

AFCRL - 68 - 0496 (I) :

CONTRACT AF 61(052)-833

7th MAY 1968

FINAL SCIENTIFIC REPORT

MULTILAYER FILTERS FOR THE REGION 0.8 TO 100 MICRONS

1st NOVEMBER 1964 - 7th MAY 1968

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PROJECTS AGENCY'S PROJECT VELA-UNIFORM.**

AD-679 564

Abstract

A study of materials for multilayer evaporated film filters for the region 1-100 μ has yielded several successful high index-low index combinations, notably PbTe/ZnS (5 - 25 μ) and Ge/CsI (10 - 80 μ).

Filters of $\frac{1}{2}\%$ bandwidth, centred to $\frac{1}{4}\%$ can be produced on a routine basis for the 15 μ region. Band pass filters of 10% bandwidth between 50 μ and 100 μ have been achieved and evaporated film and mesh techniques have overlapped in the 60-70 μ region. Good blocking filters with edges out to 70 μ have also been obtained with performance better than other methods of filtering for this region.

Foreword

The programme, initially, was conducted at Reading University and Queen Mary College (University of London) with a major sub-contract at Grubb-Parsons, Newcastle-upon-Tyne. After eighteen months Dr. Seeley's group moved from London to Reading (Department of Applied Physical Sciences).

The programme, outlined in the Introduction, demanded integrated and continuing activity from some dozen workers in the three locations ; the Investigators wish to record their satisfaction and appreciation of the contributions made by all members of the team to the total effort, as listed below.

We found this close combined operation to be stimulating and instructive; to give examples (i) a rotating source assembly was taken up by Grubb-Parsons with very rapid effects on filter production efficiency (ii) successful development of the Ge/CsI system was contributed to and implemented, very quickly, equally by industry and the Universities.

Our thanks are due to the following :-

at Grubb-Parsons:

Mr. G. M. Sisson	Managing Director
Mr. H. A. Macleod	Head of Thin Films
Mr. F. Ritchie	Research Physicist
	(working on the Contract) - Sections 4.1, 4.2, 5.4.

at Reading University:

J. J. Thomson Physical Laboratory

Mr. R. E. V. Chaddock	Research Student (supported by U. K. Ministry of Technology) - Sections 2, 3.1, 3.3, 4.5.
Dr. G. E. Peckham	Lecturer - some computer programs in Section 5.2.

Foreword (contd.)

Mr. P. J. Ellis	Electronic Engineer
Mr. G. D. Holah	Research Student (supported by the Contract, latterly).

Department of Applied Physical Sciences (formerly at Queen Mary College)

Mr. C. S. Evans	Research Student (supported by the Contract)- Sections 3.2, 4.3, 4.4.
Mr. R. Hunneman	Experimental Assistant (supported by the U.K. Science Research Council).
Dr. Heather Liddell	Lecturer at Queen Mary College (worked on the Contract for a period) - Section 5.3 and some computer programs in Section 5.2.
Dr. V. R. Costich	Visiting Scientist, for a period (under support of U.K. Science Research Council).

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LIST OF ABBREVIATIONS AND SYMBOLS

H	is unit thickness (quarterwave at λ_0) of high index
L	is unit thickness (quarterwave at λ_0) of low index
λ	is wavelength (given as μ , microns)
σ, ω	is frequency, or wavenumbers
κ	absorption index
n	refractive index
R	is reflectivity
T	is transmittance
A	is absorption given by $1 - (T+R)$
F.P.	is a Fabry-Perot filter
D.H.W.	is a Double Half-Wave filter
N	is density of charge carriers
ρ	is amplitude reflection coefficient
t	is amplitude transmission coefficient
s	is the complex frequency variable
g	is the Tchebyshev-design parameter
ϵ	is layer phase-thickness at a specified frequency
h	is the (amplitude) ripple parameter
F	is a numerical merit function

Chemical symbols are used for materials

1. INTRODUCTION

1.1 General Statement of the Problem

The programme laid down in the contract document is as follows :-

- (i) to establish the optical properties of materials suitable for use in multilayer filters in the region 0.8 to 100μ ;
- (ii) to design, fabricate and test filters using suitable materials that have been established above;
- (iii) to design and fabricate two each of multilayer filters at 75 , 85 and 95μ with half widths of 10μ or less and transmission capabilities of 50 per cent or better.

This programme resulted from consideration of the proposals made by the Investigators to the U.S.A.F. which themselves arose from a consideration of the state-of-the-art at the time and some definite practical requirements in the light of the possibilities for progress that the Investigators could recognise. The situation at the beginning of the research is summarised as follows :-

- (a) For infra-red wavelengths less than 10μ , and particularly around 4μ and up to 7μ , efficient multilayer band pass filters of down to 1% bandwidth (percentage of the wavelength or frequency of the centre of the pass band) were already established. Materials used were notably Ge/SiO, which system cannot be used longer than 7μ .
- (b) Some filters between 10 and 20μ , notably low pass filters of Ge/ZnS were known. These tended to use excessive numbers of layers and hence very narrow bandwidth filters are difficult to manufacture accurately. The problem is insufficient index contrast.

(c) Virtually no multilayer filters for the range 20 - 100 μ were available. The requirements current at that time, which were urgent and had considerable impact upon subsequent progress, can be illustrated by some examples.

(i) The need for very narrow band filters for temperature sounding radiometry from spacecraft required filters of $\frac{1}{2}$ -1% bandwidth at 10 - 16 μ .

This carries the secondary requirement that all radiation outside the bandwidth in the range 1 - 30 μ shall be rejected to such an extent that the radiation inside the pass-band dominates the energy passed at all wavelengths (say 95%).

We took this as the basic test problem in (ii) to set a standard of performance.

Established design methods (e.g. Seeley & Smith ^(1.1)) then immediately show that adequate rejection performance can only be obtained for a suitable bandpass filter with an index ratio between high and low refractive index component films of about 3.3 when around 10 films are used in the design. If the index contrast falls to say 1.6 (Ge/ZnS) the number of films rises, for similar performance, to 22 or above. The importance of obtaining a large contrast is therefore clear and is magnified for very narrow bandwidth filters since fewer layers have then to be accurately controlled in thickness. This is important since thickness control is the major difficulty in basic construction once suitable filming materials are known.

(ii) The existing filtering techniques for the range 10 - 100 μ in normal laboratory Spectroscopy were highly

unsatisfactory - this is best illustrated by the number of different techniques employed, viz. scatter plates, immersed alkali halides, grids, gratings, selective choppers, black polythene, white polythene, etc., etc. It is clear that if multilayer techniques of equal efficiency to that already obtained for $10\ \mu$ could be used from $10 - 100\ \mu$ the latter situation would be revolutionised. This would have considerable impact, for example, on grating spectrometers, interferometers and other instruments. The main filter type here is the low (frequency) pass with complete high frequency blocking. This is also a necessary component of narrow band filter systems.

- (iii) The test problem proposed by the U.S.A.F. of 10% filters in the range $75 - 95\ \mu$, with interest in measuring interstellar hydrogen emission. Again this acts as an excellent test problem particularly oriented to the problem of stacks of very thick layers.

For all these problems, given a pair of promising materials, the difficulty of controlled manufacture to optical thickness from (say) $4\ \mu$ to $50\ \mu$ is considerable, particularly when 15 layers or more are required, and the thickness control must be sometimes to 1 part in 600.

1.2 Method of Approach

Established fabrication technique is, of course, thermal evaporation; from previous work we know that the substrate temperature was likely to be the crucial parameter and also the most easily varied. We therefore gave considerable attention to this in the context of test monolayers (Sections 3.1, 3.2, 4.4) as well as in manufacture of filters (Sections 4.1, 4.2, 4.3) where special problems arise due to the necessity for simultaneous thickness control and uniformity. It will be seen that this policy proved correct and

led to very significant advances in thickness and performance.

The second parameter most accessible is choice of material, or indeed of combination of materials. This is so since the basic 'building block' of all multilayer filters is the high index film deposited between layers of low index films and as already shown the index contrast is very important.

The question of materials immediately raises the question of transparency. Absorption can be an advantage in providing natural and extensive blocking of parts of the spectrum, particularly the short wavelengths (by the semi-conductor absorption edges) and a severe disadvantage when high transparency is needed over a wide spectral range, and most substances have vibrational absorption in the 10 - 100 μ region. A series of materials therefore had to be examined and the choices could only partly be made on a systematic basis (Sections 2 and 3).

The difficulties were sufficient to encourage us to investigate alternatives to thermal evaporation, some of which could lead to radical change in the technology (Section 3.3, 4.5). This is particularly so for wavelengths greater than 50 μ where meshes can be used. (Other work on meshes has been mainly for wavelengths greater than 100 μ).

The materials for this region offer the possibility of high index contrast (shown to be greater than 3.1); this makes the present practice of design by "successive approximations" potentially inadequate for some cases (e.g. low pass filters). Exact design procedures may well become superior in these cases. This aspect is investigated in Section 5. One result is the use of 'fractional thickness' layers in a design and the first practical realisation of such a filter is reported (Section 4.3.2).

Towards the end of the programme, lack of reproducible transparency of a high-index filming material was isolated as a problem and detailed attention was turned to the deposition conditions for this material and their influence on its semiconducting properties: post-deposition heat treatment

has also been revealed as having possible importance. This work (Section 4.4) is continuing.

A number of useful computer programmes have been generated (Section 5.2) two of which - simulated 'BUILD-UP' of a monitoring record during manufacture, and 'random error studies', have given direct practical help in the manufacturing process in addition to new information.

2.3 A note on equipment

Two coating plants at Reading and one at Grubb Parsons were used; the majority of coating time was incurred at Grubb Parsons. Vacuum grating spectroscopy from 1 - 100 μ of resolution 1 cm^{-1} at Reading was extensively used in both filter and monolayer assessment.

The Grubb Parsons activity, fully integrated with the rest of the programme, is described in Sections 4.1, 4.2, 5.4. Spectroscopy and interferometry was also provided at Grubb Parsons.

References for Section 1

- 1.1 J.S. Seeley, S.D. Smith, Applied Optics 5 (1966) p. 8

2. OPTICAL PROPERTIES OF MATERIALS FOR THE RANGE 1 - 100 μ : GENERAL

2.1 Introduction

We commence with a general discussion of absorption processes and review the likely materials. Absorption in the component materials of a multilayer interference filter has two important consequences :-

- (1) First is the attenuation of incident energy, which must be minimal in the filter pass band and is helpful in the stop band.
- (2) The real part (n) of the complex refractive index is not independent of the imaginary part (κ), but is related through the Kramers-Kronig relations, for example :-

$$n^2(\omega) - 1 = \frac{-2}{\pi} P \int_0^{\infty} \frac{(2n\kappa)_{\omega'} \omega'}{\omega'^2 - \omega^2} d\omega',$$

where the absorptive term $2n\kappa$ is integrated over all frequencies ω' at which there is absorption. Therefore, the second consequence of absorption is that n is controlled by it at all other wavelengths; dispersion is illustrated schematically in Fig. 2.1. A high index contrast (~ 2.5) between adjacent layers of a multilayer is very desirable.

A study of absorption processes in solids is therefore an essential prerequisite to the selection of suitable materials. It is discussed in this Section, and one basis is a consideration of the interatomic forces which are operative, because these directly control the effects of lattice vibrations through the allowed photon-phonon and phonon-phonon interactions. Naturally, the mechanical properties are also dependent upon these forces; in particular, hardness and melting points determine whether a material is useful

evaporated layer is possible. Systematic tendencies in the mechanical properties parallel those of optical behaviour.

2.2 Electronic Absorption

The absorption due to the presence of free electrons or holes is continuous in nature; it increases in magnitude approximately as the square of the wavelength, according to the simplest of theories. Overlapping valence and conduction bands in metals mean that for these materials it is prohibitively high throughout the entire infra-red. Metal layers are not used as reflecting surfaces for interference filters in this region, as they usually exhibit a large ratio of absorptivity to reflectivity (but see Section 4.5).

When the electron energy bands become separated, as in semi-conductors, the extent to which free carriers are present depends on the width of the forbidden gap, at any given temperature and in the absence of impurities. Direct (constant K vector) and indirect (phonon assisted) transitions are possible. Electrons excited to the conduction band (usually thermally) are capable of further energy gain from infra-red radiation. They go into higher states giving rise to free carrier absorption. For these intrinsic semi-conductors, as the gap becomes larger the carrier effects become less important, until we go over to the insulators with very large forbidden band gaps. The importance of the interatomic forces in this respect, and therefore the interatomic distance, is in the manner in which they influence the gap. This is shown in Fig. 2.2 for two different structures.

As the position (E_G) of the absorption edge moves further into the infra-red, the integrated absorptive contribution to $n^2(\omega)$ shows a tendency to increase. This is illustrated (see Table 2.1) for the group IV elemental substances diamond, silicon and germanium with infra-red refractive indices of 2.4, 3.42 and 4.0 respectively. Moss^(2.1) has shown that the empirical relation :-

$$E_G n^4 = \text{a constant}, \quad (2.1)$$

is a general rule which is quite well obeyed.

The presence of impurity levels in the forbidden gap usually dominates the free carrier density and hence controls the free carrier absorption through both concentration and mobility. It is the problem of free carrier absorption which primarily dictates the requirement of high purity.

2.3 Lattice Absorption

2.3.1 Introduction

In the far infra-red, once the free carrier concentration has been sufficiently reduced, both the absorption and dispersion are primarily dominated by lattice processes. In order for the radiation to couple with phonons the following conditions must be satisfied :-

- (1) Conservation of energy.
- (2) Conservation of crystal momentum.
- (3) Associated with the phonons involved, there must be an electric moment which has a non-zero component of the right frequency in the direction of the electric vector, when summed over all unit cells. In other words, there must be a coupling mechanism.

2.3.2 Reststrahlen absorption - harmonic forces

Because the momentum of the infra-red photon is very small, Reststrahlen absorption occurs for the crystal momentum $K \sim 0$, in other words near the centre of the Brillouin Zone. This is true for the harmonic approximation of independent normal modes of vibration. The requirement of a non-cancelling dipole moment restricts the process to materials with some degree of ionicity, and hence to compounds of electropositive and electronegative element. (Electronegativity is the power of an atom to attract electrons to itself).

Positions of the Reststrahlen peaks are subject to the homology rule (2.2) :-

$$\omega_o = \sqrt{\frac{f}{\bar{m}}} \quad \text{where } \bar{m} \text{ is} \quad (2.2)$$

the reduced mass of the elements, and f is a force constant. This rule provides a useful means of comparing the strength of the effective force constants from material to material. Table 2.2 shows observed peak positions for a number of ionic materials, and also predicted positions on the basis of constant f (taken as that for NaCl). The predictions are surprisingly good, for such a very simple model, in the case of the halides. Agreement to better than 15% holds for nearly all of these; the force constants therefore vary by less than 30%. Semiconductors naturally show much poorer agreement; the force constants must typically be increased by a factor of four over that of NaCl to approach agreement with observed peak wavelengths.

The model can be very much refined by considering the distribution of electrons. Attempts have been made ^(2.3) to define effective electronic charges and compressibilities for halides, in terms of the observed Reststrahlen frequencies and the high- and low-frequency dielectric constants. The assumption is made of a uniform electric field E_e , which represents the coulombic interactions of the ions and is given by :-

$$E_e = E + \frac{4}{3} \pi P, \quad (2.3)$$

where P is the (uniform) polarisation. Approaches such as this are of limited success, because of the distortion of the ions by overlap forces; P is then no longer uniform. Distortion electric moments are consequent upon these forces. Shell model calculations have been applied with some success ^(2.4, 5) to these studies of force constants.

Non-linear effects are a consequence of taking account of the electronic structure around the masses of the atoms, which are only linked by simple harmonic 'springs' in the classical model. These effects influence

the extent of the absorption. In the harmonic model, the absorption is a 'δ' function. Non-linear effects produce damping, that is, the dissipation of energy through the other modes of vibration of the lattice, which are no longer independent. In other words, phonon-phonon interactions may take place. From the point of view of this discussion, the important consequence is the absorption of energy, which can take place over large ranges of wavelength. It is the extent and strength of these processes, which control the use to which various materials can be applied in filters. This is considered in the remainder of Section 2.3.

2.3.3 Reststrahlen absorption - damping

Broadening of the fundamental lattice peak is introduced into the classical dispersion theory by the use of a damping constant γ . For the absorptive part ϵ_2 of the complex dielectric constant, this gives :-

$$\begin{aligned}\epsilon_2(\omega) &= 2\pi\kappa \\ &= \frac{4\pi N e^* (\omega \gamma)}{V m^* (\omega_0^2 - \omega^2)^2 + (\omega \gamma)^2}\end{aligned}\quad (2.4)$$

where N/V is the number of cells per unit volume, and e^* is an effective charge.

At $\omega = \omega_0$:-

$$\epsilon_2(\omega) = \frac{(\epsilon_s - \epsilon_\infty) \omega_0}{\gamma}\quad (2.5)$$

which illustrates the 'smearing out' of the 'δ' function, and also the increment in dielectric constant. γ determines the breadth of the peak at half-height, and Table 2.3 shows this quantity for some common halides and semiconducting compounds. Some high- (ϵ_∞) and low-frequency (ϵ_s) dielectric constants are included.

The atomic origins of the damping are believed to be two.

- (1) Higher-order (anharmonic) terms in the potential energy of the lattice are involved.
- (2) Distortion of the ion occurs, because of the long range coulombic forces and short range overlap effects.

Higher-order terms in the expansion of the dipole moment M (which couples phonons to the photons) must be considered. This expansion is again carried out in terms of the normal coordinates :

$$M = m_o q_o + \sum_{ij} m_{ij} q_i q_j + \sum_{ijk} m_{ijk} q_i q_j q_k + \dots \quad (2.6)$$

It is usually stated ^(2.2,6) that the damping close to ω_o is predominantly due to the second-order potential; higher-order moments are then associated with absorption further removed from the peak. However, Szigeti ^(2.7) has demonstrated that a second-order moment automatically gives rise to a third-order potential (Φ_{ikm}) in a polar crystal, as a result of the energies associated with long range internal fields. The contribution to Φ_{ikm} is of the order $\frac{4\pi}{V} m_o m_{ij}$; large second-order moments m_{ij} should produce a contribution to one-phonon damping.

2.3.4 Higher-order phonon absorption processes

The laws of conservation of energy and momentum can be satisfied in processes which involve more than one phonon (cross-modulation from non-linear effects); the sum of the momenta must be zero. Absorption is then observed at the corresponding sum and difference frequencies for energy conservation, if there is a coupling mechanism. The latter is provided by the electric moment.

Absorption of energy can be considered quantum mechanically as dependent upon the square of the transition probability between states

$\psi(n)$ and $\psi(m)$. This probability is determined by the matrix element of the dipole moment M :-

$$M_{nm} = \int \psi(n) M \psi(m) dt$$

The multi-phonon absorption therefore can depend not only upon the high-order terms in M , but also upon those in the expansion of the potential, which appear as coefficients in $\psi(n)$ and $\psi(m)$.

Two situations are distinguished :-

- (1) Firstly, in ionic materials there is a first-order term in M , and the cross-modulation from the non-linear terms in the potential, results in sidebands to the Reststrahl^(2. 8).
- (2) In purely covalent materials such as the elements silicon and germanium, the first-order moment is zero; there is no Reststrahl, and absorption depends entirely upon the existence of higher-order terms in M ^(2. 9). These terms are from the distortion of the electron clouds.

Both effects are weaker than the Reststrahlen absorption. Clearly, in materials with any degree of ionicity both processes contribute.

In the covalent series - diamond, silicon, germanium - the strength of the two-phonon absorption progressively increases, presumably with the distortion moment. A similar tendency is observed, with greater second-order absorption, in ionic materials. Such absorption therefore appears to increase with the masses of the constituents.

Because multi-phonon absorption involves phonons over a range of energies from different branches of the dispersion curves, this absorption is extensive in wavelength. Two-phonon energy summation processes occur down to half the Reststrahlen wavelength; more precisely about half that of the longitudinal mode. Third-order processes are of negligible intensity. Difference processes occur up to a wavelength determined by the smallest

allowed energy difference, and are therefore likely to occur at very long wavelengths. They are usually weaker than summation bands, because they depend upon phonons created by the infra-red photons. The long wavelength bands also tend to be more featureless. (This is a property of the combined density of states for the difference of two dispersion curve branches).

The Reststrahl absorption in ionic materials is usually sufficiently strong to dominate the other effects discussed. The free carrier, multi-phonon and anharmonic effects are frequently of about equal importance in practice and in the "thick" evaporated films relevant to filters in this work we also find scattering effects, falling rapidly with wavelength of rather comparable strength to the secondary effects.

2.4 Selection of Materials

2.4.1 Introduction

This is carried out in two stages, which are :-

- (1) High Index materials, $n > 4.0$.
- (2) Low Index materials, $n < 2.5$.

For each class, the usable wavelength range of suitable materials can be established. We could make the criterion for absorption :-
 $\kappa < 0.0024 - 0.004$ ($3 - 5 \text{ cm}^{-1}$ at 100μ) for F.P. structures of 7% halfwidth and 30% transmission. Limits for use in the low-pass filters can also be estimated.

Mechanical properties have to be considered and those materials which cannot be fairly easily evaporated using resistive heating (melting points $> 1000^\circ\text{C}$) are eliminated. Techniques for evaporating materials of higher melting points are generally too slow for the deposit thicknesses required. These methods include electron-beam bombardment and sputtering. Materials which are poisonous (or otherwise unpleasant) have not been more than briefly considered.

2.4.2 High indices

Different semi-conductors vary considerably in refractive index (Table 2.1). Ge, Te, InSb, Pbs, PbSe, and PbTe are high index materials. Of these, the covalent elements Ge and Te were felt likely to have the most extensive useful range.

The absorption coefficient of pure Ge^(2.10) is less than 15 cm^{-1} at all wavelengths (beyond the energy gap) except between $27 - 30 \mu$ where the TO + TA combination modes reach 34 cm^{-1} at room temperature. Here, α exceeds the tolerable value for 7% half-width F.P. filters. For filters of $\frac{1}{2}\%$ bandwidth, α is excessive from 15μ to at least 35μ . Any effect upon low-pass filters is tolerable. The extent of free carrier absorption at long wavelengths was required to be investigated in film form.

Te is birefringent; this can be a disadvantage in applications involving polarised light. (The radiation reflected from gratings is polarised to some degree). The position and shape of a filter pass band may therefore depend upon the state of polarisation. Te is toxic.

InSb has a narrow band gap. This is employed to make a natural low-pass filter at 7μ . It also provides good blocking for longer wavelength filters. Free carrier absorption becomes evident beyond 20μ . The Reststrahl is at 50μ - the middle of our region of interest. The use of this material is therefore restricted.

PbTe has the highest index and lowest melting point of the Pb series. Since it is the heaviest, it also has the longest wavelength Reststrahl - at 318μ . The low frequency dielectric constant has been estimated^(2.11) to be 412 ± 40 , from which the ratio $\epsilon_s/\epsilon_\infty$ is about 13. This is very large, even compared with the halides (Table 2.3). The extent of the lattice absorption requires investigation, but the (single-phonon optically forbidden) LO mode is known to be at about 90μ . It has already been remarked that the strength of the multiphonon bands shows a tendency to increase with the atomic masses. Refractive indices exhibit a similar trend, which is

favourable for high indices. (For low indices, light atoms are indicated, but this also means short wavelength Reststrahls and is one reason for the severity of the low index problem, which will later become apparent).

There is also disagreement in the literature over the PbTe refractive index; this is partly because of free carrier absorption, which needs investigation for films. Further work is discussed in Section 4.4.

Decrease in the interatomic forces in a class of materials is reflected by the melting points. There is a parallel tendency in the electronic absorption edge positions and refractive indices. The following table shows this for a series of tellurides

	ZnTe	CdTe	PbTe	SnTe	HgTe
M. P. ($^{\circ}$ C)	1239	1098	971	780	670
E_G (ev)	2.15	1.5	0.25		0.2
n		2.6	5.6		

It appears that SnTe and HgTe have very high indices. However, the band gap of HgTe is small and the free carrier absorption may be of the same order as that of InSb ($E_G = 0.17$ ev). SnTe can be expected to have a small gap. These materials - in very pure form - would therefore be worth trying, at least for short wavelengths.

2.4.3 Low indices

The "low" index covalent elements are diamond and Se; the energy gap of Sn is too small. Se is very soft, in addition it shows considerable absorption ^(2.12). Absorption of 0.2 cm^{-1} at 10μ rises to 30 cm^{-1} at 60μ , and 100 cm^{-1} from 65μ onwards.

Low index 'weak Reststrahl' semi-conducting compounds of table 5.1 are SiO and ZnS. These materials absorb heavily from 10μ and 25μ

respectively. Two-phonon bands in $\text{ZnS}^{(2.10)}$ are about 50 cm^{-1} from 16μ . This is tolerable for 7% half-width filters; Fabry-Perots of $\frac{1}{2}\%$ would only be possible at selected wavelengths in this region.

Table 2.4 presents the melting points and some indices of ionic compounds. Many of these are low index materials, particularly the halides. Because the Reststrahlen absorption is especially strong in halides, it is essential to use those materials which have their lattice bands at very long wavelengths. This rule is easier to apply than for semiconducting compounds, because here the force constants are typically less (section 2.3.2). Table 2.2. suggests Rb, Tl, and Cs salts, especially the heavier iodides and bromides. Tl salts have very broad Reststrahlen bands $^{(2.13)}$, which were expected to somewhat reduce their usefulness. Their indices are also the highest of the alkali halides. CsBr has reached the value $n = 0.002$ at 50μ $^{(2.14)}$. CsI possess the limiting absorption at 80μ $^{(2.15)}$ for the F.P. filters with 0.07 half-width. Low-pass filters with 70% transmission would seem possible to 90μ . These wavelengths appear to be the longest at which highly ionic materials can be used for low indices layers.

Another class of low index materials is the metal oxides. Unfortunately, these have very high melting points. Classes of more complex materials, such as GaCl_3 , have suspiciously low melting points for stable and reasonably hard films.

In order to make satisfactory low index layers for wavelengths beyond 80μ , it seemed that it would be necessary to use materials beyond their Reststrahls - and thus accept the tendency of low index (light) compounds to have short wavelength fundamental lattice modes. The problem is to keep the index increment minimal. The relatively weakly ionic semi-conductors offered reasonable possibilities for this, and also for the question of two-phonon difference band absorption.

2.5 Summary

A study of absorption processes in the infra-red is given to indicate the classes of materials with the indices appropriate for multilayer filters. The number of materials which are likely to fall below the upper limits of absorption over the entire region of interest, and also possess useful indices, is limited. A series to cover all or part of the region has been discussed; the important results are summarised :-

(1) High indices

Ge (4.0) was expected to be useful for the whole region, except for very narrow filters in the two-phonon region ($15 - 35 \mu$).

PbTe (~ 5.3) is useful up to the wavelength at which Reststrahlen or free carrier absorption becomes strong. This, and also the index were required to be established.

(2) Low indices

For short wavelengths ZnS (2.3) and SiO (2.0) may be used. Two-phonon processes in ZnS are known to be too strong for filters narrower than 7% in regions between 16μ and the Reststrahl.

Alkali halides of heavy atoms offered the best possibilities for covering the whole region; Rb, Cs, and Tl salts were suggested. CsBr may be used for 7% half-width filters only to 50μ . CsI extended the region to 80μ . This appeared to be the limit, the possible exceptions being the heavier Tl salts, but these are known to have broad fundamental lattice bands.

Beyond 80μ , for pure materials of known optical properties, it appeared necessary to return to 'weak Reststrahl' semi-conductors.

Use of these, and other materials, depends upon the ability to film them in the necessary thicknesses. The fundamental and practical aspects of this problem are covered in the following Sections.

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3. PROPERTIES OF SUITABLE MATERIALS IN THE FORM OF "THICK" MONOLAYERS, 1-100 μ

3.1 Evaporated monolayers

3.1.1 Introduction

The layers of a multilayer interference filter are conventionally produced by evaporation; the thickness deposited upon the substrate is monitored optically, and is usually a quarter- or half-wave thick at the wavelength for which the filter is pitched. Design possibilities for interference filters are practically limitless, provided sufficient layers may be employed. Quantitatively this often implies that a filter with less than ten layers is poor. At wavelengths much beyond 20 μ , such structures are not readily achieved. The main factors responsible for this are :-

- (1) Inadequately good performance of the layers; in particular from absorption, scattering and non-uniformity.
- (2) The inability to achieve the correct thickness for each layer of a design.
- (3) Mechanical problems with 'thick' evaporated layers.

It is a combination of these three which restricts the usable designs.

In the present chapter, the fundamental aspects underlying these problems are first reviewed. These suggest how evaporation conditions influence both mechanical and optical properties and how they may be adjusted in film production to achieve layers of the best performance.

References to the fundamental aspects of the physical processes involved in the growth of 'thick' evaporated layers are given (3.1 to 3.8) and in (3.11) R. E. V. Chaddock, PH.D. Thesis (Reading) 1968.

An experimental study of thick evaporated monolayers follows.

3.1.2 Experimental

The films described in this sub-section were produced on one or other of three evaporation plants. These evaporators were conventional in most respects, but were equipped with the following facilities :-

- (1) Thickness monitoring is carried out continuously during deposition, by reflection of infra-red radiation from monochromators operating at short wavelengths. The number (N) of interference fringes and the wavelength (λ) to which the monochromator is set, give the optical thickness (nd) at that wavelength :-

$$nd = N \frac{\lambda}{2}.$$

Optical thicknesses at longer wavelengths are predicted when the dispersion is known; determination of dispersion data is one aim of this work.

- (2) All plants have substrate holders, which can be heated with filaments or cooled by means of the circulation of compressed air. These replace the usual rotating 'cage' which is used to aid film uniformity. Substrates are firmly clamped to the holders and temperatures measured with thermocouples.
- (3) Two of these plants (at Reading) were used for initial testing of filming materials and preliminary evaluation. The remaining plant (at Grubb Parsons Optical Works, Newcastle-upon-Tyne) was used to establish best evaporation conditions (for that plant) with selected materials. It possesses a rotating source system mounted on slip-rings, to perform the necessary spatial integration for uniformity of films. The thicker films (those on wedged substrates) and the dielectric multilayers described in this chapter were produced on that plant by Mr. F. Ritchie.

3.1.3 The Optical Properties of Thick Evaporated Films

3.1.3.1 The monolayers

These single layers are up to 60μ in optical thickness, for the following reasons :-

- (1) This is the upper limit of layers required in the spectral region less than 120μ .
- (2) The properties of thick films tend to magnify any problems encountered at smaller thicknesses and so are a useful method of studying such effects.
- (3) Consideration of such thick layers gives a useful method for predicting the behaviour of equal thicknesses of material, arranged perhaps in 30 layers, but used at short wavelengths.
- (4) Expense and production difficulties place this upper limit.
- (5) The method of computing optical constants which is employed, relies upon measurement of the interference fringe pattern. At longer wavelengths this demands thick films. A range of film thicknesses was used to increase the number of independent pieces of data.

3.1.3.2 Method of analysis

Film optical thickness (nd) can be directly obtained from measurements of the positions of extrema in the transmission interference pattern using :-

$$2nd = N\lambda, \quad (3.1)$$

where n is the index at the extremum of order N at wavelength λ , and d is the geometrical thickness. The order is identified directly if measurements extend to the first fringe at long wavelengths, or otherwise by using the closest integer to :-

$$N = \frac{\lambda_{N+1}}{\lambda_N - \lambda_{N+1}}. \quad (3.2)$$

Care must be exercised with the latter method in case of a linear variation of n with wavelength (3.9).

Reduction of data to refractive indices demands the determination of n (at one wavelength in the range of measurements) or d .

Determination of refractive index

This may be obtained from the transmitted intensities. A convenient treatment is from the analogy to transmission line theory. For one film (index n) on a substrate (index n_p) bounded by a medium of index n_o , the following results obtain for the case of no absorption. At a wavelength for which the film is $\frac{\lambda}{4}$ thick, the reflectance R at the lumped interface n_p/n_o is :-

$$R = \left(\frac{1 - Z}{1 + Z} \right)^2 \quad (3.3)$$

where $Z = \frac{n^2}{n_o n_p}$.

The reflectance R_1 at the other interface n_o/n_p is :-

$$R_1 = \left(\frac{1 - \frac{n_p}{n_o}}{1 + \frac{n_p}{n_o}} \right)^2 \quad \text{using the same argument.} \quad (3.4)$$

Transmission T of the substrate and film is given by the Airy sum for the case of no multiple beams in the substrate, that is for an average over several substrate fringes :-

$$T = \frac{(1 - R_1)(1 - R)}{1 - R R_1} \quad (3.5)$$

The accuracy of the value of n so determined is limited by the accuracy of T ; this absolute intensity is subject to scaling errors. A more accurate method is to use the fringe relative amplitude (ratio of $T_{\lambda/4}$ to adjacent extremum $T_{\lambda/2}$). A half-wave film has no influence upon the reflectivity of the system, and the transmission is that of the substrate only :-

$$T_p = \frac{(1 - R_1)^2}{(1 - R_1^2)} = \frac{1 - R_1}{1 + R_1} \quad (3.6)$$

The fringe amplitude is therefore ideally the same value as the relative transmission at $\frac{\lambda}{4}$:-

$$T/T_p = \frac{(1 + R_1)(1 - R)}{1 - RR_1} \quad (3.7)$$

This function is plotted against n in Figure 3.1 for the case of $n_p = 3.42$ employed throughout this work.

For the case of non-zero absorption, but small and slowly varying, a graphical method exists ^(3.10) for determining n using a process of successive approximations and transmission measurements of fringe intensities. This takes much of the labour out of the much more complicated computations, but involves the initial calculation and plotting of nomograms, and accurate intensity values.

Absorption of a magnitude tolerable in interference filters (for example, $A = 5\%$ in 30μ of film corresponds to $\alpha \approx 17 \text{ cm}^{-1}$) is not the major contribution to errors in our measurements of refractive index. These errors will be discussed shortly. Analysis of interference fringes within more highly absorbing regions is of limited interest for our purposes. The extent of absorption is determined from the transmission data presented later.

Determination of geometric thickness

Two methods were employed for the films in this work. These were the 'Talysurf' and the microscope interferometer.

- (1) A Talysurf produces direct traces of surface profiles. Since a tracking stylus is employed, there is danger of film distortion giving errors of $\frac{1}{2}\mu$ and more. The procedure is to track the probe across the film (which occupied half only of the substrate in our work) on to the substrate. Results are automatically plotted and any variations in uniformity are obvious. Care in associating the step at the film edge with the film thickness needs to be taken, in case of masking effects during deposition.
- (2) A more accurate technique involves the microscope interferometer. It requires the counting of fringe shifts at the film edge using monochromatic light. Identification of the fringes with white light is necessary as a preliminary. The method is really limited to films less than about 10μ thick because of fringe observation difficulties. This is just the region where the Talysurf is most inaccurate.

3.1.3.3 Results for thick films

Table 3.1 identifies the series of films and lists the metrical and optical thicknesses at the wavelengths shown. Refractive indices, at the indicated wavelengths, are also presented.

No significant ($> 5\%$) deviations from film flatness were detected during metrical measurements; it is difficult to give a quantitative upper limit of variation because of curvature of the substrates. An error in film thickness will give an equivalent error in the appropriate absolute value of n . This is not expected to exceed 5% even for the metrically thinner films of PbTe and Ge. However, data has been plotted in the more accurate form of optical thickness in the appropriate figures, because dispersion is the important quantity for multilayer production (using short-wave thickness monitoring).

Spectra below 12μ were taken with a commercial double beam instrument, and are not generally shown in the figures. Fringe positions were used to obtain optical thicknesses down to 4μ . Above 12μ the single beam infra-red instrument was used. Films were evaporated onto half only of the

substrates, and the transmission of film and substrate, relative to the substrate, is shown in the appropriate figures. Intensities were only expected to be accurate to about 5%; the causes are as follows :-

- (1) The noise level was about 2% of the signal at long wavelengths.
- (2) There were drifts in source intensity of up to 3%; an average of two runs was taken.
- (3) Stray radiation was above 1% beyond $80\ \mu$.
- (4) Wedged substrates were used to eliminate substrate interference; some deviation of the beam was experienced. The axis of the wedge was maintained vertical as far as possible in order to prevent a change of deviation or substrate absorption on positioning it for the 'film in' and 'film out' traces. This would give intensity errors.

The filming materials are now discussed in turn.

Lead Telluride (Figures 3.2, 3.3 and 3.4)

Examination of these two PbTe films under a microscope showed the existence of hair-line cracks, which divide the films into areas of about $100\ \mu$ in dimensions. This suggests that the films consist of large flat crystallites. It was not found possible to produce films of greater than $40\ \mu$ optical thickness.

There are signs of free carrier absorption in the films; κ is about 0.001 at $13\ \mu$ and 0.005 at $20\ \mu$. F.P. filters of 7% half-width are possible up to $20\ \mu$. The absorption is greater than the permissible value ($\kappa = 0.005$) for 0.6% F.P. filters even at $13\ \mu$, using the required ten layers. If absorption is small in the low index layers, the average κ will approach a tolerable level. (This in fact is known to be true for ZnS films, see also section 2.4.2 for the effect of two-phonon absorption). At wavelengths beyond $20\ \mu$ the absorption is excessive for good multilayers, and κ is 0.3 at $50\ \mu$.

The effect of cooling the $20\ \mu$ film to 80°K is shown in Figure 3.3, where measurements are presented to the long-wave limit of the filter system for our spectrometer. At the long wavelengths these results are consistent

with the 'sharpening' of a lattice peak, rather than free carrier absorption. The existence of very heavy absorption beyond 100μ is confirmed by measurements with an interferometric spectrometer. This instrument showed no transmission to 250μ , within the instrumental errors.

The strong lattice absorption (see also sub section 2.4.1) produces appreciable dispersion of refractive index, even at wavelengths far removed from the Reststrahlen peak. Figure 3.4 indicates that at 50μ , n is about 70% of its 20μ value (5.2), rising to 5.8 at 4μ (Table 3.1). PbTe is an excellent high index material and particularly good for wavelengths less than 20μ .

Germanium (Figures 3.5 and 3.6)

An obvious feature of Figure 3.5 is the high transmission at all wavelengths. The short wave spectrum shows an increase of transmission with wavelength. This is associated with scattering at short wavelengths and in particular the interference fringes were completely absent at 4μ . All the films appeared visibly rough. In very thick films, therefore Ge is not obviously a good filming material for short wavelengths, although it is adequate in thinner layers.

Two-phonon absorption in Ge is most clearly evident in MS 29, between 300 and 400 cm^{-1} . The indicated strength of the TO + TA is about 50 cm^{-1} ($\pm 50\%$). (Note that the geometric thickness is $7\frac{1}{2}\mu$). A strong peak at 270 cm^{-1} is the TO + TA in silicon. This shows that the thickness of the (wedged) substrate was different from the 'in' and 'out' traces. Using the known TO + TA strength of 10 cm^{-1} , the change in thickness is only 0.008 cm . This confirms the remark made in section 3.1.3.3 as to the wedge effect. A further feature worthy of comment is the anomolous behaviour of MS 30 near 500 cm^{-1} . Absorption due to impurities may be the cause of this. The predictions made in 2.4.1 as regards the useful transmission range for Ge in filter form, appear to hold, provided scattering is negligible (that is above 20μ). The film optical thicknesses are shown in Figure 3.6. A systematic variation in the

experimental points is ascribed to the effect of absorption upon measured fringe positions. The index falls by about $2\frac{1}{2}\%$ between 12 and 100μ . The more accurate interferometer measurements of thickness give $n = 4.1 \pm 0.1$ at 12μ . Films of up to 40μ optical thickness are possible and so Ge is very suitable for long wavelength filters.

Zinc Sulphide (Figures 3.7 and 3.8)

Figure 3.7 demonstrates the difficulty of using this material between 20μ and 100μ . The optical thicknesses of two films are shown in Figure 3.8. An apparent strong dispersion between 200 and 100 cm^{-1} is most likely a systematic error in estimating the fringe positions. The estimated index at 100μ is 2.9 ± 0.2 ; this was confirmed by an estimate from fitting a classical oscillator to the absorption peak of a thin film (not shown). In the absence of absorption, the transmission at interference maxima is 112% for this index. (This is a consequence of the blooming action; see Figure 3.1). κ is therefore estimated to be 0.03 at 80μ ; this is an order of magnitude larger than the permissible value for the 7% F.P. filters of the test problem.

At short wavelengths, absorption is negligible below the two phonon cut-off at 16μ . The refractive index changes from 2.2 at 16μ to 2.3 at 4μ .

Silicon Monoxide (Figures 3.9 and 3.10)

The long wavelength shoulder of the SiO lattice absorption is more gentle than for ZnS. Errors in the fringe positions are less, and the long wave index is estimated to be 2.6 ± 0.2 . Transmission at an interference maximum for this index is 120% (Figure 3.1). This is indeed achieved for the thinnest film, but not for the thickest film, in which absorption at 80μ is greater than for ZnS. Since absorption in SiO is more extensive than for ZnS, and also SiO is a more difficult material to evaporate, SiO is somewhat variable in performance; this is a function of evaporation rate. Oxidation to produce a proportion of SiO_2 is known to take place during evaporation; index and absorption variations are consequent.

Caesium Iodide (Figures 3.11 and 3.12)

Scattering is evident at short wavelengths. This is not as severe as for Ge, and is negligible beyond $15\ \mu$. Absorption is apparent for the fringe at $70\ \mu$ (Figure 3.11), at which point the index is 1.55 (Figure 3.12). The relative transmission should therefore be 1.27 at this wavelength and so κ is about 0.007 ($\pm 50\%$).

Conclusions from the thick film measurements

- (1) A CsI/Ge combination* is a possibility for the region $20 - 70\ \mu$. The index ratio changes from 2.4 to 2.6. Absorption is troublesome in Ge up to $25\ \mu$ only in F.P. filters narrower than 5%. CsI places a similar limit beyond about $70\ \mu$, where absorption is becoming very strong.
- (2) ZnS/PbTe is a good combination below $20\ \mu$. Again the index contrast is about 2.5. Filters with half-widths of $\frac{1}{2}\%$ are possible below $16\ \mu$, but two-phonon bands in ZnS may be troublesome between $16\ \mu$ and $20\ \mu$.
- (3) CsI/PbTe may suffer from scattering problems below $15\ \mu$. The index ratio is 3.1.
- (4) ZnS/Ge appears to be the most promising combination beyond $80\ \mu$. The absorption is still high in ZnS for filters at $80\ \mu$. The index ratio is only 1.4.

The thick films described were all evaporated at Grubb Parsons in a Balzer plant; the measurements were made in a vacuum grating infra-red spectrometer at Reading.

3.1.3.4 Low index films

Table 3.2 summarises data on some untried materials which were selected for use as low index materials. Short wave spectra are shown in Fig. 3.13. Some of these were found to have useful optical properties and filming suitability needed to be investigated. For others, especially the

* Patent applied for.

compounds with sulphur, optical properties were unknown. The source power serves as a measure of the (Mo) boat temperature. In all cases the silicon substrates were maintained at 100°C . Optical thicknesses were monitored at 3.9μ , and the refractive indices were determined from the amplitude ratio in a non-absorbing and non-scattering region.

The Cs salts are prominent in the table for both ease of evaporation and favourably low indices. Because CsI has a long wavelength Reststrahl, it was later evaporated onto a wedged substrate, and investigated more thoroughly than the preliminary results now presented in this section.

Rb and Tl halides have less favourable indices than those of Cs. The Tl salts have very long wavelength Reststrahlen bands, and the two films therefore were measured from 3μ to 100μ . Data from 25μ is shown in Figure 3.14. Both materials show the effects of absorption beyond 70μ , thus confirming the expectation from 5.4.3. Since only half an interference fringe occurs between 25μ and 100μ for these thicknesses, it is difficult to be quantitative. However, there is considerable fringe asymmetry visible.

InI and CdI were examined because of a lack of information on their optical properties. InI shows no strong absorption band between 2.5μ and 100μ . There is some background absorption increasing the wavelength in Figure 3.15. CdI has low transmission at 3.5μ ($\sim 2\%$) increasing gradually to 8μ (not shown). Absorption is small from 8μ to 30μ , but increases out to 70μ , beyond which there appears to be a strong absorption band. Both of these materials have intermediate indices and are therefore not useful for layers.

Late in this work a number of sulphides were obtained in a pure form, and a study has been commenced. The index of AsS is between 1.8 and 2.0. (The substrate is almost perfectly bloomed on the film side, and the amplitude ratio is rather insensitive to the index, see Figure 3.1). Short wave scattering is much less serious than for CsI. In_2S_3 forms hard films (also indicated by a high melting point). The index is again between 1.8 and 2.0, and there is again less scattering than for CsI, but more than for AsS. Both of these

materials are transparent in film form to at least 25μ . Measurements to 100μ have yet to be completed at the time of writing. Sulphides warr. further study.

The lack of suitable low index materials largely enforces the present wavelength limits upon the technique. It is clear that the further materials should be investigated fully.

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3.2 MONOLAYERS DEPOSITED IN THE READING PLANT

The new materials, some already referred to, were firstly tried in a second programme at Reading, which had slightly different aims.

3.2.1 Apparatus

A re-conditioned 18 inch Edwards 18E4 Coating plant has been equipped with substrate temperature control (up to 350°C) and an infra-red reflectance monitoring system. The latter consists of various interference filters and PbS or cooled InSb photoconductive detectors. Large aperture optics are used, giving advantages in energy and signal-to-noise ratio: 800 Hz modulation is employed, which avoids any difficulty with vibration. The InSb detector is used for the deposition of PbTe monitored through a 1.5% bandwidth filter at 3.9 microns; for those depositions the chamber is fitted with KRS-5 windows. The bandwidth at 3.9 microns is sufficiently narrow to monitor one hundred quarterwave layers and up to seventy layers have so far been monitored in practice. Lack of source power prevents the evaporation of Ge. The substrate block is provided with five positions which can be monitored, shuttered or chopped by external selection.

Layer thickness is adjusted to be other than an integral multiple of monitor wavelength by a mechanical chopping arrangement. The shadowing angle of chopping is adjustable between 90° and 180° or 180° and 360° . Deposition is monitored in an appropriate even order (advantageous in respect of error correction) so that any desired accuracy of thickness adjustment can be obtained by increasing the monitored thickness in step with the chopping ratio. In principle, the limiting accuracy to the technique will be determined by the number of halfwave layers which can be deposited and the accuracy of setting the chopper blade. Calibration accuracy in respect of dispersion in refractive index, fringing errors for large and small angle chopping, etc., is satisfactory and can be inferred from results given below.

3.2.2 Deposition of new materials

Samples of the following, in most cases suitably-prepared for vacuum

evaporation from crystal-growing materials, have been test deposited. All test depositions were carried out at about 100°C from an open Mo boat on to half-covered Ge or Si substrates at a rate of about 50 Å optical thickness/sec; quarterwave optical thickness (QWOT) was monitored at 3.9 microns. In Table 3.3, comparative source power is given as an indirect indication of boat temperature; efficiency in optical thickness/unit mass is also given. Short-wave spectra of these films are shown in Fig. 3.13(a) - (j).

3.2.3 Rates and efficiencies

Important parameters in the batch production of filters are the efficiencies and permissible rates of evaporation/deposition, as affected by substrate temperature. This knowledge is particularly vital in temperature cycling (as with PbTe/ZnS) and measurements have been made on single layers of PbTe and ZnS on clean germanium substrates. Evaporated mass, source heating power, source and source/substrate geometry and (as far as possible) deposition rate were held constant. Even with precautions, the rate of evaporation is not necessarily constant and observed deposition rates were excessive at commencement and beginning to fall away at completion. An efficiency parameter has been calculated in terms of deposited physical thickness per unit volume of evaporant (in Table II). The inference drawn, so far, is that these two materials are deposited in similar mass ratio and that there is little change with temperature in the range investigated.

TABLE II

Material	Substrate temperature °C	Mass evaporated gm.	Deposition rates (Mean/extreme) °A/sec	Deposited optical thickness (Quarter-waves at 3.9 microns)	Efficiency (Optical thickness in micron per gm.)
ZnS (from howitzer source)	90	11.2	48(70/39)	15	1.35*
	155	10.3	34(67/25)	14	1.34
PbTe (from a pair of boats)	235	7.9	68(120/43)	14	1.78*
	130	8.3	39(71/25)	14	1.64

* equivalent to 2.5 microns (physical thickness) per c.c. of evaporant.

3.3 ALTERNATIVE METHODS FOR PRODUCING LAYERS

3.3.1 Introduction

A number of disadvantages are associated with the production of layers using evaporation techniques. These include the following :-

- (1) A large source to substrate distance (~ 18 inches) is used to assist film uniformity. Since substrates are one inch in diameter, the consequence is a large wastage of expensive pure evaporation material. The plant is also sophisticated and expensive.

Deposition rates are limited and so considerable time is associated with the production of thick layers ($10\ \mu$ per hour).

- (2) There are materials which cannot be easily evaporated, and which would provide excellent layers were fabrication possible. This applies especially to the difficult region beyond $80\ \mu$.

Attempts to overcome both of these difficulties are described in the present Sub-section.

3.3.2 Modification of the Evaporation Technique

It is difficult to increase the deposition rate without adversely affecting film quality, even with elevated substrate temperatures. This point has been pursued in two experiments which are now discussed.

3.3.2.1 High vapour pressure method

The apparatus is illustrated schematically in Fig. 3.16. A substrate holder is maintained at a lower temperature than the walls of the enclosure; separate heater elements and air flow cooling are employed. Deposition is monitored through the fused quartz window in the top of the evacuated bell-jar. Preferential deposition upon the cooler substrate is to be expected.

A large number of films were produced by this method (~ 30) under a variety of substrate and source temperatures. No film was obtained with

acceptable transparency, although films of 50μ geometrical thickness were attainable within 15 minutes. Most of these exhibited a granular texture.

The problems are as follows :-

- (a) The deposition rate was too high and insufficiently controllable.
- (b) This is related to (a). The temperature at the surface of the substrate was far in excess of the substrate block because of the relatively poor conductivity of the silicon substrates, and the close proximity of the source (500 watts within 6 inches).

The latter problem is a difficult one to overcome without increasing the source/substrate distance. Selenium was employed in the later experiments, since it has a very low melting point and so less source radiation was involved. The Se films were not granular (Se is usually amorphous). However they showed high absorption. This approach was abandoned.

3.3.2.2 Gas flow method

A gas flow technique was attempted with the aim of separating the source and substrate, while maintaining a high rate of deposition. The apparatus is shown schematically in Fig. 3.17. The quartz-glass tube is in a furnace with a temperature gradient. An inert gas (argon) acts as the transporting medium for evaporated PbTe. Temperatures of the substrate and source are subject to independent variation. The flow rate is also adjustable.

All the films were very hard, completely opaque, with a very granular, non-uniform appearance. This remained true even with very low deposition rates.

Further experiments were not carried out. It is suggested that a modification of this approach using a chemical reaction at the substrate may be fruitful. Examples are silicon tetrachloride and hydrogen onto a heated substrate ^(3.12) and trichlorosilane and hydrogen and a heated substrate, ^(3.13) yielding silicon layers. These methods seem somewhat dangerous.

3.3.3 Bulk Layers

The possibilities for using materials in bulk form are now explored. Thickness must now be reduced to the value required, instead of the alternative approach of building up the layer.

3.3.3.1 Lapping

Substrate materials must possess high transparency even in the relatively great thicknesses compared to film dimensions. Ge (index 4), quartz (beyond 40 μ only, index 2.2) and especially Si (index 3.42) have been used in this work. These materials are also sufficiently strong to attempt to lap them down to film size (5-30 μ metrical thickness).

The problems are as follows :-

- (a) Controlling the final thickness.
- (b) The related problem of attaining uniformity.
- (c) Layers are strained in the lapping process and consequently need careful handling. This aggravates the question of measuring the thickness.

It is not necessary to achieve a high surface finish for adequate optical performance. In fact, surface scatter at short wavelengths assists the filter action (see Fig. 3.18). However, the strain produced with coarse abrasives demands that the last 30 μ be removed with the more gently polishing compounds. A polish is also required if films are to be deposited.

3.3.3.2 Plastic films

So far, no mention of organic solids has been made, basically because of a lack of transmission with these materials. The same is not so true of synthetic (man-made) organic materials of long chains, containing light atoms (hydrogen and carbon) and possessing crystalline properties.

Crystallinity causes a certain degree of symmetry within the chain. However, the configuration of a polymer usually forces the monomer units into a symmetrical arrangement, independently of three dimensional

crystallinity. The configuration of the asymmetric carbon atoms in a chain frequently produces a helical conformation in the chain, which may be considered as a one dimensional crystallinity. This may even exist in an amorphous sample, and both effects have their influence upon a spectrum. It is therefore possible to distinguish between 'crystalline' (three dimensional) and 'tactic' bands (that is, depending upon conformation).

Absorption bands in plastics in the far infra-red are mainly due to bending vibrations of the skeleton, and to stretching and bending vibrations of heavy atoms such as the halogens, silicon or metals. The transitions are low in frequency, and interactions within and between molecules may have a strong influence upon the frequency and intensity of absorption bands. This implies a sensitivity of far infra-red spectra to the physical state of a substance and conformation of its molecules. Strong (in molecular terms) but random interactions of dipole-dipole and also Van der Waals variety, within and between polar molecules, tend to produce a strong absorption background and broadening of absorption bands.

The intensity of far infra-red absorption in polymers can be classified into three groups :-

- (1) Structures of low polarity such as aliphatic polyhydrocarbons. These materials usually have only weak absorption bands in the far infra-red, especially crystalline polymers.
- (2) Compounds with more heavy and polar atoms (for example, oxygen and nitrogen). Polyesters, polyethers, resins and aromatic polyhydrocarbons fall into this class.
- (3) Polymers of molecules containing heavy polar atoms. These include halogenated polyhydrocarbons and silicones. The polar bonds give rise to relatively strong absorption bands, extending to long wavelengths.

Few measurements have been carried out beyond $40\ \mu$; the wavelengths below have been covered in a recent report ^(3, 14). A number of plastics,

taken as typical examples, have therefore been investigated in the course of this work. All have indices around 1.5 and as such could, in principle, provide an excellent solution to the difficult low index problem. The question of incorporation into a multilayer structure needs to be answered. Some remarks on this are relevant here, since they focus attention upon the desirable mechanical properties.

- (i) Evaporation of plastics appears not to be a feasible proposition. Polymerisation after deposition is an interesting possibility, however (Union Carbide:- "parylene" - this technique is not presently available).
- (ii) Few plastics are soluble for casting purposes. Change in structure is often experienced for those which do dissolve, and this is dependent upon solvent and conditions.
- (iii) Those available in film form are not sufficiently uniform for most filter purposes, and are only available in standard thicknesses.
- (iv) Attempts to evaporate thick high index layers onto plastics have been largely unsuccessful. This will be referred to again in Section 3.3.4.

Spectra of plastics

1. Aliphatic polyhydrocarbons

(a) Polyethylene

This is the most transparent plastic in the far infra-red. Extensive absorption bands in the near infra-red have been much employed for spectrometer filtering. There are two types of polyethylene, namely high pressure (low density) and low pressure varieties. The spectrum of the former, which is highly branched with both long and short side chains, is shown in Fig. 3.19. The absorption is about 1.4 cm^{-1} at 90μ ($\kappa = .0018$) rising gradually to 2.3 cm^{-1} ($\kappa = .008$) at 40μ . Polyethylene is chemically too stable for casting, but is available in films of a great variety of thicknesses. Mechanically, it is weak and easily distorted.

(b) Polypropylene

This is less transparent than polyethylene, as is to be expected from the heavier methyl group. Various forms exist ranging from amorphous to crystalline. Absorption bands are extensive to 50μ , where α is 3.7 cm^{-1} ($\kappa = .0015$) falling to 2.1 cm^{-1} ($\kappa = .0013$) at 80μ in the sample of Fig. 3.20. There are no suitable solvents for layer production.

2. Aromatic polyhydrocarbons

These are benzene derivatives and accordingly can be expected to be soluble in such a solvent.

Polystyrene

Polystyrene can be dissolved fairly readily in benzene, toluene, xylene, and similar solvents. It can be cast into films, the thickness of which can therefore be controlled. Transparency is reduced by rapid evaporation of solvent. Fig. 3.21 shows the spectrum of a sample of 'high impact' polystyrene. Transmission is steady beyond 60μ , although the film uniformity gives rise to interference fringes; α is about 5.5 cm^{-1} ($\kappa \approx .004$) at long wavelengths.

3. Polymers containing heavy polar atoms

(a) Polytetrafluoroethylene

Intramolecular forces in the $(\text{CF}_2 -)_n$ chain are much stronger than in the polyethylene chain. P.T.F.E. is one of the more transparent of category (3); this being related to the highly crystalline nature of this material. The specimen investigated (Fig. 3.22) exhibited a sharp absorption edge between 50μ and 60μ which is unusual. Absorption bands at $562, 323, 270, 204 \text{ cm}^{-1}$ due to CF_2 bending, twisting, wagging, and rocking modes are known. However, the complete absorption throughout the whole region is surprising. It suggests two possibilities, both of which may contribute to this situation :-

- (a) impurities
- (b) some form of structure effect.

Other samples obtained, have not exhibited this effect to such a degree. The lack of information on the individual environmental conditions during manufacture has prevented isolation of the effect.

The particular sample of the figure was used as an order sorting filter in the spectrometer; α is $\sim 2.8 \text{ cm}^{-1}$ ($\kappa \approx .0014$) beyond 60μ . P.T.F.E. is chemically very stable.

(b) Polyethylene terephthalate (Melinex)

This is a resinous polyester, with a monomer unit containing a large number of atoms. It has reasonable transmission only in thin films, which are obtainable in a variety of thicknesses; in Fig. 3.23 $\alpha \approx 37 \text{ cm}^{-1}$ beyond 70μ ($\kappa \approx 0.02$). It is strong even in very thin sheets (5μ), and is chemically stable.

(c) Polycarbonate (Fig. 3.24)

A very stable polyester of carbonic acid with poor transmission, $\alpha = 24 \text{ cm}^{-1}$ at 50μ ($\kappa = 0.01$). It is not available in films.

3.3.3.3 Layer structures

Layer structures, possessing weak (Van der Waals) forces between covalent planes of atoms, can be fairly easily cleaved. Mica, for instance, can be obtained in uniform sheets which would be admirable for multilayer stacks. Unfortunately, in this respect, mica is absorbing in the far infra-red ($\alpha = 250 \text{ cm}^{-1}$ at 80μ , Fig. 3.25), as appear to be other physically less favourable layer structures. However, this absorption coupled with near infra-red transmission, enables mica to be used as a selective chopper filter in the spectrometer (3.15).

3.3.4 Application of Bulk Layers to Multilayer Filters

The proposition is to replace some or all of the layers with low absorbing bulk material, to improve the performance. Particular emphasis is placed upon the low index question.

3.3.4.1 The silicon and polyethylene 'sandwich'

An early experiment was a structure made up entirely of bulk materials. Silicon plates were ground to 35μ thickness and a high order 'double-half-wave' P - Si - P - Si - P was assembled. The structure disintegrated before measurements could be completed, the short wavelength data is presented in Fig. 3.26. Some interference is visible; most of the 'filter' action is scatter from the four ground silicon surfaces. The result was sufficiently discouraging not to be pursued.

3.3.4.2 Worked spacers

It does not seem feasible to use bulk layers of smaller optical thickness than 50μ , except for the metrically thicker low index materials. Spacer layers ($\lambda/2$) of quartz have been tried. Fig. 3.27 shows the transmission of a sheet of Spectrosil B, which was not optically worked. It is fused quartz formed by the condensation of evaporated crystalline quartz fragments. Absorption is strong for such a thin sample and is increasing towards the lattice bands of quartz below 250 cm^{-1} . The index from the fringe spacing is 2.2.

A quarter wave layer of Ge was deposited either side of a second sample; the sharpened interference fringes can be seen in Fig. 3.28. The deposition had to be carried out in thin layers on alternate sides, because stress caused severe distortion. Further $\lambda/4$ layers, of both low and high index, were not attempted in order to increase the rejection, because the effect of absorption was already high.

Optically worked crystal quartz spacers for 75μ , 85μ and 95μ were produced: these have lower absorption levels than the fused variety. The grinding yielded very fragile components. They did not survive the coating attempts.

3.3.4.3 Plastic low-index layers

The disastrous effects of stress upon worked spacers would not apply to plastic layers. Availability of thin plastic films has already been discussed.

Deposition of thick high index Ge layers proved to be thermally too severe. Even mechanically strong and stable films of melinex shrank and crinkled; in this plastic κ is too high for F.P.'s in any case. The absorption in polystyrene is sufficiently low. Also, it can be readily cast, but incorporation into a multilayer structure is not readily accomplished. The technique has proved valuable, however, in protecting hygroscopic films, such as those of CsI. An application to blooming seems possible, although this has not yet been attempted.

3.3.4.4 Air-spacer Fabry-Perots

A final approach involves the elimination of only one film. The spacer layer can be eliminated by depositing $\lambda/4$ reflecting stacks upon substrates. Two such elements are then spaced with annular rings.

Experiments of this nature proved unsuccessful. The substrates were found to be insufficiently flat. This may possibly be a result of some distortion because of stresses in the films.

3.3.5 Summary

No satisfactory method for improving the evaporation technique was developed. The use of bulk dielectric layers does not appear sufficiently promising as a means of replacing evaporated layers at these wavelengths.

Plastic layer components did not find application for the difficult low index problem, because of the lack of a fabrication technique.

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4. FILTERS

4.1 Filter manufacture at Grubb Parsons - narrowband at 15 μ

4.1.1 Introduction

The work carried out at Grubb Parsons is described in this and the following Sections and is the result of an agreement dated 17th November 1964 between Reading University and Grubb Parsons for the development of multilayer interference filters for the infra-red. This agreement was, in fact, a subcontract of the U.S. Air Force contract No. AF61(052) - 833 to Reading University and required that a research programme be carried out with the following objectives;

In conjunction with Reading University to

- (i) establish the optical properties of materials suitable for use in multilayer filters in the region 0.8 to 100 microns;
- (ii) design, fabricate and test filters using suitable materials that have been established above ;
- (iii) design and fabricate two each multilayer filters at 75, 85 and 95 microns with half widths of 10 microns or less and transmission capabilities of 50 per cent or better.

The work carried out at Grubb Parsons can be broadly divided into two parts. Under (ii) above, the test problem chosen was the development of multilayer filters at 15 μ as proposed for the Selective Chopper Radiometer for Nimbus D. The requirements of narrow bandwidth and high rejection both demand a large number of layers in these filters with consequent high accuracy in thickness control and freedom from absorption. In addition, the filters are expected to withstand conditions of high humidity, thermal cycling and vibration. The films should therefore be strongly adherent to the substrate, hard

and unaffected by temperature and water vapour. The purpose of the first part of the work programme was to investigate the manufacturing problem of such filters and to establish methods of improving their performance.

The second part concerns the design and fabrication of the bandpass filters at 75, 85 and 95 microns. This was a considerable step into the unknown and, as a result, the properties of several materials had to be investigated in the search for suitable high and low refractive index films in this region of the infra-red. Although other methods of producing bandpass filters for the far infra-red are known, e.g. mesh filters, the work at Grubb Parsons has been concerned mainly with the extension of multilayer techniques to produce not only bandpass filters but also multilayer low pass filters.

4.1.2 Description of Apparatus

At the beginning of the contract, a set of vacuum equipment was provided to an identical pattern in use at the time. As the contract proceeded the equipment was modified, changed or superseded as a result of development investigations. The stimulus for development arose from the requirement for (a) good film uniformity, (b) more accurate control of film thickness, and (c) adequate substrate temperature control.

Deposition of the films took place by evaporation in a BA500 high vacuum plant which was manufactured by Balzers of Leichtenstein. The plant is shown in Figure 4.1. The vacuum system consists of a 520mm high x 510mm diameter water cooled stainless steel chamber which is pumped by a 600 l/s oil diffusion pump with baffle and backed by a two stage rotary pump. A liquid nitrogen cooled cold trap is also used. With the baffle the pumping speed is reduced to 200 l/s. Evaporation usually took place at a pressure of 10^{-5} torr as measured by an ion gauge in the pumping port of the vacuum chamber.

The plant was modified to permit monitoring in the infra-red by Grubb Parsons. A system of calcium fluoride windows and lenses was fitted to allow the radiation from a Nernst filament to be reflected from the substrate and passed into a Grubb Parsons M2 monochromator. The radiation was chopped

at 10Hz and detected by a Reeder thermocouple detector. The signal from the thermocouple was amplified by a Grubb Parsons TA10 thermocouple amplifier and fed into a Honeywell pen recorder so that a visual record of the reflectance changes during evaporation was available. A reflection monitoring system was chosen because germanium, which is a useful infra-red optical material, becomes opaque at about 125°C and, therefore, transmission measurements become impossible at elevated temperatures.

Temperature measurement was by means of sheathed chromel-alumel thermocouples. A Cambridge DE recorder was used to record the temperature. This instrument, with automatic cold junction compensation, has a range from 0 to 1200°C. Six thermocouples can be connected and read successively and continuously.

Optical transmission measurements at Grubb Parsons were carried out on a series of instruments. For the range 0.6 to 25 μ (16.667 to 400 cm^{-1}) a Grubb Parsons Spectromaster infra-red grating/prism spectrophotometer was used. This is a double beam instrument which can be operated in the single beam mode and has a limiting resolution of 1 cm^{-1} over most of the working range. Towards the end of the contract period two other Grubb Parsons instruments became available. These are the DM4 and the GM3. The DM4 is also a double beam optical null grating prism spectrophotometer for the region 500 to 200 cm^{-1} and has a limiting resolution of 2.5 cm^{-1} throughout its range. The GM3 is a single beam grating instrument which covers the range 200 to 65 cm^{-1} with a resolution better than 3 cm^{-1} . The GM3 is designed to be used with an air dryer and continuous purging facilities are built into the instrument. These three instruments together cover the range 0.6 to 154 μ (16.667 to 65 cm^{-1}). Many filters were measured in detail on the Reading vacuum grating Spectrometer in the range 10-100 μ by R.E.V. Chaddock. Measurements were also made using the Grubb Parsons IRIS interferometric spectrometer and the smaller Grubb Parsons - NPL cube interferometer. Both the interferometers are vacuum instruments and cover the range 10 to 500 cm^{-1} . An IBM 1130 computer equipped with an on-line graph plotter was used to calculate the spectra from the interferograms.

4.1.3 Narrowband filters-statement of Problem

Progress in the optical applications of thin films is closely connected with the development of improved film materials and improved deposition techniques. The difficulties have not been so much a lack of the theoretical as a lack of suitable film materials. There are only a relatively few materials, namely elements and simple compounds, which can be deposited by vacuum evaporation. In addition to the desired optical properties, other requirements such as adhesion, hardness and chemical and mechanical stability have to be fulfilled for successful applications. This restricts severely the number of available materials.

Apart from the alkali and alkaline earth halides which tend to be soft and water soluble, three materials are of interest for the $15\ \mu$ region. These are the semi-conductors germanium, lead telluride and zinc sulphide which have refractive indices of 4.0, 5.35 and 2.3 in the region of interest. The large index contrast between PbTe and ZnS make these two materials attractive as multilayer components, since it allows in theory, filters to be made which have high rejection over a broad region and narrow pass bands with relatively few layers.

However, in the case of semi-conductors, at wavelengths greater than their band gap, losses can occur due to scattering and to absorption caused by deviations from stoichiometry and impurities. In particular, one of the major practical difficulties inherent in all semi-conductor compounds like PbTe and ZnS is the fact that impurities, which may only be lattice defects or an excess of one of the constituent atoms introduced by the evaporation process result in free carrier absorption increasing sometimes with the square of the wavelength. Indeed, early attempts to use PbTe and ZnS in multilayer band-pass filters at wavelengths greater than $10\ \mu$ met with little success. Absorption usually cut the peak transmission to less than 1% despite the fact that single layers of ZnS and PbTe showed good transparency out to $20\ \mu$. This effect is obviously of extreme importance in thin film work because it implies that the techniques for obtaining good transparent thin films of single

materials may prove insufficient for the manufacture of multilayer structures, i.e. the interaction of the materials must also be taken into account. The question to consider then, is, which parameters have to be controlled to obtain films with highest transparency? In this, it was clear that substrate temperature plays an important part.

Absorption is not the only factor involved in narrow bandpass filters at $15\ \mu$. The design details of the three types of filter proposed for the Selective Chopper Radiometer (4.1, 4.2) are given as follows:

(a) Narrow band Fabry Perot

*Construction : L/Ge/LHLHLHHLHLH
Half width = 0.45%

(b) Double Half wave

Construction : L/Ge/LHLHHLHLHLHLHHLH
Half width = 1.7%

(c) Double Half wave

Construction : L/Ge/LHHLHLHLHHLH
Half width = 2.4%

ZnS and PbTe are used as the low and high index materials, respectively. The theoretical transmission of the three filters is shown in Figures 4.2(a), (b), (c).

The narrow halfwidths of these filters imply accurate control of film thickness. Not only must each filter be accurately positioned but also the D. H. W. filters are very sensitive to small errors in spacer thickness. The criterion that, for good D. H. W. filters, the spacer layers should not differ by more than the half width of the filter, implies a monitoring accuracy of $\pm 1.2\%$ for each spacer in type (b) and $\pm 1.7\%$ for the spacers in the broader type (c) filter. The effect of the random errors in layer thickness will be

* Substrates are denoted by their chemical symbols given between obliques.

examined in greater detail in Section 5.4.

The practical difficulties of evaporating films increase rapidly above 10μ . Such thick layers pose problems of mechanical stability since the stresses observed in evaporated films are frequently of considerable magnitude. Two of the most important causes of this mechanical stress are:

- (1) Since the substrate temperature during deposition usually differs from ambient, stresses are introduced by the different expansion coefficients of film materials and substrate.
- (2) In many cases, the greatest amount of stress is caused by the freezing in of lattice defects during the condensation process. To some extent, the stress resulting from the lattice disorder can be released by post annealing or often more effectively, by the choice of a higher deposition temperature. Since on the other hand higher deposition temperatures increase the stress resulting from different expansion coefficients of films and substrate, the optimum deposition temperature must be found by experiment.

The ideal film, therefore, must not only be transparent but also hard and strongly adherent to the substrate. It should survive environmental changes such as temperature cycling and humidity variations. In this respect the anti-reflection coating of ZnS is particularly vulnerable since it is exposed to the atmosphere. Therefore, before focussing attention on the multilayer, it was decided to carry out an investigation into the optimum conditions for films of ZnS. The ZnS was obtained as a powder from Levy West Limited.

4.1.4 Zinc Sulphide films

Films of ZnS were evaporated initially from a tantalum boat at the centre of the vacuum chamber. Later, a howitzer source was used which contained sufficient material to produce a film of at least 22μ optical thickness. Thicker layers were produced by using two such sources. A shutter was used to avoid contamination of the substrate while the evaporant is brought up to evaporating temperature. The substrate was a germanium disc 0.92 inches in

diameter and .02 inches thick.

The substrate was positioned 15.5 inches above the sources in the centre of the vacuum bell and attached to a copper block 3 inches in diameter and 2 inches thick. The block was equipped with a heater and was cooled by compressed air circulating around the block in a coiled copper tube. Thus, the substrate temperature could be held constant during the evaporation process by balancing the heating and cooling rates of the copper substrate holder. Two sheathed chromel-alumel thermocouples were embedded in the block to within .015 inches of the substrate and the Cambridge DE recorder was used to monitor the substrate temperature. The heater leads, cooling tube and thermocouples were taken out through the top plate of the vacuum chamber. The maximum attainable temperature was approximately 350°C.

Films of ZnS were deposited on germanium substrates under the following conditions:

- (a) With and without high voltage glow discharge cleaning of the substrate surface.
- (b) At substrate temperatures up to 240°C
- (c) At optical thicknesses from 3.4 microns to 43 microns.

In addition, each film was subjected to an adhesion test, using Lasso adhesive tape which has a tensile rating of 600 gms. per inch, and thermal cycling between - 196°C (liquid nitrogen temperature) and 250°C.

In general it was found extremely difficult to deposit permanent films at substrate temperatures below 80°C unless a high voltage glow discharge was used to clean the substrate. Without the ionic bombardment the films lifted very quickly after exposure to the atmosphere. At temperatures of 100°C and above, permanent films could be deposited but adhesion and resistance to thermal cycling were poor.

The glow discharge results in a marked improvement in film adhesion and durability. Initially the discharge was set up in air admitted via a needle

valve to the system with both diffusion and rotary pumps running and liquid nitrogen in the cold trap. After 10 minutes, the pressure was reduced as quickly as possible to 10^{-5} torr and the deposition of ZnS commenced within three minutes of the discharge.

The ionic bombardment enabled thick films to be deposited at any substrate temperature up to 240°C (approximately the maximum temperature which was attempted). For substrate temperatures greater than 80°C , films are very robust and show very good adhesion. They withstand immersion in liquid nitrogen and thermal cycling between liquid nitrogen temperature and 250°C . It was also found possible to deposit permanent films of ZnS at substrate temperatures less than 80°C . However, thin films (2 or 3 microns optical thickness) deposited at 50°C were rather soft and easily scratched although adhesion was good. Thicker films at 50°C were found to be more robust, showing good adhesion and a harder surface.

ZnS films produced in this manner, deteriorated rapidly when exposed in a humidity chamber to a relative humidity of 98% at 50°C . The coatings crazed and adhesion to the substrate was considerably reduced. Experiments showed that more resistant films could be produced if the evaporation was carried out in oxygen at a pressure of 10^{-4} torr. Complete resistance in highly humid conditions, however could not be achieved although the coatings which were evaporated in O_2 at 10^{-4} torr survived a relative humidity of 98% at 50°C for approximately 12 hours.

The following table lists films of ZnS with respect to their deposition temperature and optical thickness which were sent to Reading for further study (Reported in Section 3). The deposition rates are also included:

Temp. °C	Optical thickness μ	35	13.2	20	24	43
50				M(57) 43A/sec		
80					M(56) 37A/sec	
100			M(53) 64A/sec	M(54) 30A/sec		M(55) 61A/sec
120		M(49) 32A/sec	M(52) 32A/sec			
150		M(46) 64A/sec	M(51) 27A/sec			
180		M(47) 26A/sec	M(50) 23A/sec			
240		M(42) 35A/sec				

The effect of increasing the substrate deposition temperature is to increase the size of the crystallites within the film until eventually the film appears to have a whitish "bloom". For temperatures greater than 50°C this clouding increases with temperature for any given film thickness. The clouding was also found to increase with thickness for a given deposition temperature although not so rapidly at lower temperatures (less than 100°C). The clouding is accompanied by a decrease in optical performance of the films which is particularly evident at shorter wavelengths (less than 5μ), for temperatures of 180°C, but not so noticeable at longer wavelengths. The increase in crystallite size within the film can be prevented by holding the substrate at a sufficiently low temperature, e. g. 50°C. The films, however, are not very robust.

As a result of this investigation into the optimum conditions for evaporating thick films of ZnS, the following conclusions have been reached.

- (a) Very thick layers of ZnS can be deposited on germanium. Under the correct conditions, the thickness of the film appears to be limited only by the capacity of the evaporating source.
- (b) A high voltage glow discharge improves the quality of the film and is necessary for the successful deposition of films at substrate temperatures below 100°C .
- (c) The films should be evaporated in O_2 at 10^{-4} torr to obtain maximum resistance to high humidity atmospheres.
- (d) The ZnS should be deposited at as low a temperature as is possible in order to avoid crystallite growth with increasing film thickness. For a hard film a suitable temperature is 100°C .

4.1.5 PbTe/ZnS Multilayer filters at 15μ

It has already been stated that the early attempts at making bandpass filters at 15μ failed because of heavy absorption at wavelengths greater than 10μ although single layers of PbTe and ZnS on Ge showed good transparency in this region. In these early experiments no attempt was made to control such parameters as substrate temperature. It was therefore decided to investigate the effect of substrate temperature on multilayer PbTe/ZnS systems with particular reference to the narrow band pass filters proposed for the Selective Chopper Radiometer.

Experimental

The experimental set up was similar to that used for the investigation into the optimum conditions for ZnS films. The two materials were evaporated from resistance heated sources. As before, a howitzer source was used for the evaporation of ZnS and a tantalum boat used for PbTe. Both sources

were placed as near to the centre of the vacuum bell as possible and not more than 3 ins. apart. A shield between the two sources served to reduce cross contamination of the source material during evaporation. A small shutter was also used both to reduce contamination and improve thickness control of the layers. The arrangement of the sources is identical to that shown in Figure 4.3.

As before, the germanium substrate was attached to the heater block 15.5 inches above the sources. Hence both sources and substrate were fixed. The maximum substrate temperature which could be obtained with the heater was approximately 350°C . The evaporation of the materials was carried out at a pressure of 10^{-5} torr.

Monitoring

Film thickness control was achieved by monitoring the reflectance changes of the germanium substrate as the high and low index layers of the filter were deposited. Separate test glasses were not used because it was felt that greater accuracy could be obtained by monitoring the Ge substrate. However, continuous monitoring of the actual filter as it is constructed does present some problems. As a succession of $\lambda/4$ films of alternating high and low index is deposited, the reflectance of the substrate rapidly increases and the change in signal decreases (e.g. as in Fig. 4.4). Therefore, after only a few layers, there is considerable uncertainty as to the exact location of the turning points. To avoid such errors, it was decided to monitor in the fourth order i.e. between 3 and 4 μ for a first order pass band in the region 12-16 μ . The advantages of this method are:

- (1) the reflectance changes are, in theory, constant for each material (in practice, there is always a gradual change in substrate reflectance due to monitoring errors) and
- (2) the errors in layer thickness are reduced by a factor of 4.

Absorption in the PbTe between 3 and 4 μ is not a problem since the shift in the PbTe absorption edge with temperature is approximately $+5 \times 10^{-4}$ eV/ $^{\circ}$ C. Therefore, only a moderate substrate temperature rise of 100 $^{\circ}$ C, is required to move the absorption edge to 3 μ and so bring the monitoring wavelength into the region of high transparency of the PbTe.

Fabry Perot filters

A study of the variations in performance of the narrow bandpass Fabry Perot filter type (a).

$$L/Ge/LHLHL\text{HHLHLH}$$

at $\lambda_0 = 15 \mu$ has shown the degree of substrate temperature control which is required for PbTe/ZnS systems. The Fabry Perot filter is especially sensitive to absorption losses since the peak transmission is given by:

$$T_{\max} = \frac{1}{(1 + \frac{A}{T})^2} \quad 4.1.$$

where A and T are the absorption and transmission of each HLHL reflecting stack in the filter. Thus, by measuring the peak transmission, the ratio A/T which is a measure of the absorption in the filter, can be calculated. By varying the substrate temperature during deposition of a layer of PbTe on ZnS and vice versa over a series of filters, it was established that, for certain temperature, the absorption is a minimum. The table below shows the variation of A/T for the different deposition temperatures.

Filter	Deposition temperature		A/T
	PbTe	ZnS	
D16	175	100	•
D18	250	100	.43
D20	260	100	.46
D21	220	100	.49
D23	200	100	.72
D25	330	100	.69
D24	250	150	.77
D26	250	65	.67

The absorption in filter D16 was so high that it effectively obliterated the pass band. The data in the table has been plotted in Figures 4.5.(a) and (b) as two curves. Fig.4.5(a) shows the effect of varying the PbTe deposition temperature with the ZnS temperature constant at 100°C. It can be seen that a pronounced minimum in absorption occurs for a PbTe deposition temperature of 250°C. On either side of this temperature the absorption increases but more quickly for lower temperatures. Thus, the absorption at 175°C is so great no pass band appeared whereas at 330°C the peak transmission of the filter is 35%.

Similarly, Figure 4.5(b) shows how the absorption in the Fabry Perot filters is influenced by the ZnS deposition temperature. As in Fig.4.5(a) a minimum occurs for a ZnS temperature of 100°C. Also, since the slope of the curve in Fig.4.5(b) is steeper on both sides of the minimum than the slope of the curve in (a), it would appear that the ZnS deposition temperature is more critical than the PbTe temperature. Fig.4.6 shows the transmission of filter D18 which was made under the optimum conditions, viz. a PbTe temperature of 250°C and a ZnS deposition temperature of 100°C.

4.1.6 Double Half Wave Filters

Two types of D. H. W. filter were attempted. The designs are as follows:

Filter (b) L/Ge/LHLHHLLHLHLHHLLH

Filter (c) L/Ge/LHHLLHLHLHHLLH

In the construction of the D. H. W. filters (b) and (c) the main problem has been control of the film thickness. The design half width type (b) is 1.7% of the peak wavelength and the half width of type (c) is 2.4%. The criterion that, for good results, the spacer layers should not differ by more than the half width of the filter implies a monitoring accuracy of $\pm 1.2\%$ for each spacer in type (b) and $\pm 1.7\%$ for the spacer in the wider band filter. D. H. W. filters of types (b) and (c) have been constructed, using the same methods as for the Fabry Perot filters, the narrower type (b) having 60% peak transmission and the wider type (c) having 67% transmission. These results are shown in Figures 4.7(a) and (b) respectively. The success obtained with the latter supports the observation that the errors arising from the method of monitoring in the 4th order are not greater than $\pm 1.8\%$. However, there is some evidence that the temperature cycling of the substrate which is necessary, for least absorption produces significant systematic errors in the layers. The magnitude of this temperature effect can be considerable since a PbTe layer increases in optical thickness by 1.5% on cooling through 150°C whereas a ZnS layer increases by 0.7% on heating through the same range. The net result due to temperature cycling is that mismatching of the spacer layers takes place although the full build up of the temperature effect is inhibited to some extent by the random errors in monitoring. (The effect of random errors on these filters is examined further in Section 5.4). It was clear, however, that temperature cycling between 100°C and 250°C during filter manufacture could lead to serious mismatching of the layers. Experiments on the type (c) filter which, having a larger halfwidth, was the easier to make, showed that the transmission of these D. H. W. filters is increased on reducing the temperature difference by half, i. e. on depositing the ZnS at 150°C and the PbTe at 220°C. Indeed the results show that it is even possible to manufacture this

type of filter by depositing the two materials at the same substrate temperature. Filters have been made at substrate temperatures as low as 120°C although the best results have been obtained by depositing the ZnS and PbTe layers at 160°C. Maximum transmission of 80% has been achieved in this way as is shown by D. H. W. 14 (Figure 4.8).

At first, it would appear that these results contradict the measurements on the Fabry Perot filters which implied that an increase in absorption takes place for ZnS deposition temperature different from 100°C and PbTe temperatures different from 260°C. However, it is known that the effect of absorption in a D. H. W. filter is a minimum in the pass region and a maximum in the stop region (4.3) whereas in the Fabry Perot the absorption has a high and essentially constant effect. Thus, any improvement in the performance of the D. H. W. filters must be attributed not to less absorption but to more accurate monitoring. The increase in accuracy must result from the absence of any differential changes in the optical thickness of the layers which occur during temperature cycling of the substrate.

Despite the improvement in the performance of the type (c) D. H. W. filters which resulted from the removal of differential thermal treatment the monitoring technique was not sufficiently accurate to guarantee 100% success with the type (b) design. Indeed, on average, only one out of three attempts at making the latter was successful. However, the success rate was increased to better than one in two by scanning for the third order peak of the D. H. W. during manufacture. The design of the filter is Ge/LHLHHLLHLHLHHLLH and the third order peak occurs after deposition of the layers Ge/LHLHHLLH. This peak reappears towards the end of the second HH spacer and is used to complete that layer. Now calculations show that the height of the first order follows the changes in the third order peak so that the required optical thickness of the second spacer can be achieved by halting depositions when the third order reaches a maximum. Deposition of the remaining layers is monitored normally in the fourth order. Use of this technique, and deposition at a

uniform temperature of 160°C , have improved the yield of these filters to better than one in two.

Filter Production

The experiments described above show the importance of substrate temperature in the manufacture of such PbTe/ZnS multilayers. However the accurate control of substrate temperature prevented rotation of the substrate and as a consequence the layers are only uniform to 0.5%. The uncertainty in pitching the main pass band is also relatively large being approximately $\pm 1\%$ of the central frequency. Thus filters made according to the methods previously described were not acceptable for the Selective Chopper Radiometer for which filters are required to be uniform to 0.1%. The narrow band Fabry Perot filters for the S.C.R. are also required to be pitched to within $+0.5 - 1.5\text{cm}^{-1}$ of the target, the CO_2 Q branch at 667cm^{-1} . Such small tolerances on pitching and uniformity clearly could not be attained with the experimental methods described previously. The normal method of improving uniformity of deposition is to introduce relative rotation between sources and substrates. Usually the substrate is rotated; however, in this case, it was found more convenient to rotate the sources. In this way the fixed substrate can be temperature cycled with rapid heating and cooling periods in order to keep the manufacturing time to a minimum. Source rotation also allows a batch of several filters to be made at the same time since adequate uniformity is achieved over a considerable area. This, in turn, allows selection to be carried out and offers a partial solution to the problem of accurately pitching the narrow band pass filters.

The rotating source assembly consists of three copper slip rings each of internal diameter 12.0 inches and of rectangular cross section $\frac{1}{2}$ ins x $\frac{3}{4}$ ins. The two evaporation sources are mounted directly on the rings and electrical contact to each of the rings is by two high vacuum copper/graphite brushes with a total maximum rating of 150 amps. The rings are mounted vertically and are carried by a mild steel base ring which is, in turn,

supported by six bearings and rotated at about 30 r.p.m. by a rack and pinion drive. In this system the sources rotate 11.0 inches below the substrates in a circle of 12.0 inches diameter.

The multiple substrate holder consisted of a copper plate 4 inches in diameter and 3/16 inches thick. Two holes were drilled in the rim of the plate to take two thermocouples. With the arrangement of grouping six substrates in a circle around the monitor substrate a batch of seven can be made simultaneously. Each substrate is mounted in a hole in the plate and a copper plug is screwed down on the substrate. Thus, each substrate is firmly embedded in the substrate holder which is attached to the copper block by three screws. A shutter was also made to ensure accurate monitoring.

This system of rotating sources has proved to be a successful method of mass producing the PbTe/ZnS filters. Table 4.1 gives a list of the first successful batch of 5 D. H. W. type (b) filters to be made in this way. The mean peak wavelength of the four filters D. H. W. 40 A, B, C and D is 14.26μ . Hence all four are within $\pm 0.9\%$ of the mean. It is interesting to note that the maximum tolerable error in spacer thickness is $\pm 1.4\%$ for good filters.

The narrow band Fabry Perot filters have also been made successfully using the system of rotating sources. Table 4.2 lists a batch of Fabry Perot filters which were produced in this way.

As is to be expected, source rotation has improved layer uniformity. Figure 4.9 shows the theoretical distribution of evaporant from a small rotating surface source on to a parallel plane receiver. The source is assumed to rotate in a circle of radius 6.0 ins. and 11.0 ins. below the receiving plane. This corresponds to the actual source/substrate geometry in the evaporation plant. The centres of the six 0.92 ins. diameter substrates lie on a circle of radius 1.25 ins.

A comparison between the theoretical curve in Figure 4.9 and the spread in Tables 4.1 and 4.2 shows that some discrepancies exist between theory and experiment especially in the case of Table 4.2. The mean wavelength of the

batch in Table 4.1 is 14.26μ which is 1% smaller than the monitor filter, in agreement with Figure 4.9. However, the batch of six filters in Table 4.2 is at a slightly (0.2%) longer wavelength than the monitor. The scatter in the position of the peak wavelength in Table 4.2 is also much less than that in Table 4.1. The difference is attributed to slight variations in the radius of rotation of the sources in the two cases.

The Fabry Perot filters, as produced by these methods, are uniform to $<1\text{cm}^{-1}$ at 667cm^{-1} (15μ) or very nearly 0.1%. Measurements of the change in peak wavelength with position on the surface of the filter are given in Figure 4.10.

It is clear, therefore, that rotation of the sources together with scanning of the third order peak is a suitable technique for making such PbTe/ZnS multilayer bandpass filters in the region of 15μ . The main advantages are:

- (1) Adequate temperature control of the substrates is possible.
- (2) Uniformity is improved
- (3) Third order peak scanning leads to increased monitoring accuracy
- (4) Several filters can be made at once and therefore a selection is possible.

REFERENCES FOR SECTION 4.1

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4.2 FILTER MANUFACTURE AT GRUBB PARSONS - LONGWAVE FILTERS 20 - 100 μ .

4.2.1 Introduction

In this section we are concerned with the fabrication of multilayer filters (both narrow band and low pass) for the far infra-red region 20-100 μ . In particular, it will be shown how narrow band filters have been developed for 75, 85 and 95 μ under the terms of the contract and also how low pass filters have been constructed using high vacuum evaporation and multilayer techniques, to suppress the short wave sidebands of the narrow band filters.

The following Sections describe the research and development carried out on materials for the far infra-red and the techniques used (not all successfully) to produce narrow band pass and low pass filters for this important region.

4.2.2 Materials for the region 20-100 μ .

4.2.2.1 Substrates

Three materials in particular are of interest in this region of the infra-red, viz. silicon, germanium and crystal quartz. The optical properties of the semiconductors silicon and germanium have been examined recently in the far infra-red.

Both Si and Ge are co-valent and because of their chemical similarities, it may be expected that their optical properties would also correspond. This, however, is not the case. Germanium exhibits more dispersion and more absorption which, because of its reproducibility, suggests that it is due to lattice absorption which occurs at lower frequencies in germanium. For wavelengths greater than 20 μ (500cm^{-1}) silicon is superior in transparency and is therefore a better substrate material than germanium.

Crystal quartz is used extensively in the far infra-red. Its Reststrahlen band occurs between 5 and 40 μ and so it cannot be used as a substrate in that region. The optical properties of crystal quartz in the far infra-red have been measured by Roberts and Coon (4.4) and also by Russell and Bell (4.5).

Crystal quartz is birefringent and therefore substrates of quartz should be Z-cut crystals (optic axis perpendicular to the sample surface) to eliminate the birefringence. However, Z-cut crystals show absorption due to a weak lattice band at 128cm^{-1} ($78\text{ }\mu$) which lies in the region of interest. For this reason, silicon was preferred to crystal quartz as a substrate material for the bandpass filters at $75\text{ }\mu$. In addition to the consideration of transparency, the choice of silicon was influenced also by the knowledge that better, more adherent films are obtained with Si than with quartz substrates.

The index of Si is approximately 3.4 in this region of the infra-red ($75\text{-}100\text{ }\mu$). This apparently "high" value presents no problem since 'matching' to a multilayer system can usually be easily arranged.

4.2.2.2 Evaporated layers

The films in Table 4.3 have been deposited on both plane and wedged Si substrates for measurement in the far infra-red. The films were deposited on half only of the wedged substrates (wedge angle 3°) to eliminate the effects of substrate absorption and substrate interference fringes. These films were analysed at Reading University to obtain the refractive index and absorption coefficient of the materials (as reported in Section 3).

The materials were chosen because of their known usefulness in the near infra-red. The extent of absorption bands expected at long wavelengths was not known and is among the completely new data to be obtained.

Lead Telluride (Fig. 3.2)

The films of PbTe were evaporated from a resistance heated tantalum boat which was mounted on the slip ring assembly and rotated as described in Section 4.1. The substrate temperature was held constant at 250°C during deposition of the lead telluride. It was not found possible to produce films of optical thickness greater than approximately $40\text{ }\mu$ which remained on the substrate.

The index is $5.6 \pm 5\%$ at $20\text{ }\mu$. It appears to fall to 70% of this value at $55\text{ }\mu$ (note change of fringe amplitude in Fig. 3.2, presumably due to the existence of strong absorption at between 100 and $300\text{ }\mu$). The presence of this

absorption is of great interest and remains to be understood fully.) The absorption changes from 6% at $15\ \mu$ to 28% at $33\ \mu$ for the case of the $40\ \mu$ film.

Germanium (Fig. 3.5)

Several attempts were made to evaporate Ge from tantalum, molybdenum and tungsten boats. The molten Ge attacked all three metals and the films were absorbing. Absorbing films were also produced from a re-crystallised alumina crucible. Finally, a thin-walled carbon crucible was successfully used to evaporate the Ge. The carbon crucible was heated by a .004 in. thick strip of Ta and surrounded by a simple heat shield. However, the current requirements of the source were too large (~ 200 amp) for the slip ring assembly and therefore rotation of the source was not attempted. Instead the Ge source was positioned in the centre of the vacuum bell and directly below the substrates.

Generally, it was found that a high substrate temperature of 200°C was necessary for the successful deposition of thick Ge films. Presumably the high deposition temperature is effective in releasing the stress resulting from the lattice defects during the condensation process. Films deposited at lower temperatures tended to lift from the Si substrates.

There is no significant deviation of the index from the bulk value (4.0) in any film. Background absorption, i.e. non lattice in origin, is about 5% for both thinner films ; there is no evidence at these wavelengths to differentiate between scattering and free carrier effects, and both may be present. There is an anomaly in the case of the $40\ \mu$ film (MS30) at $20\ \mu$. The origin of this is unknown but may be the result of an impurity. The absorption in the $40\ \mu$ film changes from 8% at $80\ \mu$ to 15% at $13\ \mu$, suggesting the existence of scattering. Indeed, all films show very poor fringes between 2.5 and $5\ \mu$, providing evidence for some scattering. This is related to the high substrate temperatures required to produce such thick films. The high temperature results in an increase in crystallite size with the result that, for the thicker films, the surfaces appear visibly rough.

As for PbTe, films much greater than $40\ \mu$ in optical thickness could not be deposited. A $60\ \mu$ film was attempted but it had a fine crazed appearance and optically was very poor.

Zinc Sulphide (Fig. 3.7)

A Howitzer source was used to evaporate the ZnS films. The source was rotated on the slip ring assembly during the evaporation process. All the films were deposited at 100°C . It was found comparatively easy to produce films at least up to $60\ \mu$ optical thickness.

The films show strong reststrahlen absorption in the region $500\text{--}130\text{cm}^{-1}$ which produced a considerable increment in the refractive index. This is about 2.8 at $100\ \mu$ (as opposed to 2.3 at $10\ \mu$).

Silicon monoxide (Fig. 3.9)

The SiO films were evaporated in the same manner as the ZnS films i.e. from a Howitzer source rotating on the slip-rings. Again, the SiO films were deposited at 100°C and no difficulty was experienced in producing the thick films.

The same comments apply as for ZnS, the strong reststrahlen absorption producing a considerable increment in the refractive index which is about 2.6 in the far infra-red.

Both the SiO and ZnS films exhibit very good fringes between 2.5 and $5\ \mu$, which is indicative of the good uniformity and optical quality of these thick films at short wavelengths.

Caesium Iodide (Fig. 3.11)

The CsI films were evaporated from a rotating Ta boat. The substrate temperature during deposition of the films was 100°C .

Figure 4.11 shows the transmission of a $19\ \mu$ optically thick film of CsI on a plane silicon substrate. The index of the film is approximately 1.7 and the film anti-reflects the silicon. CsI is ionic and has a strong reststrahlen band at $\sim 145\ \mu$, which sets the long wave limit of transparency.

Comments

Germanium appears to be a suitable high index material for the infra-red region. Although the refractive index of the Ge is less than that of PbTe the Ge films show higher transparency over the infra-red and particularly between $70\ \mu$ and $100\ \mu$.

Of the three low index materials none has been found suitable for the whole of the region $70\text{--}100\ \mu$. ZnS and SiO films are still absorbing too heavily here to be very useful in narrow bandpass filters. They could possibly be used beyond $100\ \mu$ but the large increment in their indices which is produced by their restrahlen bands effectively lowers the index contrast between them and germanium and therefore reduces their 'efficiency' in multilayer interference filters. Caesium iodide is a good low index material below $80\ \mu$ although it has the disadvantages of being soft and hygroscopic. The CsI Restrahl absorption at $\sim 145\ \mu$ effects its transparency at higher frequencies and, although the absorption coefficient has fallen to 1cm^{-1} at $50\ \mu$, the absorption at $85\ \mu$ is too large for good bandpass multilayer filters to be possible at 85 and $95\ \mu$ using CsI and Ge as the low and high index materials, respectively. Nevertheless, CsI is an attractive low index material between 20 and $80\ \mu$.

There are certain systematic trends which make the finding of a low index material for the region $70\text{--}100\ \mu$ difficult and the complete solution of the problem is still being pursued. Most of the possible materials possess, like CsI, some degree of ionicity which results in strong restrahlen bands. The position of these bands is subject to the homology rule and most materials of low index have their band within or near the region of interest. These conclusions are essentially consistent with those in the parallel discussion in Sections 2 and 3.1.

4.2.3 Filter Fabrication

The solution of the problem of manufacturing bandpass filters at 75 , 85 and $95\ \mu$ has been pursued in two ways involving high vacuum evaporation :

- (i) Composite filters with evaporated layers on low index self-supporting spacers.
- (ii) Evaporated multilayers consisting of several evaporated layers of alternate high and low index material on a suitable substrate.

Experiments on methods (i) and (ii) which involve the extension of conventional high vacuum and multilayer techniques to the far infra-red, were carried out at Grubb Parsons.

4.2.3.1 Composite filters

The lack of a suitable low index material for the whole of the region 70-100 μ made attractive the possibility of incorporating prefabricated low index layers into a filter. Two materials were of particular interest, viz, melinex (polyethylene terephthalate) and crystal quartz. Also examined from this point of view was Spectrosil B, a variety of fused silica.

Attempts were made to incorporate these materials as the spacer layers in simple Fabry Perot filters with germanium as the high index layer. The design construction of the simplest Fabry Perot is H LL H in which two quarter wavelength thick high index layers are separated by a halfwave low index spacer. The ultimate aim was not only the construction of a simple bandpass filter but also the fabrication of more sophisticated designs from several of these units.

Melinex filters

Melinex is transparent in the region 80-100 μ (Fig. 3.23) and has a refractive index of 1.8. It is therefore a possible low index material to be used with germanium ($n_H = 4.0$). The main advantage to be gained from using Melinex is that sheets of various thicknesses and good uniformity can be easily obtained. Properly mounted, such sheets of Melinex are commonly used as beam splitters in far infra-red instruments like interferometers.

However, despite these attractive properties of the material, experiments on the possibility of using Melinex as a Fabry Perot spacer have not been successful due to difficulties with the thermosetting nature of the material. At the present time the best method of preparing Melinex film has been to bake it for a few hours at 150°C to allow for some shrinkage in advance. The film was then stretched over an annular frame and placed over a thick, slightly convex copper plate. The copper plate served to reduce the temperature of the plastic during evaporation but, even so, the coating obtained is not satisfactory since the Melinex tends to deform during lengthy or hot evaporation. Indeed it was observed, at first, that the infra-red radiation in the monitoring beam was sufficient to burn a hole in the Melinex. Later attempts employed a separate test glass for monitoring the thickness of the evaporated Ge layers.

Unfortunately, it was not found possible to deposit thick Ge films on the Melinex. Wrinkling and crazing of the film occurred after only three or four quarterwaves at 3μ of Ge had been evaporated. In this respect, the high temperature requirements for thick Ge films seem to oppose the cool conditions needed to keep the plastic stable.

Although relatively thin layers of Ge could be deposited on Melinex, none of the attempts at depositing the much thicker Ge films which are required for interference filters in the far infra-red were successful. It was therefore decided to abandon such attempts and concentrate on the other methods.

Crystal quartz filters

Quartz plates cut perpendicular to the optic axis were optically worked to form second order spacers at 75, 85 and 95 microns, respectively. The values of the refractive index of quartz in this region of the infra-red have been measured by Roberts and Coon and also by Russell and Bell. With these values the thicknesses of the plates are .0014, .0016 and .0018 in. respectively.

Initial trials with thin (.003 in) plates of Spectrosil were carried out to determine suitable methods of cleaning, holding and coating such very fragile, quartz samples. The transmission of a spectrosil plate is shown in Figure 4.12. Such plates are quite transparent at wavelengths greater than 120μ and show about 12% absorption at 95μ . The refractive index is 2.2 in this region of the infra-red.

The spectrosil was mounted, at first, in the copper block in the same manner as the germanium plates which were used as substrates for the 15μ filters. A slender suction tube attached to a small air pump was used to handle the delicate samples. When the specimen was mounted it was cleaned by holding it in hot trichlorethylene vapour to remove surface grease. Final cleaning was carried out prior to evaporation by subjecting the spectrosil to a high voltage slow discharge for ten minutes.

Evaporation of the Ge layer took place at a substrate temperature of 200°C . Twenty three quarter waves at 4μ were deposited.

After the first layer of Ge had been deposited the specimen was removed from the vacuum chamber in order to turn it over so that the second surface could be coated. When the half coated spectrosil was demounted considerable bending occurred due to stress in the thick evaporated Ge film. However, it was found that the spectrosil could be pressed flat without fracture occurring by replacing it in the copper holder and gently screwing the copper plug down onto it until it regained its original shape. The specimen remained flat when both sides had been coated with Ge.

The transmission of this spectrosil/germanium filter is shown in Figure 3.28. The second order peak transmission of 43% is quantitatively consistent with the measured absorption in the spacer layer at 100cm^{-1} . Another well defined passband occurs at $\sim 140\text{cm}^{-1}$ (71.5μ) and corresponds to the third order. Minimum transmission of 5% at 125cm^{-1} also agrees with calculations of the rejection based on indices of 2.2 and 4.0 for spectrosil and Ge respectively.

Although some success had been obtained with the spectrosil mounted in the above manner, it was decided to construct a special holder for the thinner quartz specimens in order to prevent bending taking place as the thick film is deposited. The quartz was placed in a recess between two thin copper plates so that, after one side had been coated, the other side could be presented for coating without removing the quartz from the holder. Despite this precaution, the fragile quartz did not survive coating and this approach had to be abandoned. It is thought that the stress in the thick Ge films was sufficient to crack these thin plates by forcing them against the edges of the holder. The difficulties in handling such thin plates are very great and any weaknesses due to the polishing process are immediately revealed when the plates are subjected to the stress in the coatings.

Multilayer evaporated filters

The extension of multilayer techniques from the visible and near infra-red to the far infra-red presents several problems. Apart from the obvious difficulty of obtaining suitable high and low index materials which are transparent in the region of interest, a major problem lies in the mechanical instability of the thick layers. The stress in such films is considerable and often prevents the formation of the multilayer. The magnitude of the stress can be estimated from the results of the previous section where it was observed that the spectrosil spacer was distorted by a quarter wave of Ge at $90\ \mu$.

From the point of view of filter stability, therefore, it is clearly desirable that the high and low index materials in the multilayer should have similar mechanical properties. This is not always possible and, indeed, in the case of Ge and CsI the mechanical properties of the materials are very different. Table 4.4 below lists values of some of these properties for Ge, Si, CsI,

Early attempts at bandpass filters in the far infra-red made use of Ge/ZnS and Ge/SiO systems. Figure 4.13 shows the transmission of filter D15

between 40 and 80 μ . This is a 6 layer Ge/ZnS filter on a Ge substrate of design Ge/LHLHHLH. In the manufacture of D15 the evaporation sources were stationary on either side of the centre of the vacuum chamber and 3.0 ins. apart. The Ge substrate was attached to the copper block 15in. above the sources and the substrate temperature was held constant at 100°C during deposition of the layers. The filter was originally intended to be centred at 60 μ and each quarter wave layer consisted of $9\lambda/4$ at $\lambda = 6.667\mu$.

Filter D15 demonstrated the feasibility of Ge/ZnS interference multilayers for the far infra-red and, later, more attempts were made to produce evaporated multilayers at 75 and 85 μ . These were mainly Ge/ZnS systems although one attempt (which failed) was made at a Ge/SiO filter. In particular, the following designs were tried:

D. H. W.	Ge/ <u>LL</u> HLH <u>LL</u> H
F. P.	Ge/ <u>LHLHLL</u> HLH

The table below lists the filters and deposition temperatures of the materials. The ZnS source was rotated during manufacture of filters L. W. 5. - L. W. 9 to improve the uniformity. For the earlier filters the sources were stationary, as in D15. Monitoring took place at 4 μ .

Filter	Design	Temperature °C		Comment
		Ge	ZnS	
D15	Ge/LHL <u>HHLH</u> at 60 μ	Ge 100	ZnS 100	Intact but appears rough
L. W. 1	Ge/LLHL <u>LLH</u> at 75 μ	Ge 200	SiO 100	Filter lifted from substrate
L. W. 2	"	Ge 200	ZnS 100	Intact but rough
L. W. 3	"	Ge 200	ZnS 100	Intact but rough
L. W. 4	Ge/LHLHLL <u>LHLH</u> at 85 μ	Ge 160	ZnS 160	Intact but rough
L. W. 5	"	Ge 100	ZnS 100	Filter lifted
L. W. 6	"	Ge 100	ZnS 100	Intact but rough
L. W. 7	Ge/LHLH <u>HHLH</u> at 85 μ	Ge 160	ZnS 100	Not completed because of contamination
L. W. 8	"	Ge 160	ZnS 100	Intact but rough
L. W. 9	Ge/LHLLH	Ge 120	ZnS 100	Lifted

From these results it would appear that Ge/ZnS multilayers have a greater chance of survival if the Ge deposition temperature is high. Thus, all four filters (excluding L. W. 7 which was not completed) in which the Ge was deposited at a substrate temperature of 160°C, or higher, survived, whereas only 50% of the remainder were successfully completed.

The importance of substrate temperature control to the survival of these multilayers for the far infra-red is further demonstrated by an experiment which was carried out during the manufacture of the Ge/SiO filter L. W. 1. A second substrate was placed in a conventional workholder and allowed to rotate around the copper block to which was attached the primary substrate. During evaporation the temperature of the copper block was controlled so that the Ge was deposited at 200°C and the SiO at 100°C. No temperature control of the rotating substrate was attempted. Although both filters failed to survive, it was observed that the multilayer on the rotating substrate lifted after the layers Ge/LLH had been deposited. On the other hand, the filter on the copper block did not lift until the final layer.

Filters L. W. 3 and L. W. 4 are typical of the Ge/ZnS multilayers which were successfully completed and the results of measurements with a Grubb Parsons IRIS interferometer are shown in Figures 4.14 and 4.15. The maximum and minimum values of transmission (corrected for transmission of the Ge substrate) are given below:

Filter	Type	Tmax %	Tmin %	Peak Wavelength μ
L. W. 3	D. H. W.	30	8	84
L. W. 4	F. P.	30	7	92

The Fabry Perot filter L. W. 4 is the more symmetrical and is, as expected, narrower with a halfwidth of about 10 μ . Some allowance was made in the manufacture of these filters for the increase in the refractive index of ZnS due to its restrahl. Generally, the optical thickness (at the monitoring wavelength) of the ZnS was around 10% less than the corresponding thickness of the Ge layers. For example, each quarter wave of ZnS in L. W. 4 consisted of 19 $\lambda/4$ at $\lambda = 4 \mu$ whereas each Ge quarter wave was 21 $\lambda/4$ at $\lambda = 4 \mu$. In this way it was attempted to minimise the mismatching between the layers

Although the results of section 4.2.2 indicate that thick Ge layers should be deposited at about 200°C such a high temperature is not desirable for ZnS

deposition. Accordingly L. W. 3 and L. W. 4 were deposited under different substrate temperature conditions in order to discover whether a compromise temperature existed and what difference, if any, it produced in the filters. The surface of both multilayers, in fact, appeared visibly very rough and when examined under a microscope the final Ge layers showed signs of preferential growth at many isolated sites. Indeed all the multilayers had the same rough surface. The scattering due to the surface roughness is possibly a contributory factor in the poor optical performance of these filters.

It may be argued that a high Ge deposition temperature leads to the layers becoming increasingly more granular. However, in that case, it would be expected that LW6 should be noticeably less rough than, say LW3, which is contrary to observation. It is more likely that the roughness of the layers is due to the nature of the evaporation process since it can be shown that the surface roughness of a random deposit increases as the square root of the film thickness. Certainly, neither ZnS source rotation to improve uniformity of deposition nor reduction in deposition temperature (both of which were employed in L. W. 6) significantly improved the surface or optical performance of the multilayers. The transmission of L. W. 6 is shown for comparison with L. W. 4 in Fig. 4.16.

It has been demonstrated that it is possible to evaporate stable Ge/ZnS multilayers of up to eight layers at $85\ \mu$. This is possibly due, in a large part, to the peculiar nature of ZnS films which are under compressive stress as opposed to the tensile stress in films of other materials such as Ge and SiO₂. Thus, the inclusion of ZnS in a multilayer will tend to have a stabilizing influence. However, the optical performance of these multilayers is, in general, poor and they only approximately resemble bandpass filters. Their poor performance is possibly a result not only of the scattering in these thick films but also of the absorption which measurements showed is still present in ZnS films at these wavelengths.

Ge/CsI multilayers

Caesium iodide is a useful infra-red material which is often employed as a prism in spectrometers out to $50\ \mu$. As CsI is ionic with its Restrahls at $\sim 145\ \mu$, the limit of its long wavelength transparency, in bulk form, is given by its cut off at $70\ \mu$. However, it will be shown that, in thin film form, Ge/CsI multilayers can provide useful filtering at $75\ \mu$ and also in the case of low pass filters still transmit at $100\ \mu$.

The transmission of a film of CsI on silicon was measured with the IRIS interferometer and is shown in Figure 4.11. The optical thickness of the CsI layer is $19\ \lambda/4$ at $\lambda = 4\ \mu$. It can be seen that the silicon is anti-reflected by the CsI layer. The interference fringes due to the substrate are also evident in this Figure.

Figures 4.17 and 4.18 show the transmission of the 2 and 4 layer stacks Si/LH and Ge/LHLH, respectively, where Ge forms the high index layers and CsI forms the low index layers. The transmission of these stacks corresponds to $n_H/n_L \approx 3.1$ or $n_L \approx 1.3$ if n_H is taken to be 4.0. Thus, it would appear that the index of the CsI films is quite low in the region 70-100 μ . That the refractive index at $75\ \mu$ must be considerably less than the near infra-red value can also be seen from the fact that the spacer layer of filter LW19/3 consisted of $49\ \lambda/4$ at $\lambda = 3.46\ \mu$. Since the filter pass band occurs at $73\ \mu$, and not $85\ \mu$ as one would expect in the absence of dispersion the change in index, or dispersion, is $\sim 13\%$.

Thus, CsI is a useful low index material and, by itself, can be deposited on Si in thick layers without difficulty (the $19\ \mu$ film in Fig. 4.11 was deposited at a substrate temperature of 100°C). However, initial attempts at combining CsI with Ge in multilayer stacks met with failure due to the multilayers lifting and crazing. It can be seen from Table 4.4 that the thermal diffusivity (h) of CsI is much less than that of Ge and so temperature gradients in the CsI films will be correspondingly greater. Also the linear coefficient of expansion (α) of CsI is almost ten times greater than Ge. This leads to considerable stress being present in the multilayer due to unequal expansion in the layers. On the other hand the lower value of Young's

modulus for CsI will tend to reduce the stress in the CsI film compared with the stress generated in the Germanium layers.

It is not surprising, therefore that difficulty was experienced with these two materials since their mechanical properties are so very different. Nevertheless, it was found possible to produce stable multilayers by depositing the Ge layers at 200°C and CsI at 100°C. This is in agreement with the earlier results for thick Ge films and suggests that a degree of annealing had taken place in the Ge layers. In addition, there is some evidence that the rate of change of temperature is important. This is to be expected since the materials such as CsI which have a high linear coefficient of thermal expansion are subject to thermal shock. The direction of temperature change is also important as these materials can withstand a more rapid rate of rise than the rate of temperature drop.

In the manufacture of successful blocking filters (see next section) like LW15 the average rate of temperature rise was 4.5°C/min and the rate of temperature drop was held at 3°C/min. However, even these rates were thought to be excessively high and in the narrow band pass filters LW19 the rate of temperature rise was reduced to 2°C/min. The temperature was decreased gradually by allowing the multilayer to cool down overnight from 200°C to ambient.

Even with these precautions, failures still occur and further work is required on the problem. *

Long wave bandpass filters

Figures 4.19 and 4.20 show the results of four bandpass filters, two each aimed at 75 and 85 μ which were produced by the high vacuum evaporation of Ge and CsI layers on silicon substrates.

The filters LW19/3 and LW19/1 in Figure 4.19 are multilayers of the following design.

* Similar difficulties are reported in Sections 4.3 and 4.4 as a result of experience in the Reading plant.

PL/Si/LHLLHP'

where L is $\lambda/4$ of CsI at 75μ

H is $\lambda/4$ of Ge at 75μ

P is a thin protective coating of Ge (evaporated) covered by a thin brushed on layer of polystyrene.

P' is a thin protective layer of brushed on polystyrene.

The protective polystyrene layers were formed by brushing on to the surface of the filters a 4% solution of polystyrene in toluene. The solvent rapidly evaporates leaving a thin layer of polystyrene on the filter. Some protection from the atmosphere is desirable for these filters because of the softness and hygroscopic nature of CsI.

Each L and H layer consisted of $25 \lambda/4$ at $\lambda = 3.46 \mu$ and the spacer LL layer was $49 \lambda/4$ at $\lambda = 3.46 \mu$. Monitoring was carried out in the odd orders to give the maximum change in reflectance. This is important since deposition of the layers was monitored continuously on one substrate and the scattering, which increases with total deposit thickness, tends to reduce the signal amplitude progressively.

Peak transmission of both filters is greater than 50% and therefore meets the specification.

Halfwidths of the filters are also within specification. Filter LW19/3 has a halfwidth of 5μ and LW19/1 has a halfwidth of 5.5μ .

Table 4.5 summarises the performance characteristics of each filter.

The last column of Table 4.5 gives the effective reflectance (R) of the quarter wave layers in each filter as calculated from the halfwidth using the equation

$$\frac{\Delta \lambda^{\frac{1}{2}}}{\lambda} = \frac{(1 - R)}{\eta \sqrt{R}} \quad (4.2)$$

If the quarter wave stacks in the filter are considered separately as in the following system

$$\text{Si} / \begin{array}{c} \text{LH} \\ \text{II} \end{array} ; \underline{\text{LL}} ; \begin{array}{c} \text{H} \\ \text{I} \end{array}$$

and if the low and high index layers are assumed to have indices of $n_L = 1.3$ and $n_H = 4.0$, respectively then the reflectances of systems I and II are given by:

$$R_I = 0.72$$

$$R_{II} = 0.85$$

The effective reflectance of the filter to be used in equations 4.1 is therefore $\sqrt{R_I R_{II}} = 0.785$ which is in good agreement with the last column of Table 4.5.

Thus it is clear that the refractive index of CsI is very low at 75μ compared with its value of 1.8 in the near infra-red. This is due to the influence of the CsI Restrahl at 145μ .

Figure 4.20 shows the results of bandpass filters LW23/4 and LW23/5 at 85μ . The design of these filters is similar to LW19, viz :

$$\text{Si} / \text{LH} \underline{\text{LL}} \text{H}$$

where, as above, L is $\lambda/4$ of CsI at 85μ

H is $\lambda/4$ of Ge at 85μ

and the reverse substrate surface was not anti-reflected.

The transmission of filters LW23/4 and LW23/5 is poor and, on correction for substrate transmission, not greater than 20%. This poor transmission is mainly attributed to the increase in absorption in the CsI layers at 85μ as a result of the CsI restrahlen band and no attempt was made to produce a Ge/CsI multilayer bandpass filter at 95μ .

Thus, these results indicate that the limit of usefulness of CsI in narrow band filters is approximately 75μ . At this wavelength the refractive index of

CsI is ~ 1.3 but is rapidly decreasing with wavelength as the influence of its Restrahl becomes stronger.

Far infra-red low pass filters

The specification did not state any integrated rejection requirements. All the filters have a minimum transmission of about 2% in the rejection region on either side of the pass band but have wide transmission bands well away from the fundamental passband - beginning at 58μ or less. The increasing absorption in the CsI limits the long wavelength transmission (at least out to 140μ) but a series of blocking filters has also been developed to reject the short wavelength radiation.

These blocking (or low pass) filters are in two parts as follows:

- (i) A Ge/CsI/crystal quartz short wave blocking filter which blocks by multilayer interference and germanium absorption at all wavelengths up to 5μ and with quartz absorption between 5 and 40μ .
- (ii) A Ge/CsI multilayer low pass filter on silicon with a sharp edge at 60μ which blocks by multilayer action from 40 to 60μ .

The combination of these two can remove all the sidebands in any of the filters at 75μ .

Figure 4.21 shows the transmission of filter AQ/2 which provides blocking at all wavelengths shorter than the natural crystal quartz cut on at about 40μ . The design of this multilayer is

$$\text{IV III / Q / I II}$$

where Q stands for the Z cut crystal quartz substrate of diameter 1 inch and thickness 1.5mm and systems I - IV are four multilayer quarter wavelengths Ge/CsI stacks as follows:

- System I : 11 layer stack HLHLHLHLHLH
 where H is $\lambda/4$ Ge at $\lambda = 3.88 \mu$
 L is $\lambda/4$ CsI at $\lambda = 3.88 \mu$
- System II : 12 layer stack LHLHLHLHLHLH
 where H is $\lambda/4$ Ge at $\lambda = 1.65 \mu$
 L is $\lambda/4$ CsI at $\lambda = 1.65 \mu$
- System III : 11 layer stack HLH LH
 where H is $\lambda/4$ Ge at $\lambda = 2.45 \mu$
 L is $\lambda/4$ CsI at $\lambda = 2.45 \mu$
- System IV : 12 layer stack LHL LH
 where H is $\lambda/4$ Ge at $\lambda = 4.7 \mu$

The rejection regions of these four multilayer stacks overlap to provide blocking from $1.5 - 5.5 \mu$. In this region the transmission is less than 0.0005. The germanium content of the multilayers is sufficient to absorb all wavelengths shorter than 1.5μ .

The transmission of AQ/2 closely follows that of the crystal quartz between 40 and 100μ , being not less than 90% of the transmission of quartz at any point in this region. That adequate blocking of the short wavelength radiation is provided by this filter was demonstrated by using it in an interferometer and obtaining a good interferogram with no other auxiliary filters present. However, the CsI in the multilayer stacks results in significant absorption beyond 100μ so that the region of high transparency of this type of filter is limited to $50-110 \mu$, approximately. At these wavelengths the transmission is about 35% and maximum transmission of nearly 60% occurs in the region of 80μ .

These points are illustrated by Figure 4.22 which gives the far infrared transmission out to 50 cm^{-1} of filter AQ/4 as measured by the interferometric spectrometer. The increase in absorption due to the CsI Restrahl is evident from the steady decrease in transmission beyond 120 cm^{-1} . The

maximum absorption is seen to occur at 65cm^{-1} . Thereafter, the transmission increases.

In addition, Ge/CsI multilayers were developed to block the sideband transmission of the narrow band pass filters between the quartz cut on at about $40\ \mu$ and $60\ \mu$. These filters consist basically of an 11 layer tapered stack on a silicon substrate. Figure 4.23 shows the transmission of an early filter LW15/2 which was made to the following design;

$$\text{PL}' / \text{Si} / \frac{\text{L}}{2} \text{HLHLHLHLH} \frac{5\text{L}}{2} \text{P}$$

where L is $\lambda/4$ CsI at $\lambda = 38\ \mu$
 H is $\lambda/4$ Ge at $\lambda = 38\ \mu$
 L' is $\lambda/4$ CsI at $\lambda = 75\ \mu$

and P is a thin protective layer of evaporated germanium.

The peak transmission of LW15/2 is 71% at $70\ \mu$ and the stop region (5% transmission points) extends from $28.6\ \mu$ to $47\ \mu$. For comparison, the theoretical transmission of the 11-layer tapered stack on a semi-infinite silicon substrate, i.e. the design $\text{Si} / \frac{\text{L}}{2} \text{HLHLHLHLH} \frac{5\text{L}}{2}$, is also shown in Figure 4.23. For the purpose of the calculation, the value of the high and low index layers were assumed to be 4.0 and 1.6, respectively and the filter was taken to be centred at $38\ \mu$.

The theoretical edge is seen to be 2.5 times steeper than the cut on slope of filter LW15/2. This relatively poor edge of LW15/2 is attributed mainly to poor uniformity of the evaporated layers since in the manufacture of the filter both the evaporation sources were stationary near the centre of the vacuum chamber. The stationary sources, although placed within 3 inches of each other, results in a thickness gradient across the filter surface.

The deposition temperatures of the high and low index layers in LW15/2 were 200°C and 100°C , respectively.

Although filter LW15/2 demonstrated the feasibility of Ge/CsI low pass blocking filters for region of the infra-red, it did not fully suppress the side-

bands of the bandpass filters since the cut on point occurred at $\sim 47 \mu$. To be completely effective the blocking filter should have a sharp edge at around 56μ and its stop region should extend to wavelengths shorter than the quartz edge.

Figure 4.24 is filter LW26/2 showing the full extent of the stop region, $175\text{cm}^{-1} - 295\text{cm}^{-1}$ or $57 - 36 \mu$, which links up with the quartz cut on. The design of LW26/2 is identical to LW15/2. The edge has been shifted to 57μ by adjusting the thicknesses of each L and H layer to be $13 \lambda/4$ at $\lambda = 3.46 \mu$. (In LW15/2 the L and H layers consisted of $11 \lambda/4$ at $\lambda = 3.46 \mu$).

The CsI source was rotated on the slipring assembly during the manufacture of LW26/2. This resulted in better film uniformity, although the Ge source was stationary at the centre of the vacuum chamber, and the edge of LW26/2 is much steeper than LW15/2 and comparable to the theoretical slope in Fig.4.23. Again, maximum transmission is 52% at 132cm^{-1} ($\sim 75.7 \mu$) and, thereafter the transmission decreases steadily to a minimum of 2% at 65cm^{-1} due to the influence of the CsI Restrahl.

In the design of filters LW15/2 and LW26/2, the final layer is a thick film of CsI which serves the purpose of anti-reflecting the filter at 75μ . This has several disadvantages as follows:

- (a) The thick final layer of CsI results in increased absorption at longer wavelengths
- (b) CsI is a soft material and thus easily marked and damaged
- (c) The index of CsI film at 75μ is about 1.3 and therefore too low to provide complete anti-reflection of the multilayer and silicon substrate at these wavelengths.

Accordingly, it was decided to replace the final CsI layer with the 3 layer system $\frac{H}{2} L \frac{H}{2}$ where L and H are $\lambda/4$ at $\lambda = 38 \mu$. In this way

- (i) The total amount of CsI in the filter is reduced by almost 20%
- (ii) The final layer is a thick hard germanium layer

- (iii) The equivalent index of the combination $\frac{H}{2} L \frac{H}{2}$ at 75 μ is higher than the CsI index and provides a better anti-reflection coating for both silicon substrate and multilayer.

Figure 4.25 illustrates the transmission curve of filter LW28/1 as measured by the Grubb Parsons - NPL cube interferometer. The design of LW28/1 is as follows:

$$P \frac{H'}{2} L' \frac{H'}{2} / Si / \frac{L}{2} HLHLHLHLH \frac{L}{2} \frac{H'}{2} L' \frac{H'}{2} P$$

where L is 13 $\lambda/4$ at $\lambda = 3.46 \mu$.

H is 13 $\lambda/4$ Ge at $\lambda = 3.46 \mu$.

L' is 11 $\lambda/4$ CsI at $\lambda = 3.46 \mu$.

$\frac{H'}{2}$ is 5 $\lambda/4$ Ge at $\lambda = 3.46 \mu$.

$\frac{L}{2}$ is 7 $\lambda/4$ CsI at $\lambda = 3.46 \mu$.

and P is a thin brushed on coating of polystyrene to give added protection from atmospheric moisture.

According to Fig. 4.25 the peak transmission of LW28/1 is 58% which is significantly higher than LW26/2. Also the transmission at longer wavelengths is generally greater than that of LW26/2 although it follows the same pattern i.e. decreases to a minimum at 65cm^{-1} (154μ). These improvements, together with the added advantage of having a robust germanium layer on the exterior of the multilayer, would seem to justify the use of the $\frac{H}{2} L \frac{H}{2}$ combination as an antireflection system.

Filter LW28/2 was manufactured in the same way as LW26/2. The CsI source was rotated on the slip ring assembly and the high and low index layers were deposited at 200°C and 100°C , respectively. The cut on points (5%T) of these filters are almost identical at 57μ and the rejection region extends to 36μ to link up with the quartz cut off. Thus, the combination of these filters with the quartz filters like AQ/2 are sufficient to fully suppress all short wavelengths sidebands of the narrow bandpass filters at 75μ .

An interesting example of the capabilities of these multilayer interference filters is illustrated in Figure 4.26 which shows the interferogram obtained with the filter combination LW19/1 + LW28/2. Filter AQ/2 was not required for this experiment since the Golay detector in the instrument (the Grubb Parsons - NPL cube interferometer) had a quartz window which, with black polythene, provided auxiliary filtering below $40\ \mu$. The resultant transmission consisted of only a narrow band at $75\ \mu$ with a halfwidth of $5\ \mu$.

For a monochromatic line source the interferogram is simply a sine wave and if the detected radiation consists of more than a single wavelength the interferogram becomes progressively more complicated. Thus, although there is a gradual decrease in amplitude away from the central maximum (which corresponds to zero path difference), the persistence of the fringes over a considerable path difference in Fig. 4.26 is a good indication of the monochromaticity of the spectral energy distribution produced by the filter combination. It is believed that this is the first time such an interferogram has been produced with multilayer interference filters.

4.2.4 Summary

These results show that evaporated multilayer interference filter techniques have been extended to produce narrow bandpass filters at $75\ \mu$ which meet in part the specifications of the contract. In addition, low pass blocking filters have been made with useful transmission out to $100\ \mu$ and sharp-cut on edges at $60\ \mu$.

Altogether, eighteen bandpass and low pass filters for the far infra-red have been produced using *Ge and CsI as the high and low index layers, respectively. These are held at Reading University and are listed in Table 4.6. The Reststrahlen absorption in the CsI prevents this material from being used in bandpass filters beyond $75\ \mu$. More work is therefore required to find a suitable low index material for wavelengths greater than $75\ \mu$.

* Patent applied for.

REFERENCES FOR SECTION 4.2

- 4.4 Roberts and Coon. J. Opt. Soc. America, 52 (1962) p.1023
- 4.5 Russell E. E and Bell G. E., J. Opt. Soc. America, 57 (1967)p.341.

4.3 EVAPORATED FILTERS AT READING

4.3.1 Blocking filters to link with crystal quartz

A multilayer filter (or filters) is required to link with the lower edge (at about 5 microns) in crystal quartz to provide integrated rejection for all wavelengths below 40 microns, low pass action being required above 40 : microns. Calculations show the following:-

Materials	Index Contrast	Stopband width (infinite stack)	Stacks required	Location (microns)	Transmission in absorption region.
PbTe/ ZnS	2.33	52%	1 of 16 layers	3.93	About 3×10^{-4} at 3.5 microns in 8 layers of PbTe
PbTe CsI	3.1	68%	1 of 12 layers	3.60	Sufficiently low in 6 layers of PbTe
Ge/CsI or CsBr	2.33	52%	3 of 12 layers		Sufficiently low in 18 layers of Ge below 2 microns.

A single stack relying on absorption in the PbTe layers is clearly an attractive proposition provided that either longwave transparency of PbTe and ZnS is satisfactory or PbTe/CsI is a viable combination. Furthermore, if this stack is deposited on a high index substrate, antireflection will be required at selected lowpassing wavelengths. (Use of quartz substrates, which can be assumed to be of sufficiently low index not to require antireflection would, therefore, be preferable. In fact, test depositions proceeded on Ge before crystal quartz substrates were available). Measurements of low-passing performance at shorter and at longer wavelengths are shown in Figs. (4.28-4.36) for the following combinations:-

- (a) SUB : (LH)⁸ [L ■ ZnS H ■ PbTe]
- (b) As (a) but with a single antireflection layer of CsI at 75 microns, and 85 microns on both faces, and (for comparison) with antireflection of CsBr.
- (c) SUB : (LH)⁶ [L ■ CsI H ■ PbTe]
- (d) Antireflection (both faces) of CsI, at 75 and 85 microns.

It should be noted that (a) (b) and (c) require the addition of quartz to make a 50 micron blocking filter (see insert on Fig. 4.29).

Conclusions are drawn from these measurements as follows:

- (1) A single stack of PbTe/Zns provides blocking to better than 10^{-3} in combination with quartz for the complete range to the far side of the absorption band in quartz. Its longwave pass properties are suitable to 80 microns.
- (2) PbTe/CsI as a combination gives low transmission and it is not viable at present for reasons which are not apparent.*
- (3) CsBr is not suitable for use in longwave filters.
- (4) CsI provides no increase in transmission as an antireflection layer beyond 80 microns. It should be noted with reference to this fact that the spectral location of peak transmittance vs physical thickness of applied antireflection coatings is in overall proportion to the measured dispersion in CsI (see Fig. (3.12))

4.3.2 Equal ripple lowpass filter employing fractional layers

A test deposition has been made of the PbTe/ZnS design discussed in Section : 5.1.4. The intention was to provide improved lowpassing at 12

* In spite of a variety of deposition temperatures which extended between 80° and 120°C for the CsI and between 200° and 300°C for the PbTe. Mechanical instability (i.e. peeling) gave considerable difficulty.

microns in comparison with a standard filter (i. e. modified Herpin-type design) made with the same materials. On the basis of the monitoring wavelength pre-selected at 3.9 microns. deposition in fourth order can yield all necessary fractional thicknesses (as shown in Table :5.1) with a chopping angle adjustment^r varied between 160° and 280° (the monitored substrate remaining unchopped throughout). The chopper is constructed from a pair of sectorised discs (angle $\sim 180^{\circ}$), one of which is rotatable with reference to the other, attached to a coaxial pair of rotary seals (1" dia. and 1/8" dia.) the whole being locked and rotated at 500 r. p. m.*

The fractions needed here have been assessed in respect of possible thickness corrections for :-

- (i) temperature coefficients of refractive index and thickness for deposition at 160°C ;
- (ii) angle of incidence of monitoring beam equal to 20° ;
- (iii) dispersion in refractive index inherent in monitoring a 12 micron filter at 4 microns;
- (iv) polar distribution of evaporant at filter, and monitor, positions.

Corrections required to reduce possible errors in thickness due to the above are as follows:

* The arrangement can also be used with a disc of fixed angle and the filters described in the previous Section were monitored in second order and chopped back in thickness by a factor of about 2:1 to obtain the desired location.

+ See Section 3.2.1.

	PbTe	ZnS
(i) Temperature	+1.4%	-0.7%
(ii) Angle of incidence	0	+1.2%
(iii) Dispersion	-4.5%	-3.5%
(iv) Polar distribution	+1.4%	-1.0%
Nett	-1.7%	-4.0%

As a result the fractions for PbTe were increased by 2.3% and the chopping angles (of obscuration) recalculated as:-

Layer	1	2	3	4	5	6	7	8
	L	H	L	H	L	H	L	H
Fraction	.545	.815	.905	.955	.985	.985	.99	1.0
Angle in 4th order	251°	191°	177°	161°	165°	155°	161°	153°
Layer	9	10	11	12	13	14	15	
	L	H	L	H	L	H	L	
Fraction	.99	.98	.96	.92	.805	.545	2.0	
Angle in 4th Order	163°	157°	169°	169°	197°	247°	About 8th order	

The filter was deposited at 160°C, without temperature cycling, to minimise errors in monitoring associated with temperature-induced thickness changes. Measured performance is shown in Fig.(4.37) and is seen to be in good agreement with calculation (Fig.(5.5)); the improvement over the more usual Herpin-type structure offers encouragement for future development.

Also of interest for possible use with this, or other designs, is the antireflection coating L:HH:SUB (or STACK:SUB) which in comparison with L.SUB, has improved transmission (Fig:(4.38)) and (for L ■ ZnS, H ■ PbTe SUB ■ Ge or Si) improved mechanical and environmental ruggedness .

4.4 EVAPORATIONS AT READING:

"Annealing" of PbTe monolayers and multilayers

4.4.1 Preliminary depositions

Evaporations of the 15 micron Fabry-Perot filter (a production item for the Radiometer as described in Section: 4.1.3) were made:

- (a) to verify the temperature-cycling conditions were genuinely optimum on transfer into a different evaporating plant;
- (b) to determine ultimate transparency and pitching-accuracy.

Immediate problems arose merely in attempts to reproduce results obtained in the Balzer plant at Grubb-Parsons, particularly with regard to transparency. Careful checks were then made of any possible relevant parameter, and the following aspects (although essential in good vacuum practice and therefore to be retained in the present case) were eliminated as not being those parameters which are vital for the deposition of PbTe:-

Continuous trapping of pump-vapour, outgassing vapours etc.
with liquid N₂.

Rigorous chamber cleaning before all evaporations, plus use of fresh source material.

Evaporation of multilayers with no significant loss of vacuum (e.g. overnight).

Shielding of hot Cu surfaces as protection against re-evaporation.

Separate shutters over sources.

Cooling, at chamber input, of hot-running heating electrodes.

The above represents, as a combination, more stringent precautions than is usual at Grubb-Parsons. It follows that semi-absorbing layers are not the result of chemical contamination, other than from within the source charge itself (i. e. boat material, loss of stoichiometry within the charge, contamination by other evaporating material). Next, checks were instituted to collect on to subsidiary substrates various combinatory layers during the evaporation of a complete filter. This was done for the following break-down (not necessarily from the same evaporation):-

10 LAYER FABRY-PEROT	SUB :	L : H : L : H : L : HH : L : H : L : H
SUBSIDIARY FABRY- PEROT		L : H : L : H : L : : L : H : L : H
		L : H : L : H : : : H : L : H
		L : H : L : : : L : H
* ALL ZnS LAYERS		L : : L : : L : : L : : L :
* ALL PbTe LAYERS		: H : : H : : HH : : H : : H

* For comparison with layers continuously evaporated in absence of other source.

Results (Fig:(4.39)) showed without doubt that lack of transparency (i. e. absorption) was the sole difficulty, distributed throughout the PbTe layers; further, that contamination across layer interfaces is not important and any cross-contamination between sources is not significant. Absorption in the PbTe was deduced to be due to the presence of excess free carriers. Again a check was made with PbTe from the same batch of material, as between evaporation in the two plants, that the difficulty was solely introduced at Reading. By calculation, on a basis of optical measurements to be discussed later, free carrier concentration in the best Grubb-Parsons production filters is lowered from about $10^{19}/\text{cm}^3$ (p-type) in source material (as prepared) to less than $10^{18}/\text{cm}^3$, and probably about $10^{17}/\text{cm}^3$, after deposition. Since further variations in the evaporation/deposition processes did not suggest

themselves, post-deposition heat treatment was tried on some test filters of inferior transparency. Immediate success was obtained as is shown in Table 4.7 below and in Figs: 4.40 (a), (b), (c).

Detailed "annealing" experiments were carried out on the basis of these preliminary results.

4.4.2 Detailed Annealing studies

4.4.2.1 Introduction

Information sought may be summarised as:-

- 1) Are improvement of transparency and shift expressible as changes in optical constants and are they separable in n and k ?
- 2) Can the annealing be described as a rate process (i.e. function of time $\times$ temperature) product ?
- 3) What is the physical nature of the process with reference to any threshold temperature, interrelation with the evaporation/deposition processes and initial conditions ?

4.4.2.2 Filters (first measurements)

Further experiments were made on the Fabry-Perot filters referred to in the previous sub-section with the intentions a) to determine whether tight specifications on peak positions could be met by using annealing as a technique for fine tuning adjustment, b) to determine the changes in optical constants. (With reference to b, filter characteristics are much more sensitive to small changes in comparison with single film transmittance characteristics).

Conditions used for the annealing were largely determined by the need to ascertain the presence, or absence, of threshold temperature(s) and the influence of the (time. temperature) product. Each experiment was carried to a conclusion before making a measurement, so that the process consisted of insertion into a pre-heated furnace for a chosen time interval, removal and natural cooling to room temperature. For insertion, the filters were mounted

on a copper block and the furnace was pre-heated to 4°C above the desired temperature. On one sample, a series of sixteen stages of heating were carried out in durations of about 10 minutes at temperatures progressively increasing from 130°C to 320°C : spectral measurements between 5 and $25\ \mu$ were taken after each stage of heating. Results from these experiments are not presented in detail : the following conclusions were drawn:-

- a) A threshold is not found by progressive increase of temperature
- b) annealing takes place more rapidly the more elevated is the temperature, which is typical of a rate process;
- c) although Fabry-Perot peaks are most sensitive as a measure of optical constants, improvement in transparency was first noted at the longest wavelength (in the extreme (i. e. lowpass) wing) confirming that the reduction in absorption is associated with a free carrier mechanism. Subsequently, the 1st order peak (at 15 microns) improves and lastly the 3rd order (at 5 microns). Shift in peak(s) occurs simultaneously with increase in peak transmittance, although the possibility remains that peak shift becomes immeasurably small before the whole of the improvement in transparency has been obtained. (Accuracy in shift measurement which was available was only $\pm 0.5\%$, i. e. insufficient at present to permit a final judgement on this point).

4.4.2.3 Filters (second measurements)

Another series of anneals were carried out at various constant temperatures. It was convenient to start at the temperature (270°C), found in the previous series, at which the 1st order peak became evident. A total time of 180 minutes was applied at this temperature, after which further changes did not take place and the process was regarded as complete : spectroscopic measurements were made at intervals (with cooling as before) of about 2 minutes. When the changes became difficult to measure the interval was increased to 4 minutes and 8 minutes (at 20 minutes and 52 minutes respectively). The consistency of a description of the annealing as a rate process was

checked by repeating the experiments at 298°C and 316°C (with 2 minute intervals) with other filters from the same batch. Detailed results of all measurements are presented in Figs: 4.41(a) and (b). (It should be noted that, in comparison with the first series, measuring accuracy had been improved by expanding the wavelength scale and using polystyrene and CO₂ calibration on the Perkin Elmer 237 instrument: non-uniformity was eliminated as a source of possible error and annealing temperature may be taken as accurate to $\pm 1^\circ\text{C}$).

Variations in total (time.temperature) conditions to complete the annealing process is as follows:-

Temperature °C	271	298	316
Completion time, mins. (on 1st order Fabry-Perot peak)	180(+20)	24(+3)	9(+2)

4.4.2.4 Discussion

No single best temperature is apparent for the annealing process, at least for the batch of filters deposited at 260°C. Using the description of a rate process, Fig.4.42 shows a plot of annealing time (log scale) vs temperature⁻¹: it is apparent that $\log_e(\text{time}) = K.T^{-1}$ within experimental error and in the range investigated. On the assumption that reaction rate varies as $\exp - \frac{\Delta E}{kT}$, the "apparent activation energy", ΔE , is 0.83 eV. The extent of the induced changes in optical constants can be inferred from the quantitative peak shifts and increases in T_{max} . A convenient way to make this deduction is to use computations of spectral performance for various fixed n and k assumed for the PbTe, with the assumption that all PbTe layers are affected by annealing but none of the ZnS layers. these computations (ranging between 5.40 and 5.64 for PbTe refractive index, and between 0.001 and 0.5 for PbTe

absorption index) are summarised in Figs:(4.43,4.44) .

The present status of annealing as a "tuning" technique may be summarised as unproven. Filters with low $T_{\max}$ do not always improve after heat treatment and filters deposited in a different situation (i. e. at Grubb-Parsons) either do not need the treatment or appear to be unresponsive to it. However, no apparent harm appears to result from this post-deposition heat cycling and it is still possible that tiny shifts (say about 1cm^{-1}) may be reproducibly obtainable after further development. This could become very significant.

The result is itself of interest and, interpretable in terms of semi-conductor properties, provides useful parameters for studying the effects of different conditions on film deposition. This line has been pursued on the simpler problem of monolayers.

4.4.3 Free carrier properties in PbTe as modified by annealing

4.4.3.1 Introduction

Measurement of vacuum deposited multilayers and single films have indicated that the major problem with structures containing lead telluride is one of excessive free carrier absorption . Further, it has been found that heat treatment alleviates this problem to some extent. It is of interest and use to understand the processes involved, to find carrier concentrations, sign of carriers and mobilities in freshly-deposited films and heat-treated films; hence, perhaps, to find a method of monitoring heat treatments, and find processes to give optimum values of optical constants.

Early work on PbTe summarised by R. A. Smith (4.6) found control of carrier concentration somewhat troublesome. Lawson (4.7, 4.8) added excess lead to produce controlled concentrations of electron carriers but always found p-type material. Oxygen impurity in the materials used to prepare PbTe was found to be the root of the trouble; it doped p-type and caused excess lead to be rejected. Using pure materials, excess Pb gives n-type material, excess Te

gives p-type. Certain impurities give rise to doping as follows:-

O_2 , Ag, Cu, Ni,	(Excess Te)	p-type
Bi	(Excess Pb)	n-type

Samples of PbTe heated in vacuo have been reported sometimes to become n-type.

N-type PbTe heated in Te vapour has been made p-type, and the same result has been achieved with O_2 . (4.9, 4.10). Te and O_2 are both group VI elements of valency two, lead is group IV but forms many bivalent compounds.

PbTe is reported to have the NaCl cubic structure with cube side $6.34\overset{O}{\text{\AA}}$.

PbS and PbTe are IV - VI compounds of similar structure and properties. It has been found that oxygen can be diffused into these materials turning n-type to p-type as with PbTe, but also that oxygen can be diffused out in vacuum (4.6).

Shogenji and Uchiyama (4.11) found resistivity of p-type PbTe increasing when heated in air and attribute the effect to evaporation of Te.

Experiments to determine equilibrium amounts of excess Pb and excess Te in various conditions have been performed. Brebrick and Gubner (4.12) heat-treated PbTe in vapours of Pb and Te at various temperatures until equilibrium was achieved. Pb vacancies and Pb interstitials are said to be the most plausible defects in each case. They find decreasing carrier concentrations with decreasing temperature, but increasing equilibrium times. For instance, excess lead produces,

$$\begin{array}{l} 3.3 \times 10^{18} \text{ elec./cm}^3 \text{ in equilibrium with vapour at } 775^\circ\text{C. and} \\ 7.7 \times 10^{17} \quad " \quad " \quad " \quad " \quad " \quad " \quad " \quad 500^\circ\text{C.} \end{array}$$

If Te or Pb is in excess of equilibrium at a given temperature it is suggested that it can be precipitated out internally at dislocations or grain boundaries as neutral atoms. Scanlon (4.13) studied this process in detail for p-type crystals with excess Te, annealing at a succession of decreasing temperatures to find an equilibrium at each. Annealing was in air, but he

reports the same results have been found in vacuum.

For p-type films it is possible to monitor thermoelectric power at annealing temperature; thus Scanlon could observe when equilibrium was reached. The thermoelectric characteristics of n-type materials are such that this is not possible for them.

His results are summarised below:-

Anneal Temp.	Time to equil.	N per cm^3	Mobility $\text{cm}^2/\text{V sec}$	ohms cms.
Initial cond.		3.1×10^{18}	870	.0023
385°C	2 hrs	1.2×10^{18}	760	.0067
370°C	4 hrs	1.0×10^{18}	680	.0091
294°C	40 hrs	4.8×10^{17}	690	.019
245°C	70 hrs	3.3×10^{17}	730	.026
208°C	400 hrs	1.95×10^{17}	790	.041
181°C	1000 hrs	1.5×10^{17}	715	.058

Experiments were performed on n-type PbTe rich in Pb which were annealed, quenched and measured, summarised below:-

Anneal process	Total Time	Room Temp. measurement of N
Initial sample		$7.8 \times 10^{17} \text{ elec/cm}^3$
At 400°C for 15 hrs.	15 hrs	$4.8 \times 10^{17} \text{ elec/cm}^3$
At 400°C for further 20 hrs	35 hrs	$2.5 \times 10^{17} \text{ elec/cm}^3$
At 400°C for further 24 hrs	59 hrs*	$9 \times 10^{16} \text{ elec/cm}^3$

* There was no evidence that equilibrium was being approached.

4.4.3.2 Experiments on Vacuum deposited monolayers

Properties

In comparing the properties of source material, deposited film and heat-treated film, variable parameters involved can be considered as the following:-

N = No. of free carriers per unit volume

m^* = effective mass of carriers

μ = mobility of carriers i.e. drift velocity per unit field

The important electrical properties of semi-conductors are:

I Electrical Conductivity $\sigma = N \cdot \mu \cdot e$

II Hall Coefficient $R_H = (N_e)^{-1}$

The optical properties due to carriers in a semi-conductor can be fairly well described by Drude-Lorentz classical theory of damped charged particles, if the damping constant, g , is simply related to the relaxation time τ , characteristic of the scattering process.

III $g = \frac{1}{\tau}$

In terms of this parameter the real and imaginary parts of the refractive index $n - i\kappa$ are given by,

IV $n^2 - \kappa^2 = n_L^2 - \frac{Ne^2}{\epsilon_0 m^* (\omega^2 + g^2)}$

V $2n\kappa\omega = \frac{g Ne^2}{\epsilon_0 m^* (\omega^2 + g^2)}$

where ω is frequency, n_L = refractive index contribution of the lattice in the absence of carriers. Lattice absorption is assumed small.

At high frequencies, $\omega^2 \gg g^2$,

and for low absorption $n^2 \gg \kappa^2$.

These reduce to simple relations for n and κ :-

VI Dispersion of refractive index

$$n^2 = n_L^2 - \frac{Ne^2}{\epsilon_0 m^* \omega^2}$$

VII Absorption index

$$\kappa = \frac{g Ne^2}{m^* \epsilon_0 2n \omega^3}$$

From IV and VI it will be noted that n can fall to zero at a frequency ω_p , known as the plasma frequency. This condition gives rise to complete reflectivity at the surface. From IV, ω_p is given by

VIII Plasma frequency

$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^* (n_L^2 + \kappa^2)} - g^2$$

or making the approximations of VI

IX

$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^* n_L^2}$$

A further optical property of interest is the rotation of the plane of polarisation of an electromagnetic wave by the free carriers.

X Faraday Rotation

$$\theta = \frac{N}{(m^*)^2} \cdot \frac{Be^3 \ell}{2nc \epsilon_0 \omega^2}$$

θ = angle of rotation

B = field

ℓ = path length in the sample

To compare optical data with electrical data there is the following relationship

XI

$$\mu = \frac{e}{m^*} \cdot \frac{1}{g}$$

though a note of caution should be introduced here.

Much of the early work on PbTe was performed on evaporated layers. They were found to be polycrystalline with static properties differing from those found by high-frequency electrical measurements due to the presence of grain surfaces (4.11, 4.15). Thus with such films static electrical measurements might be a useful guide to optical properties but could be confusing.

Conclusions from the annealing of filters

In further studies on annealing of monolayers a temperature of about 320°C has been consistently used, and the results of filter behaviour have been used to indicate the time required for a complete anneal, i. e. about 5-10 minutes. Longer periods have been used and no sign of overshooting an optimum have yet been observed. One further result on a filter should be mentioned here: in view of results in the furnace, a filter was left in the evaporation plant and raised in temperature to 320°C for 20 minutes. The absorption of the sample was high and was subsequently reduced by heating in the furnace, in air, at 320°C for 20 minutes. Improvements in thermal contact in the evaporation plant, increasing temperature to 350°C and time to 2 hours yielded the same negative result in vacuo, though the filter was later treated successfully in air.

This same sample after treatment in air, was re-annealed in the evaporation plant at 350°C and a pressure of 10⁻⁵ Torr for 2 hours to see if there was any reversal of the annealing process. No change in transparency was observed after this treatment except that absorption near 9 μ, usually found in silicon substrates, was reduced. (This absorption is due to oxygen on the surface of the silicon giving rise to an Si - O stretching mode). Thus oxygen was removed from the bare face of the substrate but presumably not from the PbTe if in fact any was diffused in previously.

Resistance Measurements

Resistance changes on annealing in air were examined as a convenient method of monitoring progress at high temperatures. A circular film was evaporated onto a quartz disc and contact made at opposite ends of a diameter using nickel wire and silver paint.

The film was 8 quarter-waves thick at 3.92μ i.e. about 1.3μ in physical thickness. This film was annealed in air and its resistance measured at intervals of 1 minute. The results are summarised in Figure 4.45 where resistance is plotted versus time, and temperature of a copper substrate holder is plotted versus time. In a circular film resistance is related to resistivity, by

$$R = \frac{\rho \pi}{2 t}$$

t = film thickness, ρ = resistivity.

The behaviour could be interpreted as a decrease of resistivity as temperature increases, which is reversed presumably when annealing becomes significant. The annealing leads to a change of carrier concentration and/or mobility which at 320°C reaches new saturation in under 10 minutes. Thus R becomes constant. Finally on cooling the resistance increases in agreement with its original trend on heating, leaving resistivity changed by a factor of 1000 in this particular film. The film was cycled from room temperature to 320°C twice subsequently in air, and though it became apparent there was a large thermal lag between thermometer and film the final part of Fig. 4.45 was broadly reproduced: Resistance fell, on heating, to about 10^5 ohms at 320°C and increased to 1.2×10^7 ohms at room temperatures, only 600 times the original value. The slight lack of correspondence on recycling might be due to surface oxydation. (This always occurs when PbTe is held at 320°C in air, without strongly affecting the optical transmission).

A resistance of 10^7 ohms for this film gives resistivity 840 ohm cm. The initial value of 2×10^4 ohms yields resistivity 1.68 ohm cm.

Even the latter is rather high compared with Scanlon's (4.13) values for crystals, he estimates intrinsic crystals to have $N_i = 2 \times 10^{16}$ carriers/cm³ and $\rho_i = 0.2$ ohm cm.

This large discrepancy may be due to crystallite structure of the film and large number of pinholes present in this particular sample. However, resistance measurement does seem to afford a more sensitive and easily monitored parameter than optical transmission for the purposes of following an annealing process.

A set of films on glass were prepared of approximately 2μ thickness with no pinholes apparent (Run No. 73). Owing to some curious fluctuations in resistance of a circular film at 320°C , these films were scribed and scraped away to leave a rectangular strip which did not fluctuate in the same manner. One of these samples was heated in a tube at a pressure of 0.1 - 0.2 Torr, to 335°C and held for 30 minutes. The contacts came loose and were remade after cooling, thus the slight increase in resistance (figure 4.46) may not be significant. The sample was heat treated in a nitrogen atmosphere containing approximately 99.5% N_2 with no effect. Finally on heating in air an immediate rise in resistance was observed taking about 5 minutes to complete at 335°C . This time is of the same order as times involved in filter examinations. Thus it appears that presence of oxygen is a necessity in the annealing process. It remains possible that an alternative process might occur over a very much longer period in vacuum - though such a process might be obscured by a slow oxygen process at reduced pressure. It would be of interest to heat-treat a film at high oxygen pressure to see if, or how, the rate of anneal and final state depend on the oxygen pressure.

Hall effect measurement

An unannealed sample from the same batch as the sample of Fig. 4.45, again in rectangular strip form, was contacted by silver paint. The dimensions were approximately 18 mm. by 1.8 mm., again about 2μ thick.

Twin contacts on one side of the strip enabled voltage at zero field to be "backed-off" and Hall voltage of 63 (± 5) was measured in a 5 kilogauss field with a current of 2.01 ($\pm .01$) ma. The change in voltage in the field occurred with a time constant of several minutes, which was not explained, and coupled with some drift in voltage across the sample at zero field led to some uncertainty in the result. This was limited as far as possible by interpolation between readings before and after applying the field.

Relation II was used and yields a value of carrier concentration directly. The sign of the Hall voltage showed the material was n-type with $N = 3.8 \times 10^{16}$ electrons/cm³.

In practice relation II is usually found to require a constant r such that

$$R_H = r. (Ne)^{-1}$$

r varies between 1 and 2 depending on the scattering mechanism (4.6). Thus the value given for N is probably a lower limit.

The resistance of the sample yielded a resistivity value of $\rho = 0.65$ ohm. cm.

From relation I and the value of N just calculated this gives a mobility of $\mu = 250$ cm²/Volt. sec., if r is unity, otherwise this value is reduced inversely with r . In all events μ is low and indicates either strong scattering or strong grain boundary effects in the film. Putting this value in relation XI yields $g = 8 \times 10^{13}$ sec⁻¹ for damping constant. This is equivalent to the frequency of 23.3 micron radiation and such a value would prevent approximations neglecting g^2 , as used in relations VI and VII for wavelengths > 10 microns. However, any infra-red measurement may well yield a smaller value of g , and the most significant deduction from this measurement is that the carriers are n-type in the film.

The state of the source material

The suppliers of the PbTe used, check each batch for carrier concentration by measuring the Seebeck coefficient using a hot probe method. Results are consistent from batch to batch to within about 25% and samples have p-type carriers with a concentration of approximately 10^{19} per cm^3 . (4.16). It would appear therefore that vacuum evaporation changes the sign of the doping of the PbTe.

Optical Measurements

All the relations for optical quantities involve m^* , the effective mass of the carriers, and this quantity has been determined by several investigators by measurements on single crystals, with carrier concentrations which have been determined electrically.

Walton, Moss and Ellis used Faraday effect measurements (4.17) to determine m^* for n-type PbTe.

N	m^*
3×10^{17} per cm^3	.086 $\pm$.004
2×10^{18} per cm^3	.130 $\pm$.008
3.3×10^{19} per cm^3	.132 $\pm$.010

All at room temperature.

Values for p-type material have been determined by Dixon and Riedl (4.18) and Lyden (4.19) using reflectivity measurements (plasma effect) and by Stiles, Burnstein and Langenberg using Cyclotron Resonance (4.20).

N	4.8×10^{19}	3.6×10^{18}	3.5×10^{18}	3.3×10^{18}	3×10^{17}
m^*	.18	.11	.10	.11	.042
Ref.	4.18	4.19	4.18	4.19	4.20

All at room temperature.

The variations in m^* over the range quoted are small enough to enable fitting of fairly accurate values of N to determination of N/m^* but if the carrier concentration given by our Hall measurement of 3.8×10^{16} electrons per cm^3 is accurate and typical, then there will clearly be some uncertainty in N because available m^* does not extend so low in a region where considerable change can be expected.

Faraday effect measurement

A sample of about 4μ physical thickness was evaporated onto Germanium held at 260°C for Faraday measurement. In transmission the film showed heavy free carrier absorption transmission falling to a few per cent at 25μ . The Faraday Rotation at 30 kilogauss was $4.1^\circ (\pm .8^\circ)$ at 551cm^{-1} (approx. 18μ). This leads to an n-type concentration of 4.2×10^{18} per cm^3 assuming $m^* = .13$. This is very much higher than the Hall measurement result (different sample), for which the conditions of deposition were similar in all respects except that the Faraday sample was on Germanium at 260°C and twice as thick as the Hall sample deposited on glass at 300°C .

The value of N from the Faraday rotation is slightly suspect however as the rotation may have been partly due to the Germanium substrate; the Germanium was about 0.5 mm thick (or over 100 X thicker than the film, it is conceivable that it might have of the order of 10^{16} carriers per cm^3). Any free carrier absorption it had was negligible compared with that in the film but if, as seems possible, the PbTe films exhibit a particularly strong scattering mechanism, and thus low mobility carriers, then it is possible that the film should exhibit much more absorption than the substrate, but comparable or smaller Faraday rotations.

Some typical values for Germanium are given by R. A. Smith (4.6, p. 97). For n-type carriers at 290°K , $\mu = 3900 \text{ cm}^2/\text{V sec.}$, $m^* = 0.3\text{cm}$.

Hall measurement gave values for PbTe of $m^* = .13 \text{ cm}$ and $\mu = 125 \text{ cm}^2/\text{volt sec.}$ Using relations VII and XI and estimating internal transmittances of film and substrate to be 10% and 98% respectively by neglecting

multiple beam reflection, the comparison shows the carrier concentration of Ge could be about a factor of 60 down on the PbTe film. Putting this deduced value in relation X shows rotation in the substrate might be as high as half that in the PbTe. It therefore suggests that the value of N is of the right order but up to 30% in error.

The factor of roughly 100 between the carrier concentration of the Faraday sample and the Hall sample is not seriously in dispute but needs to be explained such as by a theory for the doping involved.

Effect of Cooling on Transparency

The same sample as used in Faraday measurements was cooled to 77°K and its transmission traced from 12 μ to 20 μ wavelength. The transmission fell at the lower temperature compared with the higher. The ratio of the transmissions was strongly wavelength dependent being greater at long wave.

This suggests that the carrier mobility is governed by an ionised-impurity scattering mechanism, this being the only effect known to cause increased free carrier absorption at low temperature. (Ref. 4.6 P.148).

Dispersion/Absorption Measurements

The single films discussed so far have been deposited under conditions considered best for PbTe in multilayers with ZnS. Thus substrate temperature was 260°C for the Faraday sample, other samples were evaporated at 300°C and this appeared to give more transparent multilayers on the Reading plant than 260°C. However, our present conclusion is that single films of high transparency can be deposited at much lower temperatures than is possible in a PbTe/ZnS multilayer. This effect needs further investigation.

A correlation has also been found between transparency and modifications to the evaporating source, particularly, transparency is improved on using an indirectly heated alumina crucible in place of a metal boat. The consistency of this effect, and also the possible effect of substrates or pre-coated structure, need to be checked and correlated with tendency to "peel".

Examples of a highly-absorbing film and a relatively transparent film are given in Figure 4.47, neither of which were heat treated. From the turning points it is possible to plot refractive index as a fraction of the index at the extrema nearest to $4\ \mu$. At short wavelengths the effect of free carriers can be neglected, and it has been found that values of refractive index reported for bulk materials are little different from those of films; thus one can assign an index at short waves and plot a dispersion curve. By comparing this with theoretical plots one can deduce carrier concentrations. Figure 4.48 (a) shows some theoretical dispersion curves for Lead Telluride. A curve is given for the index in the absence of carriers; this is based on results of Walton and Moss (4.21) on a sample of 10^{18} carriers/cm³ in the range $4.5\ \mu$ to $8\ \mu$. The results fit the form of a dispersion curve (Fig:2.1) for a simple classical oscillator representing the electronic edge dispersion, and using such a model extrapolation has been made to $3.5\ \mu$ and $15\ \mu$.

The remaining curves show the effect of various carrier concentrations using the approximate relation VI and taking values of m^* quoted for n-type material. The free carrier calculations show that in fact the effect of free carriers in a sample with 10^{18} carriers per cm³ cannot be neglected completely below $8\ \mu$, and the results of Walton and Moss require some slight adjustment to account for this.

Some preliminary curve fitting has been attempted using transmission spectra of unannealed samples. Dispersion data is approximate at the moment due to inaccurate wavelength calibration but the effect of carriers is quite clear and yields estimates of the concentrations.

Three samples to which this has been applied are as follows, with the Faraday sample added for comparison :-

Sample No	Substrate	Deposition Temp.	Source	Transparency $\lambda > 15\mu$	Estimated carrier concentration from dispersion (approx.)
071/4	Si	300°C	Alumina crucible	Good	7×10^{17}
*073/4	Si	300°C	Ta boat	Poor	3×10^{18}
*079/4	Ge	200°C	Alumina crucible	Good	3.6×10^{18}
054/3	Ge	260°C	Ta boat	Poor	4.2×10^{18} (Faraday)

* See Figure 4.47.

These values are in closer agreement with the Faraday measurement than is the Hall effect sample, evaporated at the same time as 073/4.

The concentrations do not correlate with the level of absorption in the samples, in fact the best transmitting film has the heaviest dispersion. This suggests that the mobility of the carriers differs from sample to sample in a stronger manner than the carrier concentration. This is born out by an anomaly in the dispersion of 073/4 at 22.7μ (Figure 4.48(a)).

A qualitative explanation can be found in considering the effect of the approximation made in relation VI on which Figure 4.48 is based. Two quantities neglected are κ and g . If g^2 is large and becomes comparable with ω^2 the effect is to reduce the increment in n^2 (relation IV). κ^2 was also neglected and this seems legitimate from the level of absorption but as it increases to longer wave it would have the effect of increasing n .

This anomaly is not present in the dispersion of the other two samples out to 23μ but would be expected at longer wave.

Sample 071/4 has been traced in transmission from 40μ to 80μ . It is apparent from this and the 4μ - 25μ measurement that a minimum lies in the

region $25\ \mu - 40\ \mu$ (this is to be expected when the film is one quarter-wave in optical thickness since the order can be allocated from the fringe count of 12 in the monitoring of the evaporation). However, a maximum also occurs at 180cm^{-1} ($55.5\ \mu$) with a transmission of 70% of that of a bare substrate, low κ and g . Here the phase thickness of the film is less than a quarter-wave thick. This can be interpreted as approximately the wavelength where n , the refractive index of the film, is equal to that of the substrate. Above this wavelength the index is less than that of the substrate and has changed from high reflecting to low reflecting. At this point the film is effectively one quarter-wave thick again, due to a phase change at the substrate/film interface of close to 90° , which can only occur for the small value of κ indicated when n is close to the index of the silicon substrate, namely about 3.42. This corroborates the value of carrier concentration deduced from the dispersion at shorter waves, and is indicative of the range of values of g occurring in the different films. In sample 073/4 this parameter is appreciable at wave-numbers of about $400\text{-}500\text{cm}^{-1}$; in sample 071/4 it is still not appreciable at $180\ \text{cm}^{-1}$, where the condition $\omega^2 \gg g^2$ is still apparently valid.

Reflection Measurements

Some PbTe films have been measured in various wavelength ranges in the region $24\ \mu$ to $100\ \mu$ in reflection to see if the plasma effect occurs indicating $n = 0$. In terms of the approximate relation IX it should occur in this region for all the films discussed in the last section, for 071/4 in the range $70\text{-}100\ \mu$, for 073/4 and 054/3 in the range $30\text{-}40\ \mu$.

In practice 073/4 and 054/3 showed rising reflectivity from 50 to $100\ \mu$ reaching about 70%.

Two further samples measured in this way were prepared as follows:-

Sample No.	Substrate	Deposition Temp.	Source
078/4	Ge	100°C	Alumina crucible
081/3	Ge	300°C	Alumina crucible

081/3 was prepared in a similar manner to 071/4 but had a much higher absorption ; it is the only sample of the four measured to show a plasma effect (Fig. 4.48(b)). (The effect normally seen in single crystal samples is of reflectivity falling to near zero when $n = 1$ and then rising steeply to near 100% when $n = 0$ (4.22 p.150)). As these films are on high index substrates the effect is obscured and one can only search for reflectivities approaching 100%. In a sample of low absorption when $n = 1$, the reflectivity will be approximately that of the substrate, though transmission measurements suggest this is unlikely to be a legitimate assumption in these films.

In 078/4 the reflectivity was rising slightly from 80-100 μ reaching only 50%.

From these results one can deduce a carrier concentration using relation IX as follows:-

$$081/3, n = 0 \text{ at } 150\text{cm}^{-1}, N = 1.1 \times 10^{18}/\text{cm}^3 \text{ approx.}$$

The absence of high reflectivity in other samples, where one might have expected it, probably indicates the invalidity of relation IX rather than the carrier concentration values are excessively high. For, considering relation VIII, high values of K or g have the effect of reducing the plasma frequency. For values of g such that

$$g^2 > \frac{Ne^2}{\epsilon_0 m^* (n_L^2 + \kappa^2)}$$

ω_p is imaginary.

The short wave dispersion result of 073/4; already discussed, suggest that in this film ω_p would be very small or imaginary, and in view of heavy absorption in 081/3 at 20 - 25 μ wavelength, it is doubtful whether the approximate relation used to deduce N in this case is valid. Rewriting relation VIII thus:

$$N = \frac{\epsilon_0 m^*}{e^2} (n_L^2 + \kappa^2) (\omega_p^2 + g^2) \quad \text{VIII}$$

shows that since κ and g were neglected in deducing N, the value quoted is a lower limit.

Clearly this technique is limited in usefulness to samples with high-mobility carriers. The dispersion result of 071/4 indicates that this might be such a sample but this has not yet been measured in reflectivity.

Comparison with filter measurements

Measurements on most of the single films indicate a carrier concentration in an evaporated film of the order of 10^{18} per cm^3 . Reference to Figure 4.48(a) shows that the maximum change of index to be expected from a reduction in this concentration is from 5.48 to 5.64 at 15 μ , a change of 3.5%. This can be compared with the actual shift of Fabry-Perot peaks upon heat treatment. Figure 4.43 shows the shift of the peak is linear with index change in the lead telluride and for the design considered 1% change in n gave -0.735% shift in wavenumber.

Thus at 15 μ one would expect a maximum shift of about -2.57% in wavenumber in this particular 10 layer design.

The shift measured in 3 Fabry-Perots (Figures 4.41) of the same design are indeed of this order of magnitude; of course the carrier concentrations in the multilayer films may have been larger than 10^{18} per cm^3 before annealing, but it does seem possible from this observation that annealing may reduce carrier concentrations significantly below 2.5×10^{17} per cm^3 .

4.4.3.3 Conclusions

Interpretation of these measurements is incomplete. Transmission measurements, for instance, can be used to yield values of absorption coefficients and hence estimates of damping constants, or carrier mobilities at infra-red frequencies to provide further insight into differences in deposition conditions. Various of the measurements can be used to deduce the relative effects of heat treatment on carrier concentrations and damping constants

The following tentative conclusions can be drawn however:-

- 1) Evaporation appears to alter the doping of the PbTe from p-type to n-type, and under a variety of conditions there is an overall reduction in the number of carriers as a consequence of evaporation. Differences in evaporation conditions cause small differences in carrier concentrations but appear to cause large differences in the strength of carrier scattering processes.
- 2) Heat treatment in air reduces free carrier absorption and causes spectral shifts of interference fringes. A reduction in the number of carriers is required to explain these facts but the effect on scattering mechanisms is not yet known.

The significance of oxygen in the heat-treatment process is not yet understood, nor the doping changes during evaporation.

It is possible that some non-stoichiometry exists or is produced in the lead telluride. If free lead and tellurium are present in the evaporant stream substrate temperature may affect their relative sticking coefficient significantly. Thus an excess of tellurium in the source material (granules) may be altered to an excess of lead in the film, doping it n-type. Oxygen might then provide p-type doping and recombination of carriers to reduce the concentration. Different types of evaporation source may have differing temperature distributions with effect on dissociation of PbTe or relative evaporation rates of Pb and Te. Some boat materials may have a chemically reactive effect.

Different types of substrate crystals or previously-deposited layers causing different stresses in lead telluride films could yield differences in dislocation or grain size between films. Since these can assist in internal precipitation of any excess atoms (Refs. 4.12 and 4.13) free carrier properties may be affected.

Subject to the outcome of further investigation of these effects there would seem to be a genuine possibility of improved control of carrier concentration and/or mobility during deposition, with subsequent controlled adjustment by heat treatment.

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4.5 METALLIC MESH FILTERS FOR 50 μ AND LONGER WAVELENGTHS

4.5.1 Introduction

Homogeneous metal layers have been employed as reflecting interfaces for interference filters in the visible and ultra-violet regions. Throughout the infra-red the absorption of such layers is excessively high. The same is not true of metallic meshes. These have been investigated for the microwave region (4.23, 4.24) and also at submillimeter wavelengths (4.25).

Mesh elements are new to the region of this investigation. They pose some very special problems; those of section 3.3 are in many ways complementary. This is because establishment of the techniques involving reflecting meshes as components for infra-red interference filters, essentially demands solution of the problem of spacing and parallelism. Such studies were described for bulk layers; here we discuss the problem for meshes.

It is shown that the limitations of dielectric multilayers, which are basically those of a lack of adequately reflective interfaces, can be overcome.

4.5.2 Theoretical Considerations

4.5.2.1 Metal gratings

Metallic gratings have been discussed theoretically by a number of workers, for the cases of both wires (4.26) and flat strips (4.27). Gratings can be characterised by the periodicity (g) and breadth of metal elements ($2a$). Surface currents in the grating produce a scattered field. Wavelengths $\lambda < g$ give rise to diffraction effects. For $\lambda > g$, the only propagating parts of the scattered field are the zero order transmitted and reflected waves. The gratings are almost transparent for radiation polarised with the electric vector perpendicular to the elements. Parallel polarised radiation has a wavelength dependent reflectivity, which is large at long wavelengths. This property is used for the interference filters.

Approximate equations describing the reflectivity R , transmissivity T , and absorptivity A of strip gratings can be written down for $\lambda \gg g$ and $\lambda \gg a$.

as follows:-

$$\begin{aligned} R &= 1 - \left(\frac{2g}{\lambda} \log_e \sin \frac{\pi a}{g} \right)^2 \\ T &= \left(\frac{2g}{\lambda} \log_e \sin \frac{\pi a}{g} \right)^2 \\ A &= \left(\frac{g}{2a} \frac{c}{\sigma \lambda} \right)^{\frac{1}{2}} R, \quad (R \gg A) \end{aligned}$$

where σ is the strip conductivity, which accounts for ohmic losses.

The following deductions from the theory are of importance:-

1. A scaling of the optical properties of gratings with different grid constants, may be carried out through the normalised frequency g/λ
2. The a/g ratio affects the 'steepness' of the wavelength dependence; if a/g is large, R is steep.
3. Thickness of the element has a marked effect on the spectral response; for the same a/g ratio, strip gratings give a flatter response than do wire structures.

Considerable variation in the spectral behaviour of gratings is therefore possible.

4.5.2.2 Metal meshes

The two-dimensional structure of a mesh or grid exhibits the same behaviour for unpolarised radiation, as does the grating for the parallel polarised situation (4.24). Calculations have been carried out for circular apertures in a rectangular array (4.28) yielding qualitative agreement with the observed dependence of square openings (4.29). However, the formulae quoted above, allow a simpler expression of the behaviour.

4.5.3 Practical Situation

4.5.3.1 The problem

Suitable meshes for the region 50-100 μ have grid constants between 16 and 50 μ (1500-500 lines/inch). Copper meshes with 1000 L.P.I. were used

mostly in the experiments: the transmission of one of these is shown in Fig.4.49. They are manufactured by a photo-etching technique (Dainippon Screen Manufacturing Co.Ltd.), and are very fragile; fine tissue paper has about the same strength, but is more rigid. Their intrinsic weakness produces severe mechanical problems.

Meshes of 1000 L. P. I. have sufficient reflectivity for 1% half-width F. P. filters at 100 μ (Fig.4.50). This means that the allowable variation in mesh spacing is about one visible light fringe.

The practical problem therefore is twofold:-

- a) Meshes must be spaced with a high degree of accuracy and parallelism.
- b) Adequate mesh flatness must be achieved.

4.5.3.2 Early Fabry-Perot filters

Flatness of etched meshes can be achieved by stretching them over accurately machined and lapped support rings. The mesh is glued onto the outer ring; flexible collodion is good for this. Stretching is obtained by careful adjustment of the screws against the inner support ring.

A simple filter is assembled by clamping a spacer between two such formers. Fig.4.51 shows the transmission of an example employing a melinex film spacer. The peaks are third $-(185 \text{ cm}^{-1})$ and fourth-order; they are broad and of low transmission for these wavelengths. This is ascribed to the following causes:-

1. Absorption in the melinex reduces the peak intensity.
2. There is a lack of parallelism.
3. The flatness due to the formers is degraded by contact of the meshes with the spacer.

The simple expedient of cutting an area out of the centre of the plastic and reassembling, produced the improvement exhibited in Fig.4.52. The

peaks are now second and third order. A calculated upper limit for mesh absorption is 3% at 59 μ ; no correction was made for 'wedging'.

4.5.3.3 Spacers

There are two possible alternatives for solving the problem of spacing, which are discussed separately.

- a) The first is to find spacers which are sufficiently uniform over the optical area. Various plastic films were tried, but performance was always greatly inferior to that of Fig. 4.52. Polyethylene spacers absorb less than do melinex samples but are much less flat. In addition, the aim of 'pitching' on a selected wavelength was found impossible to achieve. The film thickness is unpredictable, and inclusion of dust particles over the optical area produces serious mesh distortion - this was often visible. Attempts were made to cast polystyrene spacers onto optical flats, and also onto grids stretched over optical flats - meshes supported in spacers could then be stacked. Quantities of standard solution were measured out with a calibrated syringe. This also failed. Again the problem is dust, and also stripping the film from the flat without distortion.
- b) The second approach of an annular spacer eliminates the spacer absorption problem. However, it results in a less robust structure. Ring spacers cut from plastic were found to be distorted in the process. Cleaved mica spacers were somewhat more predictable, but the basic problem was again distortion when cutting out the centre.

Production of precision filter spacers was successfully accomplished using metal shim. Distortionless rings are obtained by etching out the shape required. The simplest method is to wax completely a piece of shim. Wax

is removed with dividers from diametrically opposite sides of the metal. Acid attacks the metal along the concentric scribed lines. The ring is washed and the wax removed with a solvent, in which the spacer remains until it is required.

Thickness can be controlled by a number of methods, which follow:-

- 1) Selection over the shim area produces the best spacers.
- 2) Lapping can be carried out for steel. (Soft metals take up the abrasive and increase in thickness in a random manner).
- 3) The thickness can be increased by evaporation.

Measurements with an accurate micrometer dial gauge was adequate for this work ($\pm 1 \mu$)

4.5.3.4 A Double half-wave

Two identical spacers were used with two formers and three meshes to construct a double-half-wave type filter (Fig. 4.53). The failure to achieve the characteristic square band shape is assigned to drastically unequal half-waves. 'Kinking' of spacers intruding within the inner rings of the formers is expected to cause this. The low overall transmission is consistent also with the resulting non-parallelism.

The lesson is to confine the spacers within the contact area of the inner rings.

4.5.4 Improved Fabrication Technique*

4.5.4.1 Description

A schematic diagram of the assembly process is shown in Fig. 4.54. The frame rings are of 0.05 inch thick steel, the internal diameter is 0.8 inches and the outer diameter is 1.05 inches. One face of each is lapped accurately flat. They are kept under liquid until needed. Spacers are of

* patent applied for.

the same diameters, and their manufacture has already been described. Meshes are stretched flat on formers as before, but are then glued to the inner rings. These then provide weights to further stretch the meshes over a frame ring and spacers successively, as the structure is progressively assembled. Each ring must be cleaned and accurately positioned. The final assembly is firmly clamped; separations are then those of the metal spacers and dust has no influence. Finally, excess mesh is trimmed off, and the two frame rings glued together on the outside edges.

All attempts to 'pot' the assembly in plastic solution, in order to protect the meshes, destroyed the flatness. It would be interesting to try a plastic vapour deposition method in this context. (For example, Union Carbide-parylene). Protection can be afforded by the use of end substrates; including the back-up filters for removal of higher-order spectra, and transmission from the mesh diffraction region. These are generally necessary for application of band-pass filters.

In the case of F. P. filters there is the further possibility of stretching the meshes over substrates. They are then glued into place with a thin polystyrene layer. Spacing can be carried out in the usual manner. This method is only worthwhile for meshes with reflectivities designed for broad band filters ($\sim 10\%$) if dust is again troublesome. It also requires flat substrates.

4.5.4.2 Optical properties - Fabry-Perots

The following discussion parallels the previous Section. A number of the mesh F. P. type filters, made using the improved technique, are listed in Table 4.8. Half-widths and transmissions were measured for the image sizes and related resolutions indicated in this table. They are not corrected for instrumental broadening.

The transmission of G.4 was measured as a function of slit width. An extrapolation of half-width and peak transmission to zero slits is shown in Fig. 4.55. Remaining contributions are as follows:-

1. Cone angle broadening has a similar effect to resolution limiting. For the cone semi-angle (10°) employed, $\delta\lambda / \lambda = 0.015$. There is also a small peak position shift (~ 0.007) to longer wavelengths because of the consequent increase in effective spacer thickness.
2. Some degree of non-parallelism may still be effective in fig. 8.9. Although the extrapolation was to zero slit-width, the slit height contribution to the image size remains. A three position scan across filter G.2 is shown in Fig. 4.56. No overlap between the three image positions was permitted, and the separation between centres of the adjacent areas was 5mm. The peak shift is about $\frac{1}{2} \text{ cm}^{-1}$ between each: an image of 5mm corresponds to 7mm slits, and broadens the filter by about $\frac{1}{2} \text{ cm}^{-1}$ then. This is the main imperfection in these filters; no doubt improvements in spacer uniformity are technically feasible.
3. The remaining half-width contribution is that due to the finite mesh reflectivity. This half-width has no meaningful existence here, since it demands a parallel beam of vanishingly small cross-section. If the environmental geometry is specified, the achievable finesse can be determined from a knowledge of flatness and parallelism tolerances. The necessary mesh reflectivity is then determined for optimum filter peak transmission. Alternatively, if the half-width is set, the environmental geometry must be appropriately limited.

The transmission of three filters of Table 4.8 are shown in Fig. 4.57; the effect of the increasing A/T ratio with wavelength is apparent. Without making corrections to these spectra, an upper limit can be determined for A from $\tau_{\text{max}} = \left(\frac{1}{1 + A/T} \right)^2$. The approximation becomes worse for longer wavelengths since the 'intrinsic' half-widths are rapidly narrowing. (Finesse $\propto \lambda^2$). However an error of 10% in τ_{max} yields 5% in A, and T is not known (from Fig. 4.49) to better than 10% at 90μ . Performance of filters is summarised as follows:-

Filter	G7	G5	G6
Peak	84 μ	88 μ	93 μ
max	0.5	0.46	0.375
T	0.04	0.03	0.02
A/T	0.41	0.47	0.63
A (upper limit)	0.016	0.014	0.013

These compare with the upper limit of 3% at 59 μ (Sub-Section 4.5.3.2)

4.5.5 Multiple mesh Structures

Extension of the technique to multi-interface structures is relatively straightforward, using the method of the last section and work due to R. E. V. Chaddock is described below.

4.5.5.1 Mesh double-half-wave filters

Fig. 4.58 shows the transmission of a filter consisting of three 1000 L. P. I. meshes, with the 42 μ spacers. The deep central minimum is expected because of the large discrepancy between the effective reflectivities (R_1, R_2) either side of one half-wave (see section 4). Transmission at the minimum is given by:

$$\tau_o = \frac{T_1 T_2}{(1 - (R_1 R_2)^{\frac{1}{2}})^2}$$

where $T = 1 - R$, and absorption is neglected. For two meshes, $\tau_{\max} \sim 0.97$, and so R_1 at the centre of the D. H. W. pass band is 0.03. $R_2 \approx 1.0$ and so $\tau_o \sim 0.03$. The characteristic squarer shape of the pass-band is however demonstrated.

A system 500/1000/500 pitched for 90 μ is calculated to have a shallow central minimum of $\tau_o \sim 0.8$. The lower reflectivities of the 500 L. P. I. meshes mean that the spectrum is broader. (A. F. P. with two 500 L. P. I. meshes has a half-width of about 15% ; see Fig. 4.50 for ($\lambda / g = 1.8$). The

thin etched meshes ideal for these filters were not forthcoming in gauges other than 1000 L. P. I. at the time of these investigations. Thicker electrodeposited examples were employed as a substitute (E. M. I. Ltd., Hayes Middlesex). The resulting transmission is shown in Fig. 4.59. A shift to 115μ is believed to be a consequence of the thick meshes. No significant shift has been observed with the thin meshes. The possible spacing errors cannot account for the magnitude observed here, and the effect needs further investigation. Absorption may also be expected to have a greater influence for thick meshes. Higher reflectivities at this longer wavelength narrow the filter; however, a D. H. W. is intrinsically broader than a F. P. The observed half-width of 15% is therefore reasonable; 85% of the integrated transmitted energy is concentrated within this region.

4.5.6 Extension of the technique

In principle, the method of fabrication can be extended to any number of interfaces. Meshes replace the quarter-wave stacks of the dielectric multilayer. The essential differences between the approaches are as follows:-

- a) One mesh can provide any required reflectivity, in principle. This obviates the necessity of adjusting the number of layers or the index contrast, both of which have been shown to have very limited scope for variation. The grid constant g must be varied. Commercially, only certain values are obtainable at present. General application of the technique would appear to demand establishing a facility for production of a larger range of mesh constants. This is perfectly feasible on an industrial scale.
- b) The absorption in thin meshes is very much less than in the corresponding multilayer stacks - were these possible. This is certainly true beyond 50μ .

4.5.7 Summary

Metallic meshes have been employed to construct multi-interface interference filters. The limited region of dielectric multilayers has therefore been extended.

The results are summarised.

- 1) It has been shown that very good filter performance can be achieved using annular precision shim spacers. The spacing uniformity has been investigated, and this is certainly adequate for 5% half-width F. P. filters. In particular, the 85 μ and 95 μ filters of the test problem have been produced well within the specifications.
- 2) The measured single mesh transmission is sufficient to carry out simple design and predict performance.
- 3) Double-half-wave designs of squareband characters have been constructed.
- 4) Mesh absorption is low ($A < 1\frac{1}{2}\%$ for 1000 L. P. I. meshes beyond 80 μ). This is certainly true for the thin photo-etched meshes.
- 5) There is some indication of a thickness phase shift, which is suggested by the result involving thick meshes.

The techniques which have been developed can with advantage be extended to sub-millimeter wavelengths, where the mechanical tolerances are relaxed, and filtering techniques are still relatively primitive.

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5. MULTILAYER THEORY

5.1 Extraction of optimum design parameters

5.1.1 Introduction

The multilayer designer's present practice in attempting to meet new requirements is mostly to adapt whatever library of well-proven designs may be available to him by computation of the "successive approximation" sort. Not only is this a bad current practice, the demands of future measuring systems are certain to need a more general understanding of the theory of multilayer spectral response than at present obtains. Stages in establishing a satisfactory design would then be able to take the more natural sequence :-

System specification → "best-fit" spectral response for "feasible" multilayer → extraction of detailed data → refinement computation, re-design etc.

As stated, however, an immediate conflict is visible in the sequence between the requirements of best-fitting (which implies numerical processes) and the extraction of parameters, which should be repetitive and functional (i. e. analytic.) This conflict is basic in multilayer design and arises from the limitations on refractive index which leads to an inevitable loss of exactness at some stage. This is not to say that the overwhelming advantages of a 'synthesis' procedure such as that outlined above need be completely lost, however, and a suggested method which has been found eminently suitable for the present problem is described below.

The method, which is borrowed from the part of electric circuit theory known as design synthesis, assumes that best-fit response is, spectrally, an exact function. Necessarily, this representation is restrictive and it is fortunate the Tchebyshev / equal-ripple functions can be used since they are themselves "best fit" in the sense of the idealised Fig. 5.1. With reference to that figure the Tchebyshev functions (which are polynomial) are already optimum and are of improved spectral squareness in comparison with most numerically-extracted design data. Their use in the design of electrical

filters is a standard process to which reference may be made^(5.1); a brief summary is given in the next section.

Further advantages of the use of exact functions in filter design are summarised as follows :-

- (a) Rejection power and edge steepness can be increased without limit by increasing the number of filtering elements (i.e. layers in a multilayer).
- (b) The parameters which characterise the behaviour in the passband (ripple, peak transmittance etc.) are affected by the external conditions and can therefore be adjusted by admittance matching at the filter interfaces with any usual anti-reflection procedure.
- (c) Design parameters are described by formulae.

5.1.2 Tchebyshev response and parameters

Suppose the Tchebyshev polynomial function $T_m(\omega)^*$ is to determine a frequency response for transmissivity according to :-

$$T = |t(\omega)|^2 = 1/(1 + h^2 T_m^2(\omega)) \quad (5.1)$$

Reflectivity is therefore:-

$$|p(\omega)|^2 = 1 - |t(\omega)|^2 = h^2 T_m^2(\omega) / (1 + h^2 T_m^2(\omega)) \quad (5.2)$$

Whereas the reflectivity is a function of ω (i.e. a real variable), it is permissible to consider the amplitude coefficient as function of the complex variable $\underline{s} = \sigma + j\omega$. It is therefore possible to describe $p(\underline{s})$ by locations of poles and zeros in the $\underline{s}$ -plane, thus :-

$$p(\underline{s}) = \frac{\prod_{m=1}^m (\underline{s} - \underline{s}_{\text{zero}(q)})}{\prod_{m=1}^m (\underline{s} - \underline{s}_{\text{pole}(q)})} \quad (5.3)$$

* $T_m(\omega)$ is $\cos(m \cos^{-1} \omega)$ in the pass region and $\cosh(m \cosh^{-1} \omega)$ in the stop region; in polynomial form, $T_3(\omega) = 4\omega^3 - 3$; $T_4(\omega) = 8\omega^4 - 8\omega^2 + 1$ etc.

The location of the poles for the Tchebyshev function, the periphery of a semi-ellipse in the $\underline{s}$ -plane is shown in Fig. 5.3, and suffices to specify $\rho(\underline{s})$ completely. Next, an admittance function (Fig. 5.2) is formed for the filter using the relationship $Y_{in} = (1 + \rho(\underline{s})) / (1 - \rho(\underline{s}))$. Finally the expansion of Y_{in} into a repeating partial fraction allows parameters for a Tchebyshev-design circuit to be extracted one by one. Because of the simple geometry of Fig. 5.3, formulae exist for these parameters, shown in Fig. 5.4. The formulae represent "prototype" data for electrical, and all other, similar filters and extensive tabulations of them are available (5.2).

5.1.3 Application to multilayer design

The extraction of Tchebyshev-design parameters for a generalised multilayer cannot follow the method of Section 5.1.2 directly. Reasons for this are the more complicated variation of multilayer admittance with frequency (wavenumber) and the need to impose constraints corresponding to "practical" refractive indices: the simplest alternative procedure is to establish a detailed analogy between the circuit of Fig. 5.4 and the multilayer by manipulation of matrices. A physical identity between layers and individual circuit elements is then possible and the formulae of Fig. 5.4 can straightway be adapted to the multilayer situation.

Matrix manipulations (illustrated in the following Section for a low-pass design) lead to combined equations which appear to require iterative solution to obtain the layer parameters.* By simple approximations the complexity of the equations can be reduced; in the low pass case, for example, thickness becomes identified by a single equation for each layer. A complete example of the extraction of low and high pass design data by this approximation is given in the Section 5.1.4 for PbTe/ZnS multilayers.

* In extracting low pass parameters, layer thickness or layer index can (in theory) be considered as alternative choices for variable parameter; in extracting band pass parameters the (equivalent) index of "stacked" quarterwave layers is considered variable parameter.

Discussion of iterative procedures for obtaining data of improved accuracy, numerical refinement by use of merit functions, curve-fitting, are given later. Computed performance of the approximate designs is compared with the Tchebyshev function in Figs. 5.5; computed performance for (narrow) band pass designs * (of assumed construction :- STACK (1) : SPACER (1) : STACK (2) : SPACER (2) etc.) is also given in Fig. 5.6.

The Tchebyshev filters are important because :

- (a) Their spectral response is optimum and corresponding use of layer material is most efficient;
- (b) A reduced sensitivity to thickness errors in deposition is to be expected by analogy with Tchebyshev electrical filters which are known to be of reduced sensitivity to errors in assembly, and for which sensitivity is calculable in terms of a perturbation of the known s -plane pole/zero locations. A fully-fledged practical investigation of the implications of advantage (b) is justified and suggests the need for further examination of monitoring accuracy, deposition of fractional layers, diagnosis of cumulation of thickness errors followed by self-correction etc., to see whether an overall improvement in the deposition of edge filters can be obtained.

5.1.4 An example of the manipulation of matrices

Steps in this process are twofold:-[†]

- (1) Choice of conditions for the circuit/multilayer analogy, with regard to choice of frequency variables, etc;
- (2) Manipulations to make individual matrices identical and avoid the evaluation of the overall multilayer matrix.

* J.C. Williams, Ph.D. Thesis (University of London) 1965 and, in press, with J.S. Seeley for Proc. I.E.E.

† J.S. Seeley, Electronics Letters 1 (1965) p.265

Low pass procedure is given in detail as follows :-

Manipulations take place at the cut-off frequency, ω_c in Fig. (5.1), and variable quantities are normalised to ω_c . Admittance levels are also normalised, to the terminating admittance, so that layer refractive indices are taken as $\frac{n_H}{n_S}$ and $\frac{n_L}{n_S}$. The matrix for the circuit then becomes a product of form:-

$$\begin{bmatrix} 1 & 0 \\ g_p & 1 \end{bmatrix} \begin{bmatrix} 1 & g_q \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ g_r & 1 \end{bmatrix} \quad (5.4)$$

for the sequence of elements p, q, r (taking g parameters from Fig. (5.4)) and the corresponding multilayer matrix becomes :-

$$\begin{bmatrix} \cos\theta_p & j\frac{n_S}{n_H} \sin\theta_p \\ j\frac{n_H}{n_S} \sin\theta_p & \cos\theta_p \end{bmatrix} \begin{bmatrix} \cos\theta_q & j\frac{n_S}{n_L} \sin\theta_q \\ j\frac{n_L}{n_S} \sin\theta_q & \cos\theta_q \end{bmatrix} \begin{bmatrix} \cos\theta_r & j\frac{n_S}{n_H} \sin\theta_r \\ j\frac{n_H}{n_S} \sin\theta_r & \cos\theta_r \end{bmatrix} \quad (5.5)$$

where θ_p etc. is the phase-thickness of layer p at frequency ω_c and is regarded as the variable parameter in the extraction of data. By an exact expansion, the layer matrices are written as triple products of similar form to equ. (5.4); for HIGH layers this is :-

$$\begin{bmatrix} \cos\theta_p & j\frac{n_S}{n_H} \sin\theta_p \\ j\frac{n_H}{n_S} \sin\theta_p & \cos\theta_p \end{bmatrix} = \begin{bmatrix} 1 & j\frac{n_S}{n_H} \tan \frac{1}{2}\theta_p \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ j\frac{n_H}{n_S} \sin\theta_p & 1 \end{bmatrix} \begin{bmatrix} 1 & j\frac{n_S}{n_H} \tan \frac{1}{2}\theta_p \\ 0 & 1 \end{bmatrix} \quad (5.6)$$

and for LOW layers is :-

$$\begin{bmatrix} \cos \theta_q & j \frac{n_S}{n_L} \sin \theta_q \\ j \frac{n_L}{n_S} \sin \theta_q & \cos \theta_q \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ j \frac{n_L}{n_S} \tan \frac{1}{2} \theta_q & 1 \end{bmatrix} \begin{bmatrix} 1 & j \frac{n_S}{n_L} \sin \theta_q \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ j \frac{n_L}{n_S} \tan \frac{1}{2} \theta_q & 1 \end{bmatrix} \quad (5.7)$$

By manipulations, as described in reference † on p. 5.4, the sequence composed of an alternation of the form of equs. (5.6) and (5.7) is reduced to the original number and form of the sequence of equ. (5.4). For the qth matrix (assumed to correspond to a LOW layer) identification requires that :-

$$g_q = N_{qp} N_{qr} \frac{n_S}{n_L} \sin \theta_q + \frac{N_{qp}}{N_{rq}} \frac{n_S}{n_H} \tan \frac{1}{2} \theta_p + \frac{N_{qr}}{N_{pq}} \frac{n_S}{n_H} \tan \frac{1}{2} \theta_r \text{ etc..}$$

$$\text{where } \begin{cases} N_{qp} = N_{qr} \\ N_{pq} N_{qp} = 1 - \frac{n_L}{n_H} \tan \frac{1}{2} \theta_p \cdot \tan \frac{1}{2} \theta_q \\ N_{qr} N_{rq} = 1 - \frac{n_L}{n_H} \tan \frac{1}{2} \theta_q \cdot \tan \frac{1}{2} \theta_r \end{cases} \quad (5.8)$$

Similarly constructed equations are obtained for HIGH layers. Equations (5.8) are final and, not being amenable to further simplification, appear to require an iteration. An approximate solution is suggested, however, by a modification expressed as :-

$$N_{qp} = N_{qr} = N_{rq} = N_{pq} \approx \sqrt{1 - \frac{n_L}{n_H} \tan^2 (\frac{1}{2} \theta_q)}$$

$$\tan \frac{1}{2} \theta_p + \tan \frac{1}{2} \theta_r \approx 2 \tan \frac{1}{2} \theta_q \quad (5.9)$$

for the qth layer so that

$$g_p \approx \frac{n_H}{n_S} \left(1 - \frac{n_L}{n_H} \tan^2 \left(\frac{1}{2} \theta_p \right) \right) \sin \theta_p + \frac{2n_L}{n_S} \tan \frac{1}{2} \theta_p$$

$$g_q \approx \frac{n_S}{n_L} \left(1 - \frac{n_L}{n_H} \tan^2 \left(\frac{1}{2} \theta_q \right) \right) \sin \theta_q + \frac{2n_S}{n_H} \tan \frac{1}{2} \theta_q$$

etc.,

which, without further approximation, simplifies to

$$\begin{aligned} \text{for HIGH layers} \quad \sin \theta_p &\approx \frac{g_p}{\left(\frac{n_H}{n_S} + \frac{n_L}{n_S} \right)} \\ \text{for LOW layers} \quad \sin \theta_q &\approx \frac{g_q}{\left(\frac{n_S}{n_L} + \frac{n_S}{n_H} \right)} \\ \text{for the first layer (if LOW)} \quad \sin \theta_1 &\approx \frac{g_1}{\left(\frac{n_S}{n_L} + \frac{n_S}{n_H} \right)} \end{aligned} \tag{5.10}$$

θ is taken between 0 and $\frac{1}{2} \pi$ for low pass data and between $\frac{1}{2} \pi$ and π for high pass data. Equations (5.10) are extremely simple and can serve, at worst, as a starting point for the iteration of equations (5.8).

For an application of the method it remains to select a best value for the ripple parameter, h , when n_H , n_L , and n_S are initially chosen. The retention of the full rejection bandwidth is a prime consideration for reasons already given. Adopting this as a mandatory requirement ensures that central layers shall be of equal thickness. A further requirement then follows to fix h on a basis of uniform distribution of standing waves throughout the structure (5.3, 5.4).

$$h = \frac{\left| \frac{n_H}{n_S} - \frac{n_S}{n_L} \right|}{\left(\frac{n_H}{n_S} + \frac{n_S}{n_L} \right)} \quad (5.11)$$

Mandatory requirements also arise that $n_H > n_S > n_L$ and the odd layers, numbered from the termination, shall be of greater 'g' factor than the even. When $n_S < \sqrt{n_H n_L}$ the first layer at the substrate must therefore be LOW and when $n_S > \sqrt{n_H n_L}$ the first layer must conversely be HIGH. At the front interface, the refractive index n_S is "seen" for all frequencies less than ω_c by virtue of the admittance-transfer property inherent in all Tchebyshev-type structures. It is therefore n_S which requires anti-reflection and the addition of any subsidiary layers for that purpose can be regarded as a separate problem (if, for example, a LOW layer of suitably-chosen thickness is used, the filter must itself finish with a HIGH layer and contain an even number of layers if it started LOW).

Details of the entire procedure will now be given for the PbTe/ZnS combination on Ge substrate :-

Indices, assumed, are 5.1/2.2 on 4.0

$$\therefore h = \left| \frac{5.1}{4.0} - \frac{4.0}{2.2} \right| / \left(\frac{5.1}{4.0} + \frac{4.0}{2.2} \right) = 0.178. \quad \frac{n_S}{n_L} = 1.82, \quad \frac{n_H}{n_S} = 1.27$$

so the filter starts LOW: the LOW index is sufficiently close to $\sqrt{n_S}$ for use as anti-reflection so the filter should contain an even number of layers.

Rejection in excess of 10^4 is needed; a total of 14 layers will therefore be satisfactory since $\left(\frac{5.1}{2.2} \right)^{14} > 10^5$.

Parameter $\sinh \left(\frac{1}{m} \sinh^{-1} \frac{1}{h} \right) = .175$ for this n and h.

Extraction of parameters For 14 layers :-

q	1	2	3	4	5	6	7	8	9	10	11	12	13	14
$\sin \frac{q}{28}$	.111		.331		.531		.707		.848		.943		.984	
$\sin \frac{q}{14}$		.223		.435		.624		.783		.902		.975		1.0

and therefore

$$g_1 = \frac{2 \times .111}{.175}$$

$$g_1 g_2 = \frac{4 \times .111 \times .331}{(.175^2 + .223^2)}$$

$$g_2 g_3 = \frac{4 \times .331 \times .531}{(.175^2 + .435^2)} \quad \text{etc.}$$

Complete parameters are summarised in Table 5.1.

Except for slight rounding-off, the design parameters show a symmetrical (and typical) tapering of thickness; a similar result can be expected for other circumstances such as an odd total of layers, changes of indices, etc.

Computed performance, shown in Fig. (5.5), lies within the spectral limits set by the Tchebyshev function. Divergence from a perfect fit is not severe and can be ascribed to rounding-off in the design data, approximations in the derivation of equations (5.10) etc.. The numerical significance is negligible and it is reasonable to assume, in an actual deposition of such a design, that similar magnitudes of thickness errors would likewise be of negligible experimental significance.

An increased steepness of the remote edge of the stopping band may be discerned in Fig. (5.5), (viz: the one not dealt with in the design process, the high-passing edge for a low pass filter and the low-passing edge for a high pass filter, see Fig. (5.1)). The drawn inference (not formally proven) is that either type of behaviour can be obtained and the design data can be classified as -

- (a) of equal-ripple broadband pass (e.g. 0 to $\frac{1}{2} \pi$ etc.)
- or (b) of single-edge pass (situated above $\frac{1}{2} \pi$) of maximum steepness.

REFERENCES FOR SECTION 5.1

- 5.1 Van Valkenburg, Introduction to network synthesis (Wiley 1964)
- 5.2 Weinberg L. Network and analysis and synthesis (McGraw-Hill 1962)
- 5.3 Young L. J. Opt. Soc. America 52 (1962) p. 753
- 5.4 Levy R., Ph. D. Thesis (1966) University of London

Un-numbered references to J.S. Seeley and J.C. Williams are given in Sections 5.1.4 and 5.1.3.

5.2 COMPUTER PROGRAMS

5.2.1 Analysis

The following programs have been written, mostly in the ALGOL language :-

- (1) Transmittance (including the effects of absorption) as function of wavenumber.
INPUT: optical constants, n , k , for layers and incident/emergent media, wavenumber increment.
OUTPUT: plotted on-line (logarithmic or linear scale) using cubic interpolation.
- (2) Similar to (1), but excluding absorption, gives additional parameters such as complex reflection coefficient, refractive index and thickness of the equivalent Herpin bi-layer etc.
- (3) Two programs for refinement of design data to various criteria; one for refinement (and better fit) of initially-approximate Tchebyshev data, the other a general program using merit functions and able to proceed by generating its own initial data.
- (4) A tolerance program which calculates the sensitivity to thickness errors in selected layers, as a change in reflectivity, and variations in sensitivity with wavenumber.
- (5) A further tolerance program which assumes thickness errors in all layers, distributed randomly and calculated for a selected standard deviation.

5.2.2 Build-up computation (aid to monitoring)

An ALGOL program has been written which reproduces the reflectance as it is measured during a filter deposition (i.e. the monitoring signal). Reflectance as a function of incremental thickness (at a chosen wavenumber) through any number of layers is computed.

INPUT: refractive indices and thicknesses of layers, refractive indices of incident/substrate media, thickness increment(s).

OUTPUT: plotted on-line using cubic interpolation.

5.3 USE OF BUILD-UP, TOLERANCE, AND REFINEMENT COMPUTATION

5.3.1 Use of BUILD-UP computation

Data computed with the aid of the analytical programs are included at several places in this Report so that no further discussion of them need be given. The other programs, however, have thrown up information of intrinsic interest which will be discussed below.

The major contribution of the BUILD-UP program consists of predictions of the influence of monitoring errors and choice of monitoring wavelength. For the 10-layer production-type Fabry-Perot filter at 15 microns BUILD-UP computations were made on the assumption that layer thickness would be monitored in, or near to, 4th order at 3.75 micron wavelength. It was further assumed that deposition could be terminated at precise values of reflectance as well as at turning points, permitting the use of a monitoring (reciprocal) wavelength other than integral multiple of $(15 \text{ microns})^{-1}$: calculations were made at $\frac{6}{8}$, $\frac{7}{8}$, $\frac{8}{8}$, $\frac{9}{8}$, of λ_0 for an assumed observational error of 4% in reflectance at termination. Errors in actual thickness which result are as follows :-

Percentage errors in thickness for assumed 4% observational error in reflectivity

Layers	Monitoring wavelength (fraction of λ_0)			
	$\frac{6}{8}$	$\frac{7}{8}$	$\frac{8}{8}$	$\frac{9}{8}$
SUB				
L	4 (a minimum)	2	7 (a minimum)	2
H	11 (a maximum)	1	7 "	1
L	3 (a minimum)	4	7 "	3
H	18 (a maximum)	3	7 "	3
L	6 (a minimum)	5	7 "	5
HH	2 (a minimum)	4	3 "	5
L	21 (a maximum)	2	7 "	2
H	2 (a minimum)	1	7 "	0.5
L	12 (a maximum)	9	7 "	6
H	1 (minimum)	2	7 "	2

Therefore, if magnitude of observed reflectance errors is the sole criteria in choosing the monitoring wavelength, a non-integral multiple of $(\lambda_0)^{-1}$ may be advantageous and even a change of monitoring wavelength for crucial layers may be preferred.

BUILD-UP was used next to determine the effect of an observational error in a particular layer on the deposition of a succeeding layer where the monitoring proceeds without correction. Results are given for a bi-layer intended to be of equal thickness :-

	Monitoring wavelength(fraction of λ_0)		
	$\frac{6}{8}\lambda_0$	$\frac{7}{8}\lambda_0$	$\frac{8}{8}\lambda_0$
Error in first layer (in optical thickness)	-30° $+30^\circ$	-30° $+30^\circ$	-40° $+40^\circ$
Resulting error in second layer	$+10^\circ$ -10°	$+15^\circ$ -15°^*	-15°^* $+17^\circ^*$

* Magnitude at turning points affected.

The tendency for errors to accumulate in even-order monitoring is clearly seen above. On the assumption that these bi-layer results are typical for the second and third layers in a 6-layer Fabry-Perot filter (SUB: LHLHHLH), the following results were obtained from BUILD-UP:-

Monitoring at $\frac{3}{4}$ of λ_0

Layer	Phase thicknesses			
L	270°	270°	270°	270°
H	$270^\circ - 30^\circ$	$270^\circ - 30^\circ$	$270^\circ + 30^\circ$	$270^\circ + 30^\circ$
L	$270^\circ + 10^\circ$	$270^\circ + 10^\circ$	$270^\circ - 10^\circ$	$270^\circ - 10^\circ$
HH	540°	540°	540°	540°
L	270°	$270^\circ - 10^\circ$	270°	$270^\circ + 10^\circ$
H	270°	$270^\circ + 30^\circ$	270°	$270^\circ - 30^\circ$
Reflectance at end of monitoring	0.96	0.99	0.96	0.99
Shift of minimum, cumulative scale	$+10^\circ$	0	-10°	0
of phase-thickness		5-13		

Monitoring at $\frac{7}{8}$ of λ_0

Layer	Phase thicknesses			
L	315°	315°	315°	315°
H	$315^\circ - 30^\circ$	$315^\circ - 30^\circ$	$315^\circ + 30^\circ$	$315^\circ + 30^\circ$
L	$315^\circ + 15^\circ$	$315^\circ + 15^\circ$	$315^\circ - 15^\circ$	$315^\circ - 15^\circ$
HH	630°	630°	630°	630°
L	315°	$315^\circ - 15^\circ$	315°	$315^\circ + 15^\circ$
H	315°	$315^\circ + 30^\circ$	315°	$315^\circ - 30^\circ$
Reflectance at end	0.94	0.76	0.40	0.26

In these tables, as is common in Fabry-Perot filters, the possibility of thickness-error compensation by correction in symmetrically-placed layers on the far side of the spacer has been included.

An additional method of analysing error characteristics of filter designs is based on the method described by Baumeister (5.5). This tolerance program is most efficient when used to complement the build up method described above. A program was written to calculate the change of reflectivity produced by a percentage change in thickness of any layer in the filter over a spectral range and gives a direct indication of the spectral sensitivity of the filter to error, and also of the layers which are most sensitive to monitoring errors. The former quantity enables the designer to choose the most sensitive monitoring wavelength for the evaporation of each layer. The actual evaporation process can then be simulated using the 'build up' program.

Fig. 5.7 shows the results obtained for the ten-layer Fabry Perot Radio-meter filter using this program. As expected, the most sensitive wavelength for the space layer is at multiples of $\frac{\lambda_0}{4}$ ($-\frac{\lambda_0}{4}$ corresponds to 1.0 on the scale shown in the Figure), whereas for the other layers (i.e. those forming the reflecting stacks on either side of the spacer), the edge of the high reflecting region (about 0.63 on the wavenumber scale) appears most sensitive.

This work has not been pursued further or yet related to monitoring procedures used by plant operators.

5.3.2 Computer-aided refinement of design data

Several methods have been developed to refine given design data in order to produce a better approximation to a specified filter response (e.g. Baumeister^(5.6)). Other methods produce the design data automatically, i.e. no design data is given (Dobrowolski^(5.7), Heavens and Liddell^(5.8)), using the concept of a merit function. This is a function of the design variable parameters which may be minimised to produce an approximation to a desired curve. The program referred to in (3) of Sub-section 5.2.1 used the following form of merit function

$$F(\bar{x}) = \sum_{\sigma_K} f_K^2(\bar{x}) \quad (5.12)$$

$$f_K(\bar{x}) = f_K(x_1, x_2, \dots, x_n) = \left[R'(\sigma_K) - R(\sigma_K, \bar{x}) \right]$$

$x_1, x_2, \dots, x_n$ are the design variables (i.e. thicknesses of the layers in the filter), $R'(\sigma_K)$ is the specified value of reflectivity at wavenumber σ_K ; $R(\sigma_K, \bar{x})$ is the computed value of reflectivity at σ_K for a filter with layer thicknesses $x_1, x_2, \dots, x_n$. A value of the merit function F is obtained by summing $f_K^2(\bar{x})$ over a discrete number of wavenumbers σ_K . F may then be minimised with respect to $\bar{x}$, using a least-squares method, the minimum value of $\bar{x}$ providing the approximation required. The method may either be used to refine initial data (this would give the starting point for the search for a minimum) or it may be incorporated with a scanning procedure to produce a design completely automatically.

The method has been tried on the design of the low and high pass filters discussed in Sub-Section 5.1.4 by setting up an idealised 'squared-up' spectral response (same as in Fig. 5.1) at a number of discrete wavenumber values. Computed behaviour is compared in Figs. 5.8, 5.9, with a refinement of the data in Table 5.1 and with the locations of the chosen points. Designations for the computations were :-

Low pass and High Pass Phase Thicknesses

Layer	SUB	1	2	3	4	5	6	7	8
Index	4.0	2.2	5.1	2.2	5.1	2.2	5.1	2.2	5.1
Refinement of Table 5.1 (Low pass)		35.4°	51.6°	59.8°	60.1°	63.9°	63.1°	64.0°	64.2°
Automatic extraction		-	-	71.2°	47.5°	69.1°	57.4°	60.7°	79.5°
Refinement (High pass)		135.2°	130.9°	116.8°	109.2°	120.6°	113.9°	115.3°	114.9°
Automatic		-	-	130.0°	130.1°	128.9°	124.7°	113.9°	114.1°
		9	10	11	12	13	14	15	Air
		2.2	5.1	2.2	5.1	2.2	5.1	2.2	1.0
(Low pass contd.)		65.3°	62.0°	61.9°	58.9°	52.8°	36.1°	127.9°	
		50.8°	94.9°	55.1°	63.8°	58.9°	55.1°	162.7°	
(High pass contd.)		116.5°	117.6°	115.4°	131.6°	133.8°	137.2°	56.2°	
		125.5°	113.9°	118.5°	109.3°	119.8°	102.5°	54.5°	

The specified response (at points X in the Figures) was chosen to emphasise steepness of edge rather than overall low passing performance, but this could be altered by applying suitable weighting factors to compensate if desired. The initial design data (extraction described in Sub-Section 5.1.4) is changed very little by the refinement showing the data is very near an optimum and confirming the use of the Tchebyshev function and the method of extraction. In high pass the change produced by refinement is more marked and is probably due partly to the periodically repetitive nature of multilayer spectral behaviour (which renders it incapable of acting as a true high pass filter) and also to the scale chosen (the range $[0, 2\pi]$ in phase thickness corresponds to $[0 \text{ to } 2.5]$ in wavenumber in the low pass case, $[0.5 \text{ to } 1.5]$ in the high pass case).

An alternative specification may be provided by selecting the actual maxima and minima of the Tchebyshev curve as design points ^(5.9) instead of the 'squared-up' format. For an 11 layer filter of index contrast corresponding to a widely used visible-region multilayer combination this results in a slightly better fit of realisation to specification :-

Layer Index	SUB	1	2	3	4	5	6	7	8	9	10	11	Incidence
	1.52	2.30	1.38	2.30	1.38	2.30	1.38	2.30	1.38	2.30	1.38	2.30	1.52
*Initial thicknesses		38.3°	55.3°	63.4°	67.4°	68.4°	68.3°	68.4°	67.4°	63.4°	55.3°	38.3°	
Refined thicknesses		41.9°	52.8°	69.3°	64.5°	67.2°	68.9°	69.5°	65.5°	68.2°	49.4°	38.7°	

* extracted by method of Sub-Section 5.1.4

Results (discrete points only)

Wavenumber	0.144	0.270	0.424	0.556	0.691	0.800	0.900	0.972	1.00	1.056	1.11	1.21	1.36
Given reflectivity	0.025	0	0.025	0	0.025	0	0.025	0	0.025	0.15	0.5	0.9	0.975
Initial reflectivity	0.023	0.002	0.017	0.011	0.010	0.008	0.017	0.017	0.03	0.16	0.51	0.90	0.975
Refined reflectivity	0.025	0.004	0.027	0.010	0.017	0.006	0.017	0.015	0.026	0.15	0.50	0.90	0.975

Merit function $(F = \sum (\text{Given } R - \text{Calculated } R)^2)$:

Initially 1.028×10^{-3}

Refined 0.524×10^{-3}

REFERENCES FOR SECTION 5.3

- 5.5 Baumeister P., J. Opt. Soc. America 52 (1962) p.1149
- 5.6 Baumeister P., J. Opt. Soc. America 48 (1958) p.955
- 5.7 Dobrowolski J.A., App. Optics 4 (1965) p.937
- 5.8 Heavens O.S., and Liddell H.M., in press for Opt. Act.
- 5.9 Chen T.C., unpublished work for University of London Thesis.

5.4 RANDOM ERRORS IN NARROW BANDPASS FILTERS

5.4.1 Introduction

The problem of errors is basic to the production of any item which requires the utmost precision in its manufacture. This is particularly true for multilayer interference filters which depend for their action on the accurate positioning in space of many reflecting interfaces. During the manufacture of a multilayer filter by evaporation in a vacuum, the thickness of each layer is controlled by a photoelectric monitoring technique which is a process subject to errors in human judgement and also instrumental errors. Therefore, if it is required that the spectral transmittance of a multilayer filter conforms to rigid specifications, i. e. the transmittance should be within certain limits at specific wavelength, it is useful to know the sensitivity of the filter to errors in layer thickness.

There are several methods of examining the effect of errors on filter performance. For example, one can follow Baumeister^(5.10) and calculate the change in transmittance resulting from variations in each layer on the assumption that the other layers are unchanged. In this way, the sensitivity of the multilayer to changes in a given layer is derived. On the other hand, instead of isolating each layer in turn and examining the effect of changes in its thickness on the multilayer, one can study the effect of simultaneous changes in the thickness of all the layers. This latter approach is the one which has been adopted at Grubb Parsons. It corresponds to the situation in the manufacture of interference filters because the production process inevitably introduce errors into all the layers. The magnitude of these errors will depend to a large extent on the skill of the operator as well as the accuracy and response of the instrumentation.

In any system of instrumentation for the purpose of measurement or control there will be two main kinds of error which must be taken into account. There are what might be called "scale" errors which are known and constant and for which allowance may be made in calibration. The other kind are

variable and random and may be investigated only statistically. It is the latter random type of error with which we will be concerned below.

5.4.2 Calculational method

For the purposes of this study, the thickness x of each layer in the multilayer filter is assumed to be a random variable which is determined by the Gaussian distribution

$$y = \frac{1}{\sigma\sqrt{2\pi}} \exp - (x - \bar{x})^2 / 2\sigma^2 \quad (5.13)$$

where $\bar{x}$ is the mean (or datum) thickness of the layer, σ is the standard deviation (S.D.) of the distribution, and y is the probability or frequency of occurrence of the error $x - \bar{x}$. Since not all possible values of the error are equally probable (in fact it can be shown that 68% of the errors be within $\pm \sigma$), the error in each layer is determined by a normal (Gaussian) distribution of random numbers. This is done by means of the following method which allows the calculation of normally distributed random numbers with a given mean and standard deviation.

An approximation to a normally distributed random number Y can be found from a sequence of uniform random numbers ^(5.11) using the formula

$$Y = \frac{\sum_{L=1}^K X_i - \frac{K}{2}}{\sqrt{\frac{K}{12}}} \quad (5.14)$$

where the X_i are uniformly distributed random numbers such that

$$0 < X_i < 1$$

and K is the number of values of X_i to be used.

Y approaches a true normal distribution asymptotically as $K \rightarrow \infty$. In this case K was chosen as 12 to reduce execution time. Equation 3.2 thus becomes

$$Y = \sum_{i=1}^{12} X_i - 6.0 \quad (5.15)$$

The adjustment for the required mean and standard deviation is then

$$x = Y \cdot \sigma + \bar{x} \quad (5.16)$$

where x is the required normally distributed random number,

σ is the required standard deviation

$\bar{x}$ is the required mean.

The whole calculation of the effect of random errors on a multilayer has been programmed for the IBM1130 computer. The method outlined above has been coded in the form of standard Fortran Library sub-routines. The uniformly distributed random numbers X_i are found by the power residue method using a subroutine which is specific to the IBM1130 machine. The subroutine should not repeat its cycle in 2^{13} entries.

The normally distributed random numbers Y are then calculated from equation (5.15) and are used to compute the thickness of the layers according to equation (5.16). Each layer is therefore a normally distributed random variable with a mean thickness of $\bar{x}$ and a standard deviation of σ . These values of layer thickness are then used to calculate the multilayer performance according to the matrix methods of Abeles. The output is in the form of a graph produced by an on-line IBM 1627 plotter.

5.4.3 Results

The effects of random errors in film thickness have been examined with respect to their influence on the performance of two types of PbTe/ZnS multilayer filter.

- (i) Fabry-Perot Ge/LHLHLHHLHLH at 15μ
- (ii) Double Half wave Ge/LHLHHLHLHLHLHHLH at 15μ

These filters are identical to the basic bandpass filters required for the Selective Chopper Radiometer as described in Section 4.1.

5.4.3.1 Fabry-Perot Ge/LHLHLHHLHLH at 15μ

The Fabry-Perot filters were manufactured by monitoring in the 4th order to turning points in reflectance. On the assumption that the uncertainty in determining the turning point can be as large as 8% per quarter wave at the monitoring wavelength, the error is reduced to 2% per quarter wave at 15μ . Therefore, a standard deviation of 2% was taken for all the layers in the filter. Strictly, according to the above assumption, the error in the half wave spacer layer should be only 1% since the spacer is twice as thick as the quarter wavelength thick layers. However, it is convenient for the purpose of illustrating the effect of the random errors to assume the same S.D. for all the layers.

In the table below is listed the data for 12 calculations based on a S.D. of 2% in layer optical thickness. The results of these calculations are shown in Figures 5.10 (a), (b), (c) and (d). The second table below lists the peak wavelengths of the various filters in the first table along with their half widths as measured from the above Figures.

The standard deviation of the spacer layer, as calculated from the data is 1.98%. This of course, is in good agreement with the stipulated S.D. of 2% which was used as input data in the calculations. However, the S.D. of the shift in peak wavelength of the multilayers is only 1.2% which is 40% less than the variation in spacer thickness. This is a very interesting result since it illustrates the influence of the reflecting stacks on the position of the pass band.

Optical Thickness (fraction of 15 μ)													
LAYER	Datum	Run 1	2	3	4	5	6	7	8	9	10	11	12
1 L	. 25	. 2567	. 2534	. 2590	. 2493	. 2513	. 2496	. 2448	. 2482	. 2579	. 2436	. 2510	. 2521
2H	. 25	. 2458	. 2444	. 2468	. 2421	. 2573	. 2436	. 2474	. 2514	. 2467	. 2518	. 2510	. 2540
3L	. 25	. 2545	. 2550	. 2493	. 2552	. 2385	. 2580	. 2440	. 2437	. 2596	. 2496	. 2507	. 2556
4H	. 25	. 2530	. 2454	. 2465	. 2487	. 2450	. 2526	. 2597	. 2500	. 2465	. 2521	. 2501	. 2469
5L	. 25	. 2611	. 2554	. 2534	. 2474	. 2519	. 2526	. 2494	. 2403	. 2475	. 2493	. 2442	. 2528
6HH	. 5	. 5080	. 5003	. 4900	. 4929	. 5082	. 4759	. 5065	. 4996	. 5051	. 5125	. 4960	. 5070
7L	. 25	. 2515	. 2495	. 2463	. 2558	. 2515	. 2435	. 2511	. 2483	. 2517	. 2528	. 2464	. 2438
8H	. 25	. 2537	. 2586	. 2522	. 2555	. 2493	. 2444	. 2531	. 2408	. 2548	. 2441	. 2496	. 2538
9L	. 25	. 2406	. 2524	. 2529	. 2454	. 2424	. 2457	. 2490	. 2474	. 2521	. 2500	. 2524	. 2485
10H	. 25	. 2522	. 2558	. 2532	. 2507	. 2559	. 2572	. 2491	. 2581	. 2534	. 2507	. 2499	. 2429

F.P. data for 2% Standard Deviation

Run	Peak Wavelength (microns)	Half width (microns)
1	15.145	.097
2	15.07	.097
3	14.83	.095
4	14.92	.094
5	15.13	.093
6	14.565	.095
7	15.145	.095
8	14.87	.095
9	15.105	.095
10	15.22	.094
11	14.87	.095
12	15.15	.095

Peak wavelengths, halfwidths for 2% S.D.

The variation in half width is approximately equal to the percentage shift in peak wavelength i.e. expressed as a percentage of the mean half width of .095 microns the S.D. in half width is 1%, but this is probably due to the uncertainty in peak height since the plotter linearly interpolates between the calculated points.

There appears to be very little change in peak transmission over all twelve calculations if allowance is made for truncation of the peak in the graphs.

Hitherto it had been assumed that the position of the pass band of a Fabry Perot multilayer was determined by the thickness of the spacer half wave layer. The above results suggest that the reflecting stacks also play a significant part in pitching the filter. In fact, the errors in the stacks oppose, to some extent, changes in spacer thickness and result in an overall increase in monitoring accuracy. However, the problem of accurately pitching these narrow band filters is still very great as can be seen from the following example based on the above results.

The narrow band Fabry Perot filters for the SCR are required to be centred on 667cm^{-1} to within $\pm 0.15\text{cm}^{-1}$ which implies a pitching accuracy of $\pm 0.23\%$ in peak wavelength. If the S.D. of the shift in peak wavelength is

LAYER	Optical Thickness (fraction of $\lambda_0 = 15\mu$)												
	Datum	Run 1	2	3	4	5	6	7	8	9	10	11	12
1L	.25	.2436	.2442	.2485	.2448	.2403	.2521	.2493	.2519	.2457	.2567	.2554	.2529
2H	.25	.2518	.2480	.2429	.2474	.2498	.2534	.2421	.2541	.2572	.2458	.2501	.2532
3L	.25	.2496	.2464	.2620	.2440	.2483	.2438	.2552	.2515	.2541	.2545	.2495	.2532
4HH	.50	.5043	.4992	.5116	.5194	.4817	.4965	.4974	.4987	.5025	.5060	.5172	.5159
5L	.25	.2493	.2524	.2442	.2494	.2474	.2467	.2474	.2424	.2537	.2611	.2524	.2473
6H	.25	.2562	.2499	.2574	.2532	.2581	.2493	.2464	.2559	.2515	.2540	.2558	.2464
7L	.25	.2528	.2521	.2502	.2511	.2579	.2559	.2558	.2496	.2546	.2515	.2590	.2552
8H	.25	.2441	.2540	.2477	.2531	.2467	.2466	.2555	.2436	.2430	.2337	.2468	.2536
9L	.25	.2500	.2556	.2449	.2490	.2596	.2513	.2454	.2580	.2468	.2406	.2493	.2518
10H	.25	.2507	.2469	.2518	.2491	.2465	.2501	.2507	.2526	.2548	.2522	.2465	.2446
11L	.25	.2510	.2528	.2434	.2482	.2475	.2480	.2513	.2526	.2402	.2534	.2534	.2521
12HH	.50	.5021	.5070	.4894	.5029	.5051	.4999	.5146	.4759	.5097	.4889	.4900	.4987
13L	.25	.2507	.2438	.2457	.2437	.2517	.2459	.2385	.2435	.2498	.2550	.2463	.2512
14H	.25	.2501	.2538	.2513	.2500	.2548	.2510	.2450	.2444	.2451	.2454	.2522	.2528

1.2%, corresponding to a 2% S.D. in monitoring layer thickness, then the probability of a filter lying within the prescribed limits is only .06. Put another way, this means that only 1 filter in 17 would be expected to be centred within the stipulated tolerance. Even if the S.D. of the shift in peak wavelength is reduced by half, only one filter in eight will be within tolerance.

5.4.3.2 Double Half Wave Ge/LHLHHLHLHLHHLH

A similar analysis of the effect of random errors in layer thickness was carried out on the double half wave design Ge/LHLHHLHLHLHHLH at 15μ . Initially twelve calculations were performed using a S.D. of 2% in layer thickness as in the Fabry Perot design. The results of these calculations are shown in Figures 5.11(a), (b), (c) and (d). The data used in the calculations is given in the table below. The second table below summarises the results of the computations and lists the peak wavelength and half width of each run. In that table, the column labelled λ_p gives the wavelengths at which peak transmission in the pass band occurs; the column labelled λ_c gives the centre wavelengths of the pass band as determined from the mean of the wavelengths at which the transmission has fallen to half the maximum value. In general, λ_p is different from λ_c except for the datum case (no errors).

Run	λ_p μ	λ_c μ	$ \lambda_p - \lambda_c $ μ	Half- width μ	$T_{\max}$ %
Datum	15.0	15.0	0	.30	98
1	15.05	15.065	-.015	.307	96.5
2	15.115	15.052	+.063	.347	76.0
3	14.73	14.964	-.234	.637	19.0
4	14.985	15.12	-.135	.469	41.0
5	15.11	14.914	+.196	.576	26.5
6	14.965	14.927	+.038	.312	91.0
7	15.16	15.071	+.089	.393	58.0
8	14.55	14.731	-.181	.573	27.0
9	15.05	15.087	-.037	.318	91.0
10	14.85	15.02	-.17	.54	30.0
11	14.815	15.056	-.241	.677	18.5
12	15.00	15.126	-.126	.46	43.0

Parameters in D. H. W. for 2% Standard Deviation

Figures 5.11 (a), (b), (c) and (d) show the sensitivity of this D.H.W. design to 2% random error in monitoring. Both the peak transmission and shape of the pass band are strongly affected by errors of this magnitude. If the peak transmission ($T_{\max}$) is plotted against half width, as in Figure 5.12, the points are seen to lie very close to a smooth curve. It can be seen from Fig. 5.12 that there is an initial rapid decrease in peak transmission with filter broadening but that the rate at which $T_{\max}$ falls off is less for the broader and more degenerate filters.

Figures 5.11 (a), (b), (c) and (d) show that two peaks occur in the pass band as the filter degenerates. The separation of the peaks is given approximately by the parameter $2|\lambda_p - \lambda_c|$. When $T_{\max}$ is plotted against $|\lambda_p - \lambda_c|$, as shown in Figure 5.13, the points lie on or near to a smooth curve. Also plotted in the Figure 5.13 is the variation in halfwidth with $|\lambda_p - \lambda_c|$. For very broad degenerate filters ($T_{\max} \sim 70\%$) the halfwidth is proportional to the 'peak separation', i.e. $|\lambda_p - \lambda_c|$.

The standard deviations of the peak wavelength shift (σ_p), the centre wavelength shift (σ_c) and the average halfwidth ($\bar{w}$) have been computed from Figures 5.11 (a), (b), (c) and (d) and are $\sigma_p = 1.2\%$, $\sigma_c = 0.72\%$ and $\bar{w} = .468 \mu$. It is interesting that σ_p for the D.H.W. filters is equal to the S.D. of the peak wavelength shift of the Fabry Perots.

Only four of the twelve runs show a peak transmission greater than 70%. Therefore the accuracy in monitoring must be better than 2% to produce a good yield of D.H.W. filters of this design with $T_{\max} > 70\%$. This is an agreement with the rough rule of the thumb that the mismatch between spacers should be less than the halfwidth of the D.H.W. filter. Using this simple criterion we obtain the permissible error in spacer thickness as follows.

If x and y are the thicknesses of the two spacer layers, the mismatch is given by $f = x - y$. If the probable error in spacer 1 is $\pm \alpha$ and this is assumed to be equal to the probable error in spacer 2, then the probable mismatch is given by

$$\Delta^2 = \alpha^2 \cdot \left(\frac{\partial f}{\partial x} \right)^2 + \alpha^2 \cdot \left(\frac{\partial f}{\partial y} \right)^2 = 2\alpha^2$$

If Δ is put equal to the halfwidth then the permissible value of spacer error is

$$\alpha = \frac{(\text{halfwidth})}{\sqrt{2}}$$

$\approx 1.4\%$ for the D.H.W. under consideration.

Accordingly a further series of 12 random error calculations was carried out with a standard deviation of 1.4% in layer thickness for comparison with the 2% S.D. cases. The data used in these calculations is given in the next table. The results are shown in Figures 5.14 (a), (b), (c) and (d) and are summarised in the second table.

The standard deviations of the peak wavelength shift (σ_p), the centre wavelength shift (σ_c), and the mean halfwidth ($\bar{w}$) are given below:

$$\begin{array}{rcl} \sigma_p & = & 0.86\% \\ \sigma_c & = & 0.51\% \\ \bar{w} & = & 0.399 \mu \end{array} \quad \begin{array}{l}) \\) \\) \\) \end{array} \quad \begin{array}{l} \text{S.D. in layer thickness} \\ \\ \\ = 1.4\% \end{array}$$

In this case, five of the twelve runs had a peak transmission greater than 70%. This represents a 25% increase in yield of good filters for a 30% increase in monitoring accuracy. However, the yield of good filters is still relatively small and it is estimated that, on the basis of the above figures, a yield of good filters of 50% or greater would be obtained only with a S.D. of 1% or less in monitoring accuracy.

It is also interesting to note that the standard deviations in peak wavelength and centre wavelength are proportional to the standard deviation in layer thickness, i.e.

$$\left[\frac{\sigma_p}{\text{S.D.}} \right] = \left[\frac{\sigma_p}{\text{S.D.}} \right] = .61$$

S.D. = 2% S.D. = 1.4%

and

$$\left[\frac{\sigma_c}{\text{S.D.}} \right] = \left[\frac{\sigma_c}{\text{S.D.}} \right] = .36$$

S.D. = 2% S.D. = 1.4%

Optical Thickness (fraction of $\lambda_0 = 15\mu$)													
LAYER	Datum	Run 1	2	3	4	5	6	7	8	9	10	11	12
1 L	0.25	.2455	.2459	.2490	.2464	.2432	.2515	.2495	.2513	.2470	.2547	.2538	.2520
2H	0.25	.2512	.2486	.2450	.2482	.2498	.2524	.2445	.2528	.2550	.2470	.2501	.2522
3L	0.25	.2497	.2475	.2584	.2458	.2488	.2456	.2537	.2511	.2528	.2532	.2496	.2522
4HH	0.5	.5030	.4994	.5081	.5136	.4872	.4975	.4981	.4991	.5018	.5042	.5120	.5111
5L	0.25	.2495	.2517	.2460	.2496	.2482	.2477	.2482	.2447	.2526	.2578	.2516	.2481
6H	0.25	.2543	.2499	.2552	.2523	.2557	.2495	.2475	.2541	.2510	.2528	.2541	.2475
7L	0.25	.2519	.2515	.2501	.2508	.2555	.2541	.2540	.2497	.2532	.2510	.2563	.2536
8H	0.25	.2458	.2528	.2484	.2521	.2477	.2476	.2538	.2455	.2451	.2526	.2477	.2525
9L	0.25	.2500	.2539	.2464	.2493	.2567	.2509	.2468	.2556	.2477	.2434	.2495	.2512
10H	0.25	.2505	.2478	.2513	.2494	.2475	.2501	.2505	.2518	.2471	.2515	.2475	.2462
11L	0.25	.2507	.2520	.2454	.2487	.2482	.2486	.2509	.2518	.2431	.2524	.2524	.2515
12HH	0.5	.5051	.5049	.4926	.5020	.5036	.4999	.5102	.4831	.5068	.4922	.4930	.4991
13L	0.25	.2505	.2457	.2469	.2455	.2512	.2471	.2419	.2455	.2499	.2535	.2474	.2508
14H	0.25	.2501	.2527	.2509	.2500	.2534	.2507	.2465	.2461	.2466	.2468	.2515	.2519

DHW data for S.D. = 1.4%

Results of D.H.W. Parameters, S.D. = 1.4%

Run	$\lambda_p \mu$	$\lambda_c \mu$	$\lambda_p - \lambda_c \mu$	Half-width μ	$T_{\max}$ %
1	15.035	15.048	-.013	.30	97
2	15.085	15.039	+.046	.322	85.5
3	14.8	14.971	-.161	.524	32
4	14.99	15.083	-.093	.408	58
5	15.075	14.945	+.13	.457	41.5
6	14.975	14.950	+.025	.308	94.5
7	15.115	15.048	+.067	.347	73.5
8	14.68	14.807	-.127	.454	42
9	15.035	15.067	-.025	.31	94.5
10	14.895	15.014	-.119	.439	46.5
11	14.875	15.039	-.164	.531	31
12	15.00	15.089	-.089	.387	60

Furthermore, if $T_{\max}$ and the half width are plotted against the 'peak separation', $|\lambda_p - \lambda_c|$, the points lie on substantially the same curves as in Figure 5.13. The same comments apply to the curve of $T_{\max}$ v. halfwidth which is very similar to Figure 5.12. The conclusion must be that these curves are typical of the filter design and not dependent on the random errors at least for small values of the standard deviation in layer thickness. The small amount of scatter of the points about the curves is partly due to inaccuracies in reading from the transmission profiles in Figures 5.11 and 5.14.

The influence of the quarter wave layers on the performance of the D.H.W. filter can be considerable, as for the Fabry Perot. In particular, it has been found possible to alter the peak by varying the thickness of the final layer. Figure 5.15 shows how the passband alters the final layer thickness in the case of a filter nominally monitored at $\lambda_o = 14.82\mu$ but with a 2% S.D. in layer thickness. The data for the calculations is given in the table below.

As the thickness of the final layer is increased from $0.2 \lambda_0$, the peak transmission of the filter decreases from a maximum of 88% and the pass band shifts to longer wavelengths. Presumably, if the final layer had been monitored to approximately a quarterwave then the transmission of the pass band would have been similar to Run 2. Therefore by carefully adjusting the final layer it has been possible, in this case, to improve the peak transmission. This is very important from the filter manufacturers point of view, since it offers a certain amount of control over D.H.W. filter performance through adjustment of the final layer during deposition despite random errors in the previous layers. Inevitably, however, there is a decrease in performance compared with the perfect filter which is given by the curves in Figures 5.12 and 5.13. Both $T_{\max}$ and hence the halfwidth are affected by monitoring errors. Thus, even in D.H.W. filters with $T_{\max} = 88\%$ there is a halfwidth broadening of approximately 7%.

Layer thicknesses

LAYER	Optical Thickness fraction of $\lambda_0 = 14.82 \mu$			
	Run 1	2	3	4
1 L	.2464			
2 H	.2566			
3 L	.2557			
4 <u>HH</u>	.5075			
5 L	.2467			
6 H	.2419			
7 L	.2491			
8 H	.2492			
9 L	.2604			
10 H	.2478			
11 L	.2518			
12 HH	.5184			
13 L	.2564			
14 H	.2000	.2500	.3000	.3500

5.4.4 Summary

The Fabry Perot filter and the D.H.W. filter are two different systems and, as expected, react in different ways to random monitoring errors. In particular neither the transmission profile nor the half width of the F.P. are much affected by errors which mainly cause a shift in peak wavelength to take place. In this respect, it is interesting to note that the S.D. in the peak wavelength shift of the F.P. filter is the same as that of the D.H.W. although the centre wavelength shift (σ_c) of the D.H.W. is much less ($\sigma_c = 0.6\sigma_p$).

The peak transmission of the D.H.W. is seriously affected by monitoring errors and a relationship exists between $T_{\max}$, halfwidth and peak separation such that significant broadening of the pass band takes place even for $T_{\max} > 90\%$. For a high yield of good D.H.W. filters of this design the S.D. in layer thickness must be 1% or less which represents a considerable monitoring problem.

Finally, in both the F.P. and D.H.W. filters, the errors in the quarter wavelength thick layers play a significant part in determining the peak wavelength. It has also been shown possible to improve on otherwise badly mismatched D.H.W. by altering the thickness of the final layer.

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6. ASSESSMENT AND CONCLUSIONS

The expectation at the commencement of this research programme was that multilayer interference filter techniques could be extended to $100\ \mu$ in wavelength and that the performance and manufacturing ease of filters at shorter wavelengths, notably $10\text{-}16\ \mu$, could be significantly improved.

Such progress turned crucially upon finding suitable coating materials and the conditions under which they could be deposited in 'thick', uniform layers of accurately controlled thickness.

We summarise here the main results under the three headings of the programme.

i) Materials

A large number of materials (20) were tried, some already known, some unknown in this context (i. e. thicknesses up to $\lambda/2$ at $100\ \mu$ were required).

Of the known materials, conditions were found for improved deposition of 'thick' layers of ZnS, PbTe and Ge and their optical properties studied in detail in the range $1\text{-}100\ \mu$. Adequate mechanical properties and transparency were obtained in regions where the basic absorption processes permitted, i. e. $1\text{-}25\ \mu$ and $> 70\ \mu$ for ZnS, $5\text{-}70\ \mu$ for PbTe and almost the complete region $2\text{-}100\ \mu$ for Ge.

Important details of the processes on deposition were uncovered by studying the semiconductor properties of PbTe films as a function of heat treatment.

Of the new materials, the best to date has been CsI ($5\text{-}80\ \mu$) but several others showed considerable promise.

The systems PbTe/ZnS and Ge/CsI were shown to be very successful combinations in multilayer filters each having adequate index contrast and between them covering the region.

(ii) Filters: Performance and Production

Significant advances were made in the performance (bandwidth down to $\frac{1}{2}\%$, centering to $\frac{1}{4}\%$) and production facility for filters in the 15μ region (PbTe/ZnS). This was primarily due to adequate temperature control and use of rotating sources with consequent improvement in uniformity and monitoring accuracy and yield.

Further advances in thickness control led to the successful trial of non-integral thickness designs with promise of allowing use of the more advanced design methods.

(iii) Long wave Band pass filters and blocking filters

Fig. 6.1 shows the results of six filters, two each aimed at 75, 85 and 95μ .

The full line and the dotted line (LW 19/3, LW 19/4) are multi-layer evaporated interference filters of the following design

P L	Silicon	L. H L L H P'
	Substrate	

where L is $\lambda/4$ of CsI at 75μ .

H is $\lambda/4$ of Ge at 75μ .

P is a thin protective coating of Ge (evaporated) covered by a thin brushed-on layer of polystyrene.

P' is a thin protective layer of brushed on polystyrene.

The full and dotted lines with peaks at 85 and 95μ give the transmission curves of interference filters each with two reflecting interfaces made from photo-etched copper meshes. The spacing of the meshes is accomplished by use of worked metal shims, normally $\lambda/2$ in thickness, with their centres etched away.

All six filters have an aperture of 1" and are appropriately mounted.

Centering of filters on specified wavelengths has been achieved within $2\ \mu$ or better (i. e. acceptable in relation to the half-width specified).

Peak transmissions (56% - 40%) reasonably approach the target value of 50%; these are related to the band width. These bandwidths vary from $2\ \mu$ to $7\ \mu$, in all cases well within specification.

The specification made no mention of integrated rejection requirements. All the filters have wide transmission bands well away from the fundamental pass band region - at around $50\ \mu$ and less. We have also developed a series of blocking filters to reject all this radiation. Fig. 6.2 is a Ge/CsI low pass multilayer filter with a sharp edge at $60\ \mu$. The combination of such filters with quartz can remove all the side bands in any of the filters of Fig. 6.1.

Conclusion

These results show that multilayer interference filter techniques have been extended, continuously, to $100\ \mu$ in wavelengths and that the specifications of the contract have been met.

The Investigators foresee that further progress, comparable with that reported here, can be made in all three areas of the present work.

TABLE 2.1
Semiconductor properties

Material	Energy gap (ev)	ϵ_s	n	M. P. °C	Groups
C	5.4	5.5	2.4	3550	IV
Si	1.15	11.8	3.42	1412	
Ge	0.65	16	4	958	
Sn	0.08			232	
Se	1.6	8.5	~2.5	220	VI
Te	0.33	25	4.9-6.3	Softens 35 449.7	
BP	6.0	11.6		3000	
AlP	2.5	11.6		1500	
AlAs	2.3			1600	III - V
AlSb	1.52	10.3		1060	
GaP	2.25	8.4	3.0	1350	
GaAs	1.35	13.5	3.34	1280	
GaSb	0.69	15.2	3.7	728	
InP	1.27	10.6	2.9	1055	
InAs	0.35	11.5	3.4	942	
InSb	0.17	16.8	4	525	
SiC	3.0	10.2		2700	IV-IV
PbS	0.37	17.9	4.1	1114	IV-VI
PbSe	0.26		4.6	1062	
PbTe	0.25		5.1-5.6	971	
SnTe				780	
SiO			2.0		V-VI
Bi ₂ Te ₃	0.15			580	
Cd ₃ As ₂	0.13			721	
Cd Sb	0.48			456	
ZnO	3.2	8.5		1975	VI
ZnS	3.65	8.3	2.3	(Sublimes below 1185)	
ZnSe	2.6	5.75		1515	
ZnTe	2.15	18.6		1239	
CdS	2.4	5.9	2.9	685	VI
CdSe	1.74	4.3		1350	
CdTe	1.5	11.0	2.6	1098	
HgS	2.5	5.86		583	
HgSe	0.3	14		798	
HgTe	0.2			670	

The index refers to 5 μ ; it may be purity dependent.

TABLE 2. 2
Collected Reststrahlen Wavelengths and Predicted Positions based
On The Homology Rule

Compound	Ref	λ (μ)	λ (Predicted)	Compound	Ref	λ (μ)	λ (Predicted)
CsF	1	78.6	66.5	RbBr	6	112	105
	2	87		Tl Br	6	210	124
KF	1	52.1	58.3	CdBr ₂	7	156	88.8
	2	52.6		CsI	3,6	163	132
LiF	1,3	32.9	36.9	KI	1,6	99	89.1
NaF	1,3	40.6	52.6	LiI	1	69.4	41.8
RbF	1	62.5	64.3	NaI	6	85.6	72.2
	2	64.1		RbI	6	134	117
TlF	4	67.5	68.1	Tl I	6	193	145
BaF ₂	5	54.3	48.7				
	3	52.9		AlSb	8	31.9	76.8
CaF ₂	5	38.9	45.2	GaSb	8	44.4	109
	3	37.6		InSb	8	57.6	126
SrF ₂	5	45.6	48.2	InAs	8	47.8	110
	3	46.1		GaAs	8	37.5	98.2
AgCl	6	97.1	84.2	InP	8	32.9	80.7
CsCl	6	101		GaP	9	27.3	75.8
KCl	3,6	70.8	70.6	ZnSe	10	46.5	97.7
LiCl	1	58.5	38.8	CdSe	10	54.1	112
	2	62.9		CdS (z)	10	37.1	81.6
NaCl	3,1	61	-	CdS (w)	10	38.4	
RbCl	6	84.1	81.7	PbS	11	154	86.2
TlCl	6	159		PbTe		318	
AgBr	6	125	111	CdTe	10	66.6	127
			\	ZnTe	10	52.6	108
CsBr	3,6	135	116	ZnS (z)	10	32.3	75.9
KBr	6	86.5	83.6	ZnS (w)	10	33.3	75.9
	3	84.7		HgTe	12	85	
LiBr	1	58.5	41.2	Mg ₂ Sn	13	53	54.3
	2	62.8		Mg ₂ Si	13	36	47.6
NaBr	6	73.9	69.2	Mg ₂ Ge	13	45	52.6
				MgO	3	24.8	49.3
				ZnO	1	24.2	58.5

The predicted wavelengths are arbitrarily scaled to NaCl.

TABLE 2.3

Collected Data (290°K) From Martin, D.H., Advanc. Phys. 14, 39. (1965)

Material	γ	ϵ_s	ϵ_∞	Material	γ	ϵ_s	ϵ_∞
LiF	0.053	8.96	1.90	CuI	0.12		
NaF	0.065	5.10	1.72	MgO	0.07	9.8	3.1
NaCl	0.055	5.91	2.31	CaF ₂	0.04	6.7	2.05
NaBr	0.067	6.38	2.63	BaF ₂	0.05	7.2	2.16
NaI	0.18	7.26	3.03	SrF ₂	0.03	6.6	2.07
KCl	0.064	4.87	2.16				
KBr	0.034	4.91	2.34	AlSb	0.02		
KI	0.048	4.14	2.59	InP	0.04		
RbCl	0.076	4.92	2.19	ZnS	0.01		
RbBr	0.056	4.87	2.33	PbS	0.21		
RbI	0.058	4.94	2.62				
CsCl	0.096		2.62				
CsBr	0.042	6.67	2.72				
CsI	0.055		2.96				
TlCl	0.25	32.3	5.1				
RlBr	0.17	30.6	5.41				
TlI	0.13		6.5				
AgCl	0.40	11.1	4.0				
AgBr	0.30	13.1	4.62				

Reststrahlen Damping

TABLE 2. 4

Properties of Ionic Compounds

Material	Index	M. P.	Material	Index	M. P.
NaF	1.3	992	GaCl ₃		78
NaCl	1.5	801	GaBr ₃		122
NaBr		755	GaI ₃		211
NaI		661	AlCl ₃		189
KF		992	MgO	1.6	2800
KCl		768	Al ₂ O ₃ ·MgO		2030
KBr	1.53	728	Al ₂ O ₃	1.6	2030
KI	1.6	680	La ₂ O ₃		2315
AgCl	2.0	458	SnO ₂		1080 (dissoc)
AgBr		422	BaO	1.98	1293
AgI		555			
LiF	1.3	842	GeO ₂		1115
LiCl		606	CeO ₂		1950
LiBr		550	SnO ₂		1127
LiI		452	SiO ₂	1.4	1710(soft)
			(Synthetic)		
CsF		684	TiO ₂	2.2	1825(soft)
			SrTiO ₃	2.3	2080
			BaTiO ₃	2.4	1600
CsCl		636			
CsBr	1.67	636	GeS ₂		530(subl)
CsI	1.74	621	SnS ₂		882
RbCl		717			
RbBr		681	ZnAs ₂	n _{AV} = 3.38	
TlCl	2.1	427	GeS ₂		800(subl)
TlBr	2.2	458	GeSe ₂		707(dissoc) in water)
TlI	2.55				
CaF ₂	1.4	1360	SnSe ₃		861
SrF ₂		1190			
BaF ₂	1.45	1280	Mo ₂ S ₃		1100(dissoc)
MgF ₂	1.38	1255	Sb ₂ S ₃	2.7	
CdF ₂					
PbF ₂		855	5NaF·3AlF ₃	1.35	
BaCl ₂		950	CaCO ₃	1.5-1.6	894(decomp)
BaBr ₂		847			
CdI ₂		568	NaNO ₃	1.3	306.8
PbCl ₂		500			
PbBr ₂		373			
PbI ₂		402			

Refractive Indices are for 5μ.

TABLE 3.1

Thick films on wedged Si substrates

Film Material	Identification*	Thickness (μ)			Refractive Index		
		Optical	Metrical		Taly surf	Inter-fer	Ampli-tude data
			Taly-surf $\pm \frac{1}{2} \mu$	Inter-fer			
LEAD	MS 16	21.3(4 μ)	3 $\frac{1}{2}$	3.84 $\pm$ 0.05	6.1 $\pm$ 1.0	5.5 $\pm$ 1.0	5.8 $\pm$ 0.2
TELLURIDE	MS 15	42 (4 μ)	7	7.2 $\pm$ 0.1	6.0 $\pm$ 0.5	5.8 $\pm$ 0.3	5.8 $\pm$ 0.2
GERMANIUM	MS 28	16.7(10 μ)	4	4.2 $\pm$ 0.05	4.2 $\pm$ 0.4	4.0 $\pm$ 0.2	4.2 $\pm$ 0.2
	MS 29	30.1(10 μ)	7	7.2 $\pm$ 0.1	4.2 $\pm$ 0.3	4.2 $\pm$ 0.2	4.2 $\pm$ 0.2
	MS 30	39.3(10 μ)	9	9.9 $\pm$ 0.1	4.3 $\pm$ 0.2	3.95 $\pm$ 0.2	-
ZINC	MS 24	~20					
SULPHIDE	MS 12	43(4 μ)	19		2.26 $\pm$ 0.1	-	-
	MS 11	59(4 μ)	25		2.34 $\pm$ 0.1	-	-
SILICON	MS 5	~20					
MONOXIDE	MS 13	42.0(4 μ)	23 $\frac{1}{2}$	24 $\pm$ 1	1.8 $\pm$ 0.1	1.75 $\pm$ 0.1	-
	MS 14	63.8(4 μ)	31		2.06 $\pm$ 0.1	-	-
CAESIUM IODIDE	-	20.5(4 μ)	Too Soft	11.2 $\pm$ 0.7		1.8 $\pm$ 0.1	-

*See Table 4.3

TABLE 3.2
Low Index Films

Material	Comment
CsI	$n \approx 1.8$ (5μ)
CsBr	$n \approx 1.7$ (5μ)
TlI	$n \approx 2.6$ (6μ)
TlBr	$n \approx 2.2$ (6μ)
RbI	$n \approx 2.6$ (10μ)
RbBr	$n \approx 2.4$ (10μ)
InI ₂	$n \approx 2.65$ (10μ)
CdI ₂	$n \approx 2.9$ (10μ)
CdS	Opaque 7μ to at least 25μ
As ₂ S ₂	$n \approx 1.8 - 2.0$
In ₂ S ₃	$n \approx 1.8 - 2.0$

TABLE 3.3

Deposition of new Materials

Material	Source Power Start/ Finish Watts	Total Thickness QWOTS	Efficiency Opt. Thickness (Microns) per gm.	Refractive Index †	Comments
CsI	60	12	5.2	1.8 @ 5 μ	Shortwave scattering due to xtallinity
CsBr	85	19	5.1		
InI ₂	45	18	2.5	2.60, 4 to 10 μ	No melting
ZnSe	310 (Max available)	0	0		Reacted with boat
ZnO	320 (Max available)	0	0		Xtalline, opaque 7-25 microns
CdS	220/300	10	1.5	2.2 @ 2.6 μ	Opaque below 8 microns
CdI ₂	45	9 (estimated)	3.2	2.9 @ 10 μ	Excellent but easily reevaporable
TlI	45	13	1.0	2.55 3 to 7 μ	Excellent but easily reevaporable
TlBr	40	14	0.76	2.2 @ 3 to 8 μ	Considerable outgassing
BaS	330 (Max available)	0	0		Excellent material
In ₂ S ₃	250	10.5	1.3	1.8 - 2.0 3 to 8 μ	Slight melting
Ga ₂ S ₃	220 (Max available)	0	0		Excellent material
As ₂ S ₂	48	23	5.5	1.8 - 2.0 @ 10 μ	Excellent material
RbI	110	16	2.2	2.6 @ 10 μ	Excellent material
RbBr	94	16	2.2	2.4, 2.5 to 9 μ	Excellent material
Bi ₂ I ₃	50	4			Opaque below 6 microns poor transparency.
PbTe (open boat)	250	12	2.6*		
(Lidded boat)	95	12	1.7*		
(crucible)	75	12	3.3*		

† Preliminary measurements only, by transmission fringe contrast. (See Table 3.2)

* Evaporation data given for comparison with other materials; test depositions at 300°C for subsequent evaluation of equilibrium number of charge carriers.

TABLE 4.1

D. H. W. Filters for radiometer

Filter	Peak Wavelength (microns)	Maximum transmission %	Halfwidth (microns)
DHW 4C (monitor)	14.40	51	0.28
DHW 40 A	14.34	42	-
DHW 40 B	14.29	55	0.21
DHW 40 C	14.27	48	-
DHW 40 D	14.14	53	-

TABLE 4.2

F. P. Filters for Radiometer

Filter	Peak Wavelength (microns)	Peak transmission	Halfwidth (microns)
8172/159 A	14.61	50	
B	14.60	44	
C	14.61	47	
D	14.59	47	
E	14.60	44	
F	14.66	44	
Monitor	14.58	47	

TABLE 4.3**Thick films on Wedged Si Substrates**

Film Material	Film	Optical Thickness (μ)	Deposition temperature $^{\circ}\text{C}$
Lead Telluride	MS 16	21	250
	MS 15	43	250
Germanium	MS 28	17.7	200
	MS 29	31.9	200
	MS 30	40.7	200
Zinc Sulphide	MS 24	20	100
	MS 12	44.9	100
	MS 11	61.5	100
Silicon Monoxide	MS 5	20.7	
	MS 14	43.8	100
	MS 13	64.6	100
CsI			100

(See also Table 3.1)

TABLE 4. 4

Properties of Ge, Si, CsI relevant to Longwave filters

Material	Density g/cm ³	Thermal conduct- ivity cal/ cm. sec °C	Specific heat	Coeff. of expansion °C ⁻¹	Youngs Modulus p. s. i.
Ge	5. 4	0. 14	. 074	5. 5 - 6. 1 x 10 ⁻⁶	14. 9x10 ⁻⁶
Si		0. 309	. 168	4. 2 x 10 ⁻⁶	19. 0x10 ⁶
CsI	3. 5	0. 0027	. 048	50 x 10 ⁻⁶	0. 769x10 ⁶

TABLE 4. 5

Longwave CsI/Ge F. P. filters

Filter	Peak wavelength λ μ	Peak Transmission %	halfwidth $\Delta\lambda \frac{1}{2}$ μ	Effective R
LW19/1	74. 5	51	5. 5	. 80
LW19/3	73.	55	5	. 81

TABLE 4. 6

Summary of evaporated filters produced for the Contract

Filter	Type	Comments
AQ/1	Quartz + 5 μ edge filter	These filters provide blocking to 40 μ
AQ/2	"	
CQ/1	"	
CQ/2	"	
CQ/3	"	
AQ/6	Silicon + μ edge filter	Can be used with quartz to provide blocking to 40 μ
LW15/2	Low pass at 47 μ on silicon	Low pass filters which link up with quartz edge at ~ 40 μ Filters LS15/2 and LW15/3 do not show such a steep cut on as the others
LW15/3	"	
LW26/2	Low pass at 57 μ on silicon	
LW28/1	"	
LW28/2	"	
LW28/3	"	
LW19/1	Bandpass at 75 μ	Batch LW19 generally shows good transmission but LW23 is poor due to CsI absorption at 85 μ
LW19/3	Bandpass at 75 μ	
LW19/5	Bandpass at 70 μ	
LW19/7	Bandpass at 74 μ	
LW23/4	Bandpass at 83 μ	
LW23/5	Bandpass at 85 μ	

Filter design	3rd Order			1st Order		Wing		Heat Treatment
	T _{max} Before/After %	Peak Shift $\frac{\Delta\lambda}{\lambda}$ %	T _{max} %	Shift %	T _{max} %	Shift %		
SUB:LHLHHLH (6 layer F. P.)	31/41	+2.5	0/50	-	35/63	4.9	45 mins. at 315°C Slow cooling	
10 layer F. P.	5/13	2.2	0/12	-	23/51	3.3	10 mins. at 310°C. Rapid Cooling	
*10 layer F. P.	29/26	0.5	8/24	0.4	26/54	1.4	10 mins. at 310°C Rapid Cooling	

* Different evaporation.

TABLE 4.7

"Annealing" of F. P. Filters Containing PbTe

TABLE 4. 8
Mesh F. P. Filters

Mesh F. P.	$\lambda_0(\mu)$	d (%)	T max	Image size (mm)	Resolution
G. 1	95	3	0.4	2 x 10	200
G. 2	93.25	5.6	0.42	4 $\frac{1}{2}$ x 10	100
G. 3	96.5	6.2	0.48	4 $\frac{1}{2}$ x 10	100
G. 4	86	4.6	0.43	5 $\frac{1}{2}$ x 10	75
G. 5	87.5	6.1	0.46	3 x 10	150
G. 6	93	3.9	0.37	5 $\frac{1}{2}$ x 10	75
G. 7	84	5.2	0.50	4 $\frac{1}{2}$ x 10	100

TABLE 5.1 Equal-ripple parameters

Layer	Index	'g' parameter	$\left(\frac{n_S}{n_L} + \frac{n_S}{n_H}\right)$ etc.	Sin θ	Low Pass θ	Low Pass Fraction	High Pass θ	High Pass Fraction
SUB	4.0.							
1	Low, 2.2.	1.27	2.21	.575	35.1°	.545	144.9°	1.25
2	High, 5.1	1.44	1.82	.793	52.5°	.815	127.5°	1.10
3	Low, 2.2	2.22	2.61	.851	58.3°	.905	121.7°	1.05
4	High, 5.1	1.60	1.82	.880	61.6°	.955	118.4°	1.025
5	Low, 2.2	2.33	2.61	.894	63.4°	.985	116.6°	1.01
6	High, 5.1	1.625	1.82	.896	63.6°	.985	116.4°	1.01
7	Low, 2.2	2.34	2.61	.897	63.8°	.99	116.2°	1.005
8	High, 5.1	1.64	1.82	.902	64.4°	1.0	115.6°	1.0
9	Low, 2.2	2.34	2.61	.897	63.8°	.99	116.2°	1.005
10	High, 5.1	1.62	1.82	.892	63.1°	.98	116.9°	1.01
11	Low, 2.2	2.30	2.61	.882	61.9°	.96	118.1°	1.025
12	High, 5.1	1.56	1.82	.858	59.1°	.92	120.9°	1.045
13	Low, 2.2	2.055	2.61	.788	52.0°	.805	128°	1.10
14	High, 5.1	0.892	1.55	.576	35.0°	.545	145°	1.255
Anti-Reflect	Low, 2.2					2.0		0.5

TABLE 5.1
Equal-ripple parameters

MULTILAYER FILTERS FOR THE REGION 0.8 TO 100 MICRONS
FINAL SCIENTIFIC REPORT
(In Two Volumes)
CONTRACT AF61(052)-833

S.D. SMITH
J.S. SEELEY

University of Reading
Reading, England

ERRATA

Volume 1. Text and Tables (AFCRL-68-0496)

Page

- 2-13 Reference 2.3, for Mower read Mawer
3-11 Replace InI by InI₂
Replace CdI by CdI₂
Replace AsS by As₂S₂
3-20 4 lines from bottom ($\kappa = .0018$) should read ($\kappa = .0010$)
3 lines from bottom ($\kappa = .008$) should read ($\kappa = .0008$)
4-35 Equation (4.2) should read

$$\frac{(\Delta\lambda)^{\frac{1}{2}}}{(\lambda)} = \frac{(1-R)}{\pi \sqrt{R}}$$

- 4-46 Line 9, for 500 r.p.m. read 50 r.p.m.
Line 14, for 20° read 27.6°.
4-47 Line 2 of first table, read: -
Angle of incidence 0.4% 1.7%
4-50 Line 11 read as: - " ..as a rate process,
i.e., a function of (time x temperature) product? "
4-52 Line 6, for "non-uniformity" read "film non-uniformity"
4-54 Line 11, for "PbS and PbTe" read "PbS and PbSe"
4-55 Line 2, for "p-type films" read "p-type crystals"
4-59 Line 17, for "new saturation" read "near saturation"
4-62 Line 22, for "Burnstein" read "Burstein"
4-72 Ref 4.20, for "Burnstein" read "Burstein"

Volume 1, - Text and Tables (AFCRL-68-0496) Continued

4-74 The expression

$$A = \left(\frac{g}{2a} \frac{c}{\sigma\lambda} \right)^{\frac{1}{2}} R$$

should read

$$A = \frac{g}{2a} \left(\frac{c}{\sigma\lambda} \right)^{\frac{1}{2}} R$$

4-83 Reference 4.28 should read:

Culshaw, W., I.E.E.E. Trans. MTT-7, 221 (1959)

Reference 4.29 should read

Rawcliffe, R.D. and Randall, C.M., Applied Optics, 6, 1353 (1967).

Table 4.8 For $T_{\max}$ read $\tau_{\max}$

Volume 2, - Illustrations (AFCRL-68-0496)

Fig: 4.29 Replace "Wavenumber" by "Wavelength"

Fig: 4.31 For 70% read 50% (Transmission scale)

Fig: 4.32 Read CsI throughout in place of GsI.

Fig: 4.33 Read CsI throughout in place of GsI.

Fig: 4.37 Rear side of substrate is not antireflected.
For 40 read 400 (wavenumber scale)

Fig: 4.41 (a) Scale is time, in minutes.
Markings (d) and (e) are unnecessary.

Fig: 4.47 (a) Scale is transmittance.

Fig: 4.48 (a) 0 from Fig: 4.47 (b)
X from Fig: 4.47 (a)

Fig: 5.1 Cut-off frequencies are respectively
 ω_c , ω_c and ω_c' , ω_c .

Fig: 5.3 Replace "g" throughout by "q".

Fig: 5.4 $g_1 = \dots\dots$ not $q_1 = \dots\dots$

Fig: 5.5 Scale is relative frequency.

Fig: 5.6 Design is SUB: LHLL HLHLHLL HLHLH LL H

Fig: 5.7 Scale is relative wavenumber.

Security Classification		DOCUMENT CONTROL DATA - R & D	
(Security classification of title, how abstract and indexing annotation must be entered when the overall report is classified)			
1. ORIGINATING ACTIVITY (Corporate author) Physics Department University of Reading Reading, England		2a. REPORT SECURITY CLASSIFICATION Unclassified	
		2b. GROUP	
3. REPORT TITLE "..... Multilayer Filters for the region 0.8 to 100 microns"			
4. DESCRIPTIVE NOTES (Type of report and inclusive dates) Scientific Final.			
5. AUTHOR(S) (First name, middle initial, last name) S.D. Smith J.S. Seeley			
6. REPORT DATE 7th May 1968		7a. TOTAL NO. OF PAGES 192 in Vol. 1 6 in Vol. 2	7b. NO. OF REFS 28
8a. CONTRACT OR GRANT NO AF61(052)-833		8b. ORIGINATOR'S REPORT NUMBER(S)	
b. PROJECT NO 8662			
c. 4730		9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) IN TWO VOLUMES: Vol. 1 Text and Tables Vol. 2 Illustrations	
10. DISTRIBUTION STATEMENT This document has been approved for public release and sale; its distribution is unlimited.			
11. SUPPLEMENTARY NOTES TECH, OTHER		12. SPONSORING MILITARY ACTIVITY AIR FORCE CAMBRIDGE RESEARCH LABORATORIES L.G. Hanscom Field Bedford, MASS 01730	
13. ABSTRACT A study of materials for multilayer evaporated film filters for the region 1-100 μ has yielded several successful high index-low index combinations, notably PbTe/ZnS (5 - 25 μ) and Ge/CsI (10 - 80 μ). Filters of $\frac{1}{2}\%$ bandwidth, centred to $\frac{1}{2}\%$ can be produced on a routine basis for the 15 μ region. Band pass filters of 10% bandwidth between 50 μ and 100 μ have been achieved and evaporated film and mesh techniques have overlapped in the 60-70 μ region. Good blocking filters with edges out to 70 μ have also been obtained with performance better than other methods of filtering for this region.			

DD FORM 1473
1 NOV 63

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14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Multilayer filters, near infra-red, far infra-red, vacuum evaporation, micro-mesh, radiometry, semi-conductors.						

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