

18 AFOSR-TR-81-0615

CRITICALLY EVALUATED DATA ON OPTICAL AND TRANSPORT PROPERTIES OF SOLIDS
(Refractive Index and Absorption Coefficient of Selected Optical Materials).

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By

H. H. Li

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June 1981

FINAL TECHNICAL REPORT

on Contract F44620-76-C-0105

Prepared for

AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
Department of the Air Force
Bolling Air Force Base
Washington, DC 20332

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CENTER FOR INFORMATION AND NUMERICAL DATA ANALYSIS AND SYNTHESIS
Purdue Industrial Research Park
2595 Yeager Road
West Lafayette, Indiana 47906

AD A105059

DTIC FILE COPY

409062 81 10 2 102

-- 1 1 UP 1

-- 1 - AD NUMBER: A086658

-- 2 - FIELDS AND GROUPS: 20/6, 7/3

-- 3 - ENTRY CLASSIFICATION: UNCLASSIFIED

-- 5 - CORPORATE AUTHOR: CENTER FOR INFORMATION AND NUMERICAL DATA
ANALYSIS AND SYNTHESIS WEST LAFAYETTE IN

-- 6 - UNCLASSIFIED TITLE: CRITICALLY EVALUATED DATA ON OPTICAL AND
TRANSPORT PROPERTIES OF SOLIDS (REFRACTIVE INDEX AND ABSORPTION
COEFFICIENT OF SELECTED OPTICAL MATERIALS).

-- 8 - TITLE CLASSIFICATION: UNCLASSIFIED

-- 9 - DESCRIPTIVE NOTE: ANNUAL REPT. 1 MAY 79-30 APR 80.

--10 - PERSONAL AUTHORS: LI, H. H. ;

--11 - REPORT DATE: JUN 03, 1980

--12 - PAGINATION: 13P

--15 - CONTRACT NUMBER: F44620-76-C-0105

--16 - PROJECT NUMBER: 2308

--17 - TASK NUMBER: B1

--18 - MONITOR ACRONYM: AFOSR

--19 - MONITOR SERIES: TR-80-0539

--20 - REPORT CLASSIFICATION: UNCLASSIFIED

--23 - DESCRIPTORS: ♦OPTICAL MATERIALS, ♦ABSORPTION COEFFICIENTS,
♦CRYSTAL GROWTH, ♦REFRACTIVE INDEX, ♦IMPURITIES, ALKALI METAL
COMPOUNDS, BARIUM HALIDES, CONTAMINATION, FLUORIDES, GERMANIUM,

OPTICAL PROPERTIES, SOLIDS, CONTAMINATION, EXTRACTION, MAGNESIUM,
SILICON, STRONTIUM, SURFACES, PHONONS

--24 - DESCRIPTOR CLASSIFICATION: UNCLASSIFIED

--25 - IDENTIFIERS: SURFACE POLISHING, PE61102F, WUAFDSR2308B1

--26 - IDENTIFIER CLASSIFICATION: UNCLASSIFIED

--27 - ABSTRACT: IN THIS REPORT, EXPERIMENTAL MATERIAL ON THE ABSORPTION
COEFFICIENTS OF ALKALINE EARTH HALIDES WAS SEARCHED, COMPILED, AND
ANALYZED. IT WAS FOUND THAT THE BULK OF AVAILABLE DATA WAS
CONCENTRATED TO THE ABSORPTION EDGES OF THE MAIN TRANSPARENT REGION
FOR THE FOUR MATERIALS CALCIUM FLUORIDE, STRONTIUM FLUORIDE, BARIUM
FLUORIDE, AND MAGNESIUM FLUORIDE. AN EQUATION WAS FORMULATED TO
BEST DESCRIBE THE ABSORPTION DATA IN THE INFRARED MULTIPHONON
REGION AS A FUNCTION OF BOTH FREQUENCY AND TEMPERATURE. IT WAS
NOTED THAT THE SAME TYPE OF EQUATION IS EQUALLY VALID FOR BOTH
ALKALINE EARTH HALIDES AND ALKALI HALIDES. ALTHOUGH THE INTRINSIC
ABSORPTION WAS PREDICTED BY THE PROPOSED EQUATION, DISCREPANCIES
MIGHT OCCUR IN THE REGION WHERE ABSORPTION IS EXTREMELY LOW AND
WHERE EXTRINSIC ABSORPTIONS DUE TO IMPURITIES AND SURFACE
CONTAMINATION DOMINATE THE INTRINSIC ABSORPTION BY FACTORS RANGING
FROM FRACTIONS TO MULTIPLES OF TEN. EXPERIENCE HAS SHOWN THAT
EXTRINSIC CONTRIBUTIONS CAN BE REDUCED THROUGH IMPROVED CRYSTAL
GROWING AND SURFACE POLISHING TECHNIQUES. THE WORK ON THE
ABSORPTION COEFFICIENT OF SILICON AND GERMANIUM WAS IN PROGRESS. IT

WAS FOUND THAT THE ABSORPTION COEFFICIENT IS SENSITIVE TO THE
AMOUNT AND KIND OF IMPURITIES USUALLY BEING INTRODUCED AS DESIRABLE
DOPANTS. NUMEROUS EXPERIMENTAL MATERIAL WAS MADE AVAILABLE.
CURRENTLY THE WORK REMAINED IN THE STAGE OF DATA EXTRACTION AND
COMPILATION.

--28 - ABSTRACT CLASSIFICATION: UNCLASSIFIED

--29 - INITIAL INVENTORY: 4

--33 - LIMITATION CODES: 1

--34 - SOURCE SERIES: A

--35 - SOURCE CODE: 409062

--36 - DOCUMENT LOCATION: NTIS

--40 - GEOPOLITICAL CODE: 1802

--41 - TYPE CODE: 0

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM	
AFOSR-TR- 81 -0615		2. GOVT ACCESSION NO. AD-105059	
4. TITLE (and Subtitle) CRITICALLY EVALUATED DATA ON OPTICAL AND TRANSPORT PROPERTIES OF SOLIDS (Refractive Index and Absorption Coefficient of Selected Optical Materials)		5. TYPE OF REPORT & PERIOD COVERED Annual Technical Report 1 May 1976 - 30 April 1981	
7. AUTHOR(s) H. H. Li		6. PERFORMING ORG. REPORT NUMBER	
9. PERFORMING ORGANIZATION NAME AND ADDRESS CINDAS/Purdue University 2595 Yeager Road West Lafayette, Indiana 47906		8. CONTRACT OR GRANT NUMBER(s) F44620-76-C-0105 ✓	
11. CONTROLLING OFFICE NAME AND ADDRESS Air Force Office of Scientific Research Bolling AFB, DC 20032		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F 2308/B1	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		12. REPORT DATE June 4, 1981	
		13. NUMBER OF PAGES 25	
		15. SECURITY CLASS. (of this report) Unclassified	
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release. <i>Distribution Unlimited.</i>			
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)			
18. SUPPLEMENTARY NOTES			
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Absorption coefficient Alkaline earth halides Refractive index Silicon Optical constants Germanium Alkali halides			
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Under this contract a study was carried out of the optical properties of selected optical materials through data compilation, critical data evaluation and generation of recommended values based on available data from worldwide literature. The selected optical materials include alkali halides, alkaline earth halides, silicon and germanium, and III-V compounds. A major contribution of this work has been the exhaustive compilation of absorption coefficient and the formulation of an Urbach type equation for the multiphonon region near the infrared absorption			

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20. ABSTRACT (continued)

edge of alkali halides and alkaline earth fluorides. This equation is used to calculate the intrinsic absorption coefficient as a function of both wavelength and temperature in the temperature region of practical interest with the calculated result in concordance with the best contemporary experimental measurements for the pure samples.

Optical constants of silicon and germanium were studied in a similar manner as for the above mentioned materials. Refractive index data for silicon and germanium were exhaustively compiled and analyzed. Recommended values of refractive index for the transparent wavelength region were generated in the range 1.2 to 14 μ and 100-750 K for silicon, and 1.9 to 16 μ and 100-550 K for germanium. Generation of these values were based on a dispersion equation which best fits the selected data sets covering wide temperature and wavelength ranges. Both the refractive index and its temperature derivative are in concordance with the available data. Data compilation for the absorption coefficient of silicon and germanium is completed and is currently in the stage of data analysis and preparation of technical report which includes discussions on the absorption of lattice vibration, free carriers, impurities and absorption edges. Temperature and pressure dependences of energy gap are also presented.

The work on the refractive index of III-V compounds is currently at the stage of data extraction.

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TABLE OF CONTENTS

	<u>Page</u>
1. ABSTRACT	1
2. PERSONNEL INVOLVED WITH RESEARCH PROGRAM	1
3. INTERACTIONS	2
4. NEW DISCOVERIES, PATENTS OR INVENTIONS	2
5. STATUS OF RESEARCH EFFORT	2
5.1. Studies on the Absorption Coefficient of Alkali Halides	3
5.2. Studies on the Absorption Coefficient of Alkaline Earth Fluorides	10
5.3. Studies on the Optical Properties of Silicon and Germanium . .	10
5.4. Studies on the Refractive Index of III-V Compounds	21
6. PUBLICATIONS	21

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A. D. BLOSE

Technical Information Officer

1. ABSTRACT

Under this contract a study was carried out of the optical properties of selected optical materials through data compilation, critical data evaluation and generation of recommended values based on available data from worldwide literature. The selected optical materials include alkali halides, alkaline earth halides, silicon and germanium, and III-V compounds. A major contribution of this work has been the exhaustive compilation of absorption coefficient and the formulation of an Urbach type equation for the multiphonon region near the infrared absorption edge of alkali halides and alkaline earth fluorides. This equation is used to calculate the intrinsic absorption coefficient as a function of both wavelength and temperature in the temperature region of practical interest with the calculated result in concordance with the best contemporary experimental measurements for the pure samples.

Optical constants of silicon and germanium were studied in a similar manner as for the above mentioned materials. Refractive index data for silicon and germanium were exhaustively compiled and analyzed. Recommended values of refractive index for the transparent wavelength region were generated in the range 1.2 to 14 μm and 100-750 K for silicon, and 1.9 to 16 μm and 100-550 K for germanium. Generation of these values was based on a dispersion equation which best fits the selected data sets covering wide temperature and wavelength ranges. Both the refractive index and its temperature derivative are in concordance with the available data. Data compilation for the absorption coefficient of silicon and germanium is completed and is currently in the stage of data analysis and preparation of technical report which includes discussions on the absorption of lattice vibration, free carriers, impurities and absorption edges. Temperature and pressure dependences of energy gap are also presented.

The work on the refractive index of III-V compounds is currently at the stage of data extraction.

2. PERSONNEL INVOLVED WITH RESEARCH PROGRAM

During the progress of the work, the staff members who have contributed to the various tasks of this program are as follows:

Dr. H. H. Li
Dr. T. C. Chi
Dr. Y. S. Touloukian
Mrs. A. F. Furdyna
Mr. B. Runyan
Mr. D. J. Huff

In addition to the above effort, invaluable contribution was made by the professional personnel of the Division of Scientific Documentation at CINDAS in the literature search, data source identification and document procurement.

3. INTERACTIONS

The director of CINDAS, Dr. Y. S. Touloukian, frequently participated in meetings and conferences on the laser technology. He has discussed the recent development of high-power laser technology with the attendees and gained information useful to this research project. Dr. Touloukian has been constantly communicating with Dr. A. H. Guenther of the Air Force Weapons Laboratory at Kirtland, New Mexico regarding the subject of optical constants of laser window materials.

In addition to attending meetings on the related subjects, the principal investigator, Dr. H. H. Li, frequently discusses and exchanges information with the faculty of the Department of Physics at Purdue University on the optical and other related properties of solids. Results derived from these discussions are helpful in locating information and solving problems.

4. NEW DISCOVERIES, PATENTS OR INVENTIONS

There were no new discoveries, patents or inventions.

5. STATUS OF RESEARCH EFFORT

The objectives of this research project is to provide evaluated data and information on optical properties at high temperatures of selected solid materials of potential importance in laser window and laser-hardened material technologies.

The goals of the present work are:

- (i) to review the present state of knowledge on the refractive index and absorption coefficient of selected groups of pure solid substances related to the Air Force environment resistant material technology,
- (ii) to compile the available data and information,
- (iii) to critically evaluate and analyze the existing data, and to generate recommended values.

Selection of substances was based on the needs of the Air Force material technology and the recommendation of the National Material Advisory Board on high-power infrared-laser windows. The research program planned to continue to investigate the following groups of substances:

- (i) alkali halides
- (ii) alkaline earth halides
- (iii) diamond-structures materials (Ge and Si)
- (iv) III-V compounds (GaP, GaAs, GaSb, InP, InAs, and InSb)
- (v) (ZnS, ZnSe, ZnTe, CdS, CdSe, and CdTe)
- (vi) silver halides (AgCl, AgBr, and AgI)
- (vii) other IR materials to be selected.

We have completed the works for material groups (i) and (ii). At the time of writing this final technical report, the work on the material group (iii) is in the stage of data analysis and technical report preparation. The work on the material group (iv) is in the stage of data extraction.

5.1. Studies on the Absorption Coefficient of Alkali Halides

A typical absorption spectrum of alkali halide is shown in Figure 1. The fundamental transparent region of alkali halides is defined between the Urbach tail and multiphonon absorption. In the high transparent region, the transparency of the material is limited by many factors, notably the crystal vacancies, dislocations, impurities, surface contamination, etc. Extrinsic absorption due to these origins can be reduced through appropriate crystal growing, purification, annealing, and surface cleaning processes.

Investigation of the Urbach tail region may serve as an indicator to show the extent of impurity and/or defect contents. As generally observed, the purer the sample is, the more the validity of Urbach rule is extended to the

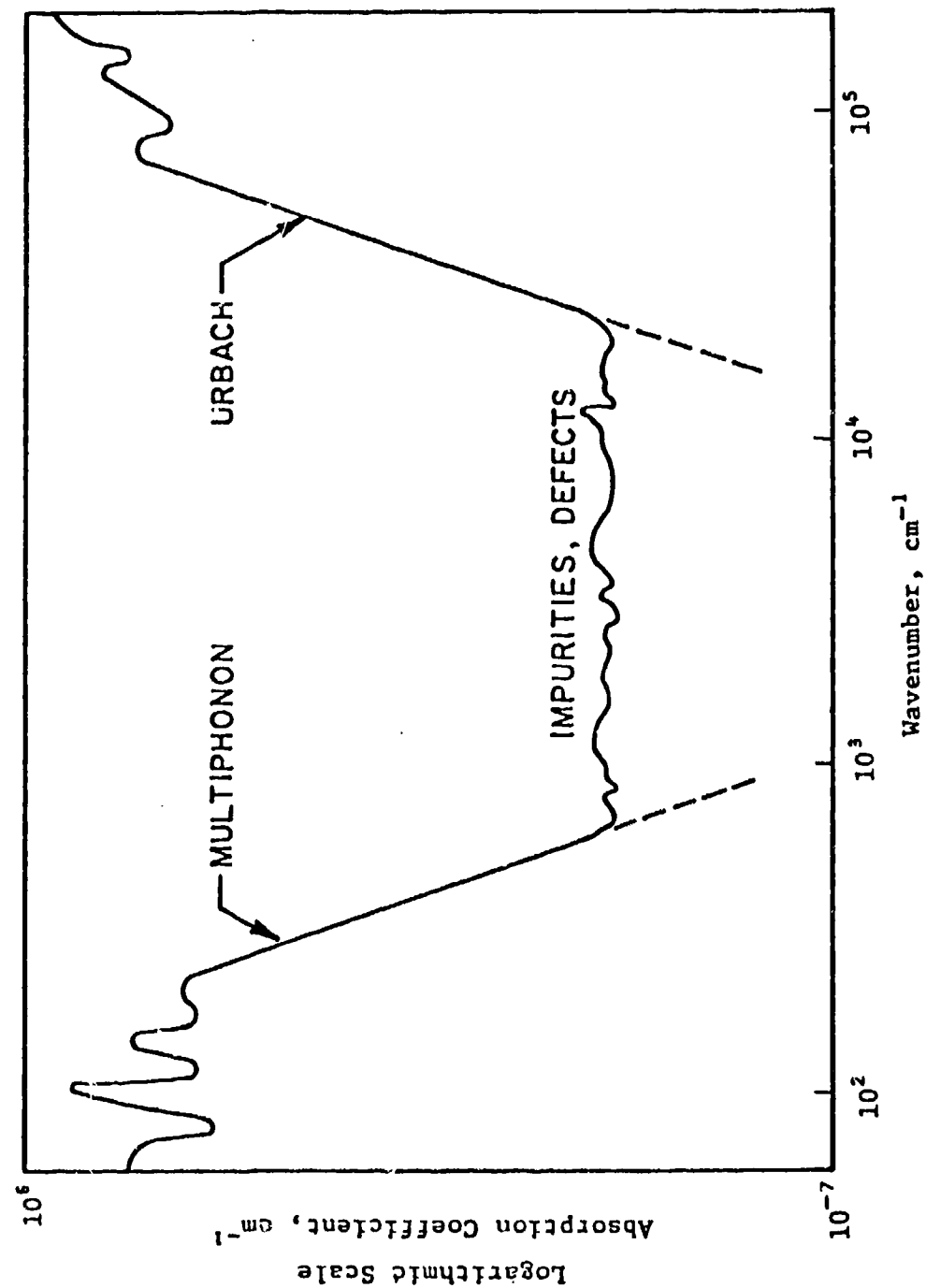


Figure 1. Absorption Spectrum of a Typical Alkali-Halide Crystal

transparent region. By comparing the absorption spectrum at the Urbach tail, the purer sample is self-revealed.

Absorption in the multiphonon region has been of current interest because of its application in windows for high-power infrared lasers. At the frequencies in this region, the total absorption can be attributed to processes involving several phonons, defect modes due to impurities, vacancies, or surface contaminations. A number of observations have been made to investigate the frequency and temperature dependence of the intrinsic multiphonon absorption. It has been found that the exponential dependence of the absorption coefficient on frequency holds for LiF, NaCl, KCl, and KBr at room temperature, i.e.,

$$\alpha = \alpha_0 e^{-W/v_0} \quad (1)$$

We have found that this exponential relation is also applicable in the cases of NaF and KI.

This exponential relation attracted considerable attention in the theoretical interpretation of such behavior. Two fundamentally different approaches have been taken to calculate the anharmonically induced absorption in ionic crystals. The most common approach requires the solution of the harmonic-lattice problem followed by a perturbation treatment of the anharmonicity. Sham and Sparks and McGill et al. employ diagrammatic techniques for evaluating the Green's functions, while Bendow et al. use the equation-of-motion method. Calculations using these methods become increasingly complex for higher-order processes and consequent simplification and approximation are made.

The results indeed predict the exponential dependence of absorption coefficient on the frequency. With regard to the temperature dependence, however, the results are not satisfactory. Discrepancies between experimental and theoretical predictions are of several orders of magnitude. In the case of NaF, NaCl and KCl, for example, theory predicts that temperature dependence of absorption coefficient follows the relation:

$$\alpha = T^{n-1} \quad (2)$$

where the values of n are 4, 6 and 7 for NaF, NaCl and KCl, respectively, but the corresponding experimental values are 2.6, 3.3 and 2.8.

In our studies, we have compiled available data for various spectral and temperature regions that have been experimentally investigated. For the data in the multiphonon region we found that the exponential relation also holds well at various temperatures and that the curves tend to converge to a point in the low frequency region. This situation is analogous to the Urbach rule in the ultraviolet absorption edge. Although the experimental temperature dependence of absorption coefficient does not agree with theoretical prediction as mentioned above, the observed trend does follow a power relation: $\alpha = T^a$ except at low temperatures. Based on these facts, it appeared that an equation of the form:

$$\alpha = \alpha_0 e^{-a(b+\nu)(c-\log T)} \quad (3)$$

could be used to represent the frequency and temperature dependence of absorption coefficient at temperatures higher than 200 K. Indeed we have found that the experimental data fit the expression satisfactorily. The best-fit values of the adjustable constants, α_0 , a , b , and c are given in Table 1.

Table 1. The Parameters of Alkali Halides

Crystal	α_0, cm^{-1}	a, cm	b, cm^{-1}	c
LiF	$10^{4.5116}$	44.08	0.002237	5.434
NaF	$10^{5.4453}$	81.87	0.00437	5.434
NaCl	$10^{4.9778}$	94.478	0.00591	5.434
KCl	$10^{6.4187}$	230.241	0.00700	5.434
KBr	$10^{4.9413}$	90.245	0.00886	5.434
KI	$10^{6.3105}$	207.251	0.0098	5.434

Analogous to the Urbach rule, the pair of constants, α_0 and b , defines the 'cross point' where the curves of α versus ν converge. The physical meaning of this point remains to be interpreted. Figures 2 and 3 provide a visual comparison of the predicted values from eq. (3) and experimentally determined data of sodium chloride. Figure 4 is a plot of the corresponding calculated values for a wider wavenumber region including the crossover point (b, α_0).

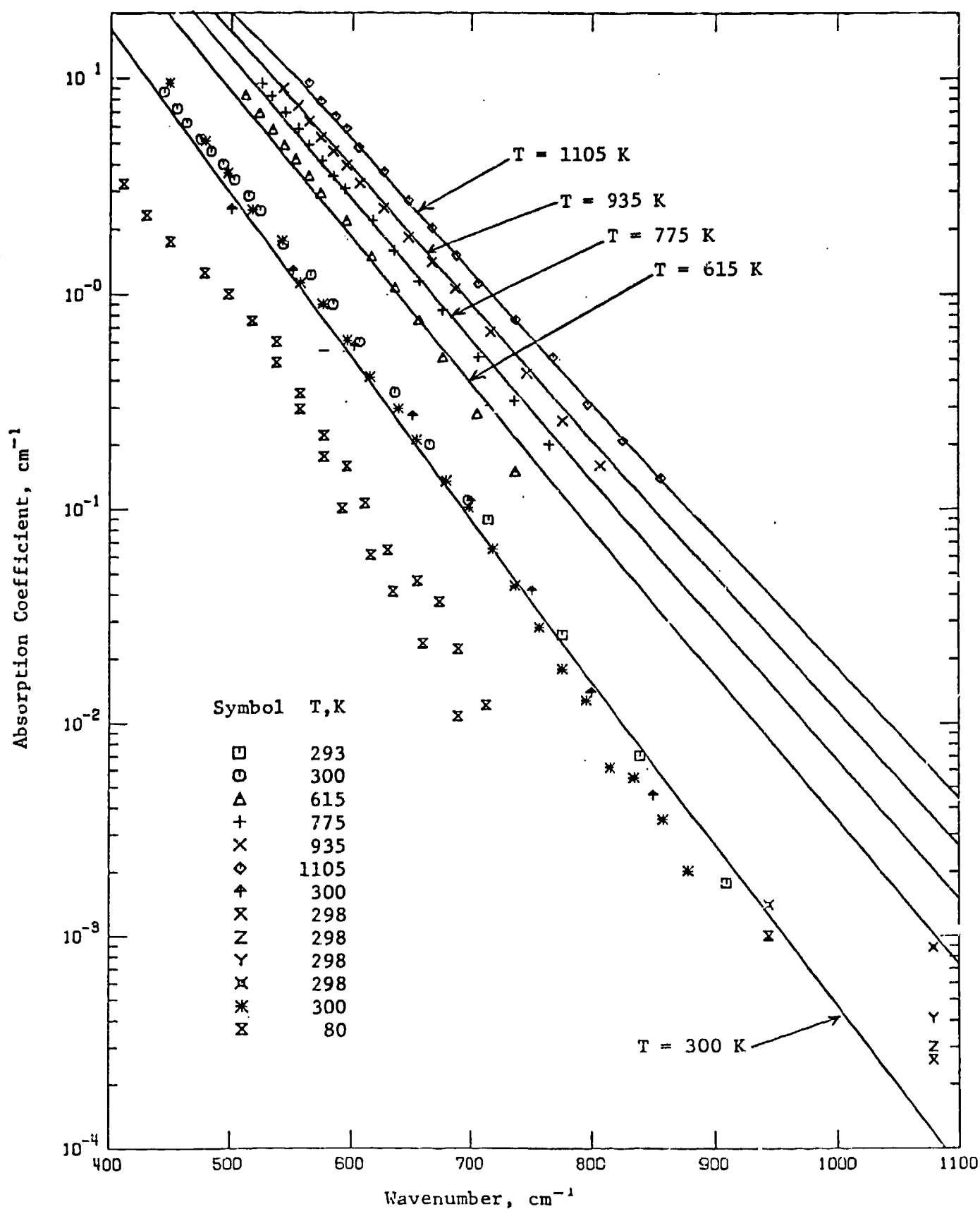


Figure 2. Absorption Coefficient of Sodium Chloride in the Multiphonon Region (Wavenumber Dependence)

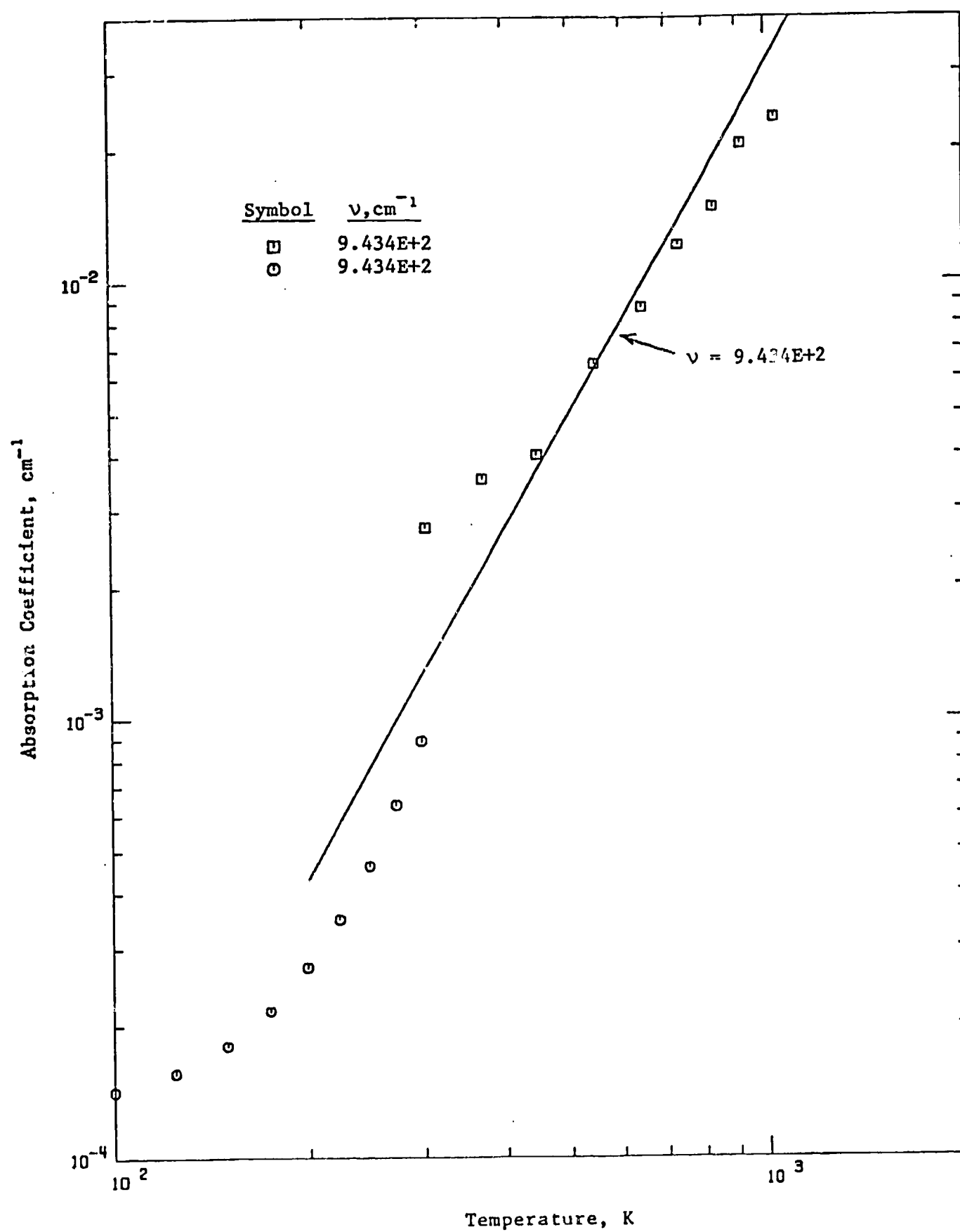


Figure 3. Absorption Coefficient of Sodium Chloride (Temperature Dependence)

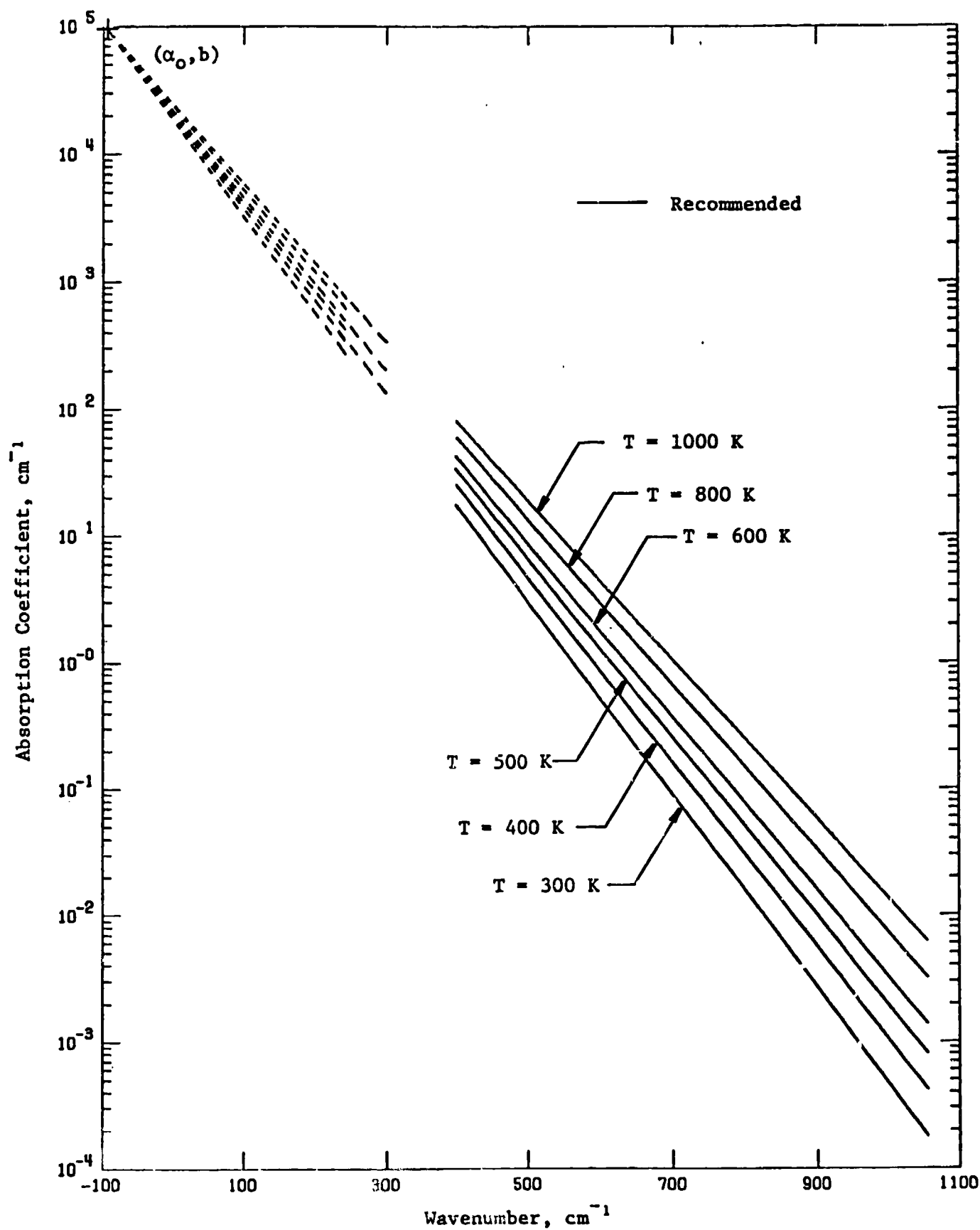


Figure 4. Calculated Absorption Spectra of Sodium Chloride

5.2. Studies on the Absorption Coefficient of Alkaline Earth Fluorides

In this study, we compiled all the available experimental data of absorption coefficient and related information useful for the Air Force material technology and laser research. In the data analysis on the available data of laser interest, we found that eq. (3) can be equally used to describe the absorption coefficient as a function of both frequency and temperature with appropriately determined constants, α_0 , a , b , and c for each material. These constants, listed in Table 2, were determined based on data evaluation, correlation, and synthesis. In the example of strontium fluoride, Figures 5 and 6 provide a visual comparison of the predicted values of eq. (3) and experimentally determined data. Figure 7 is the corresponding calculated value for a wider wavenumber region including the crossover point (b, α_0).

Table 2. The Parameters of Alkaline Earth Fluorides

Crystal	$\log \alpha$	a, cm	b, cm^{-1}	c
K_2F_2	3.1877	0.004184	-377.29	4.2901
CaF_2	3.0834	0.005251	-383.92	4.4498
SrF_2	2.9169	0.006577	-360.56	4.3288
BaF_2	2.8507	0.007349	-327.67	4.2873

As a counterpart of the Urbach rule for the uv absorption edge, we have established the expression, eq. (3), for the infrared absorption edge of alkaline earth fluorides. These expressions are of the same form and the parameters in the corresponding equations are similar. Compared with Deutsch's (eq. 1) expression, we have extended the dependence of the absorption coefficient to include the temperature in addition to frequency. It is worthwhile to point out that the equation formulated in this work is of the same type as it was found for alkali halides. Correlation techniques used for alkali halides are equally applicable in the present work.

5.3. Studies on the Optical Constants of Silicon and Germanium

The work on the optical constants of silicon and germanium is in the stage of data analysis and technical report preparation. Refractive index data are

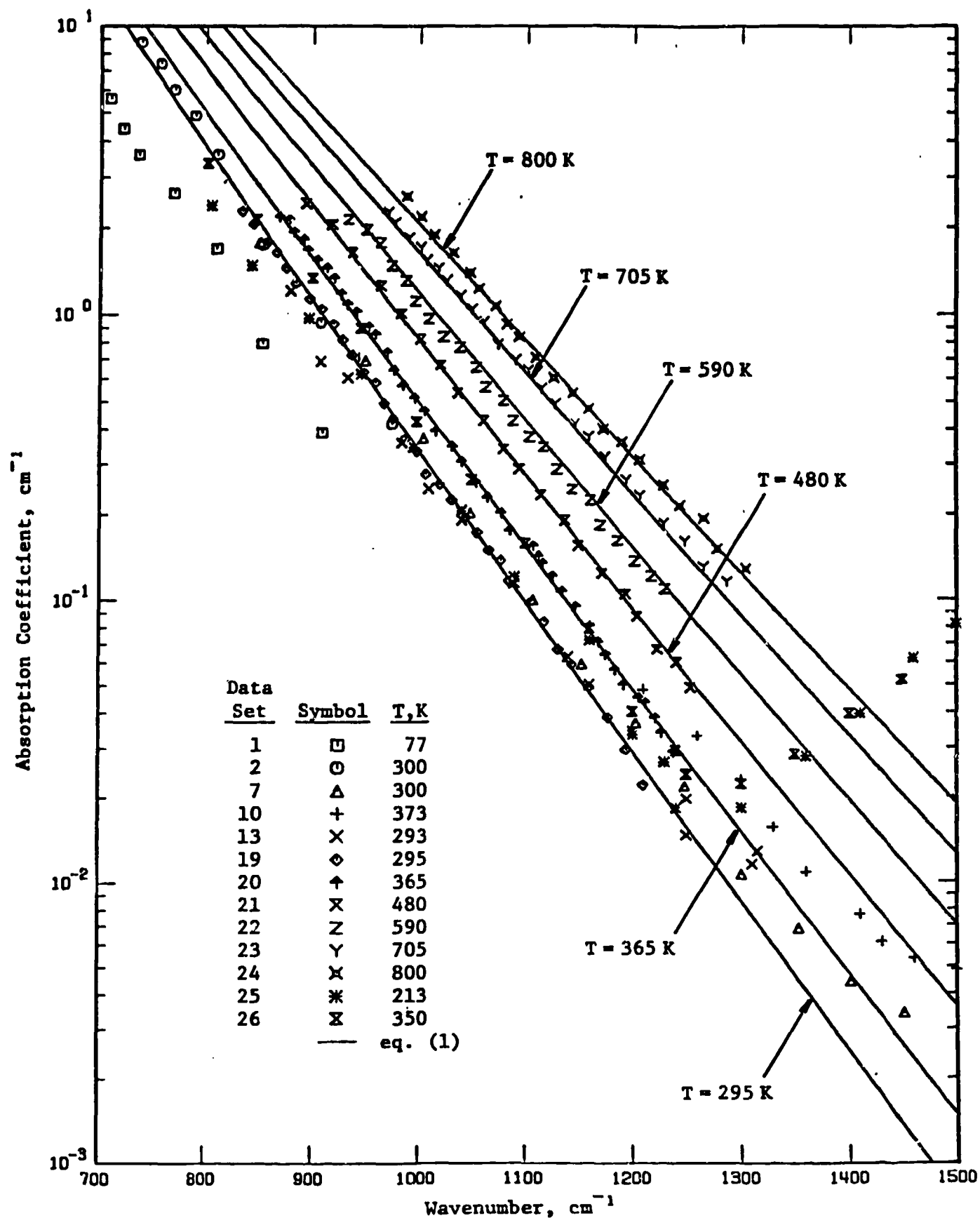


Figure 5. Absorption Coefficient of Strontium Fluoride in the Multiphonon Region (Wavenumber Dependence)

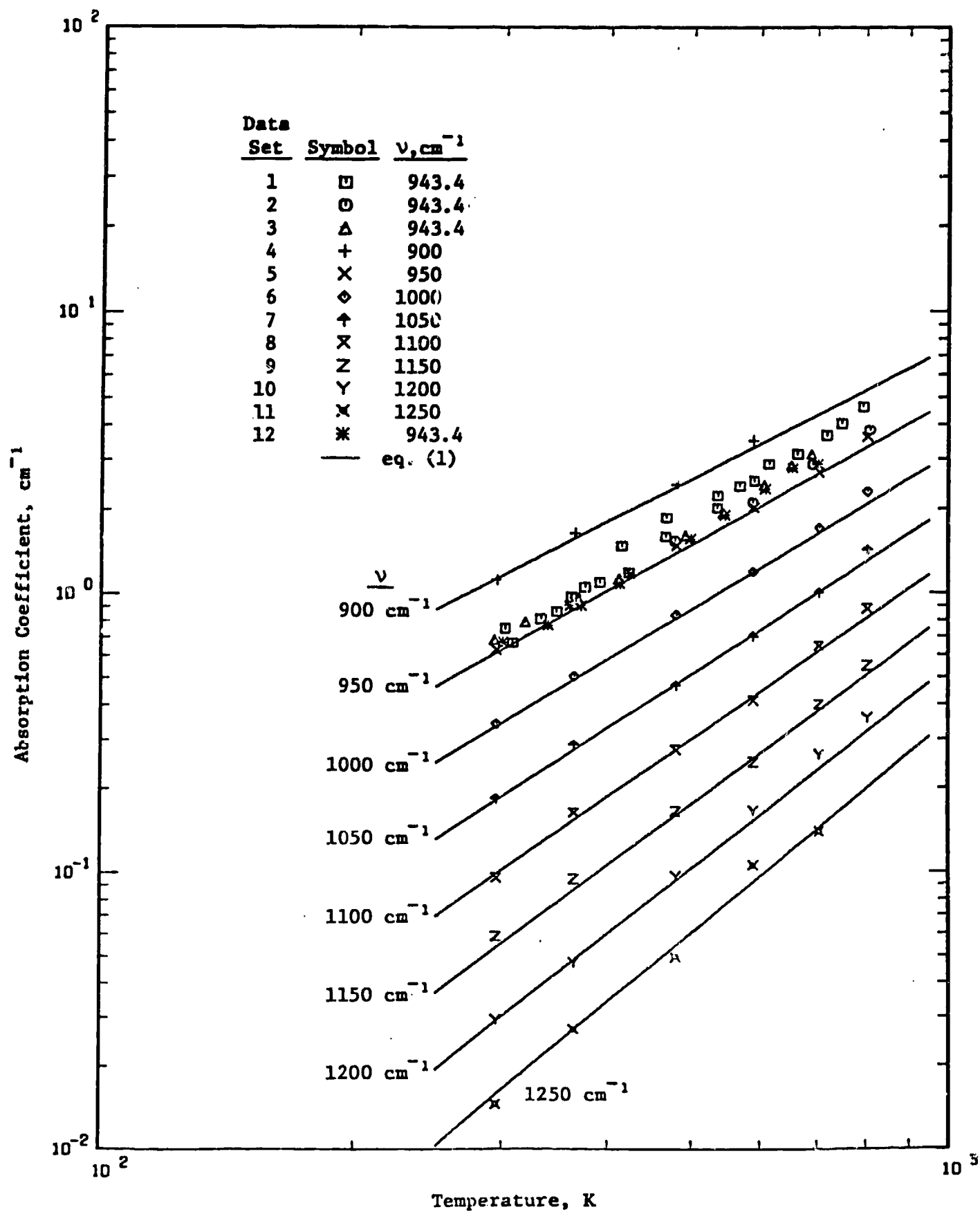


Figure 6. Absorption Coefficient of Strontium Fluoride in the Multiphonon Region (Temperature Dependence).

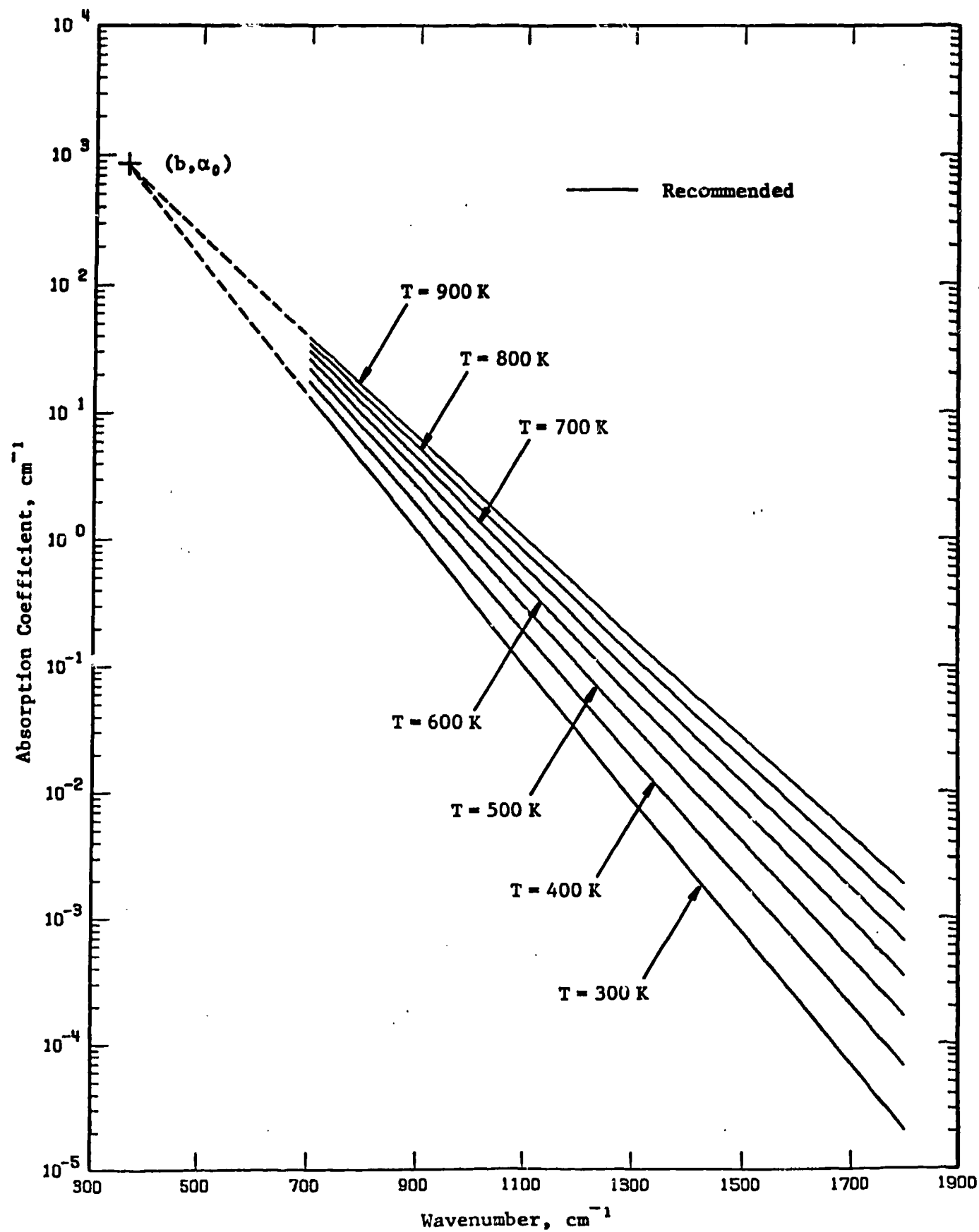


Figure 7. Calculated Absorption Spectra of Strontium Fluoride (Wavenumber Dependence)

available in the fundamental transparent wavelength region and are represented by the following dispersion equation through data analysis:

$$n^2(\lambda, T) = s(T) + \frac{L(T)}{\lambda^2} (A_0 + A_1 T + A_2 T^2) \quad (4)$$

where

λ = wavelength in units of μm ,

T = temperature in units of K,

$L(T) = e^{-3\Delta L(T)/L_{293}}$ = length of the sample at temperature T ,

$s(T)$ = static dielectric constant at temperature T , and

A_0 , A_1 , and A_2 are constants for a given material.

In the case of silicon, the parameters are:

$$s(T) = 11.4445 + 2.7739 \times 10^{-4} T + 1.7050 \times 10^{-6} T^2 - 8.1347 \times 10^{-10} T^3,$$

$$A_0 = 0.8948, A_1 = 4.3977 \times 10^{-4}, A_2 = 7.3835 \times 10^{-8},$$

and

$$\frac{\Delta L(T)}{L_{293}} = -0.021 - 4.149 \times 10^{-7} T - 4.620 \times 10^{-10} T^2 + 1.482 \times 10^{-11} T^3$$

(for temperature region 20-293 K)

and

$$\frac{\Delta L(T)}{L_{293}} = -0.071 + 1.887 \times 10^{-6} T + 1.934 \times 10^{-9} T^2 - 4.544 \times 10^{-13} T^3$$

(for temperature region 293-1600 K)

In the case of germanium, the parameters are:

$$s(T) = 15.2892 + 1.4549 \times 10^{-3} T + 3.5078 \times 10^{-6} T^2 - 1.2071 \times 10^{-9} T^3,$$

$$A_0 = 2.5381, A_1 = 1.8260 \times 10^{-3}, A_2 = 2.8888 \times 10^{-6},$$

and

$$\frac{\Delta L(T)}{L_{293}} = -0.089 + 2.626 \times 10^{-6} (T-100) + 1.463 \times 10^{-8} (T-100)^2 - 2.221 \times 10^{-11} (T-100)^3$$

(for temperatures $100 < T < 293$),

and

$$\frac{\Delta L(T)}{L_{293}} = 5.790 \times 10^{-6}(T-293) + 1.768 \times 10^{-9}(T-293)^2 - 4.562 \times 10^{-13}(T-293)^3 \quad (\text{for temperatures } 293 < T < 1200).$$

Figures 8 and 9 show the calculated refractive index and its temperature coefficient, dn/dT , of silicon, the latter is calculated from the first temperature derivative of eq. (4).

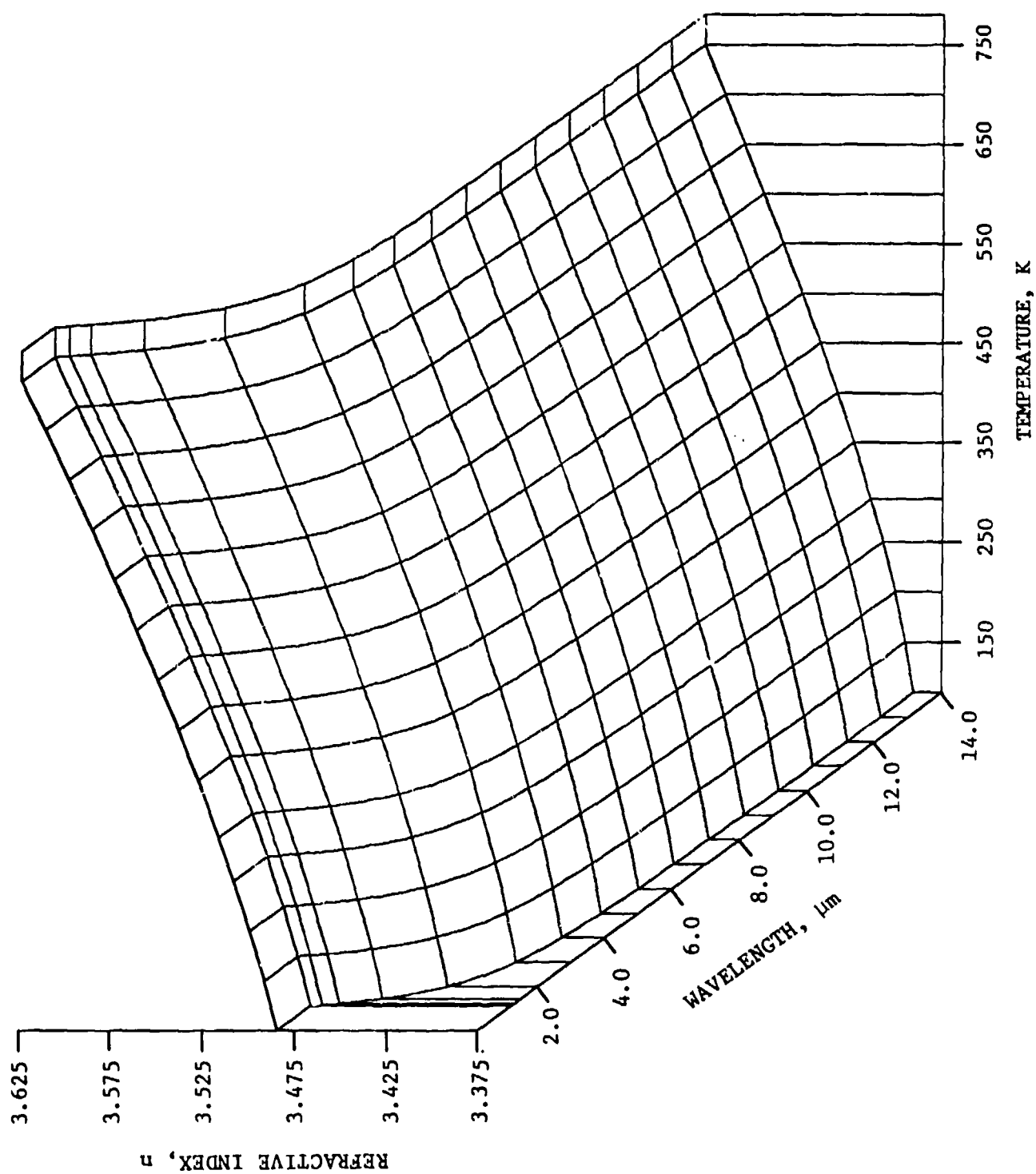
A large amount of data on the absorption coefficient of silicon and germanium are compiled and organized. Shown in Figures 10 and 11 are collective plots of all compiled data for silicon and germanium, respectively. In each of the plots there are about 5000 data points obtained from various data sources. Similarity of these two plots evidences that the absorption mechanisms in silicon and germanium are fundamentally the same as indicated in the plots. The absorption can be classified and discussed in terms of three origins, namely the electron excitation, the carrier absorption, and the lattice absorption.

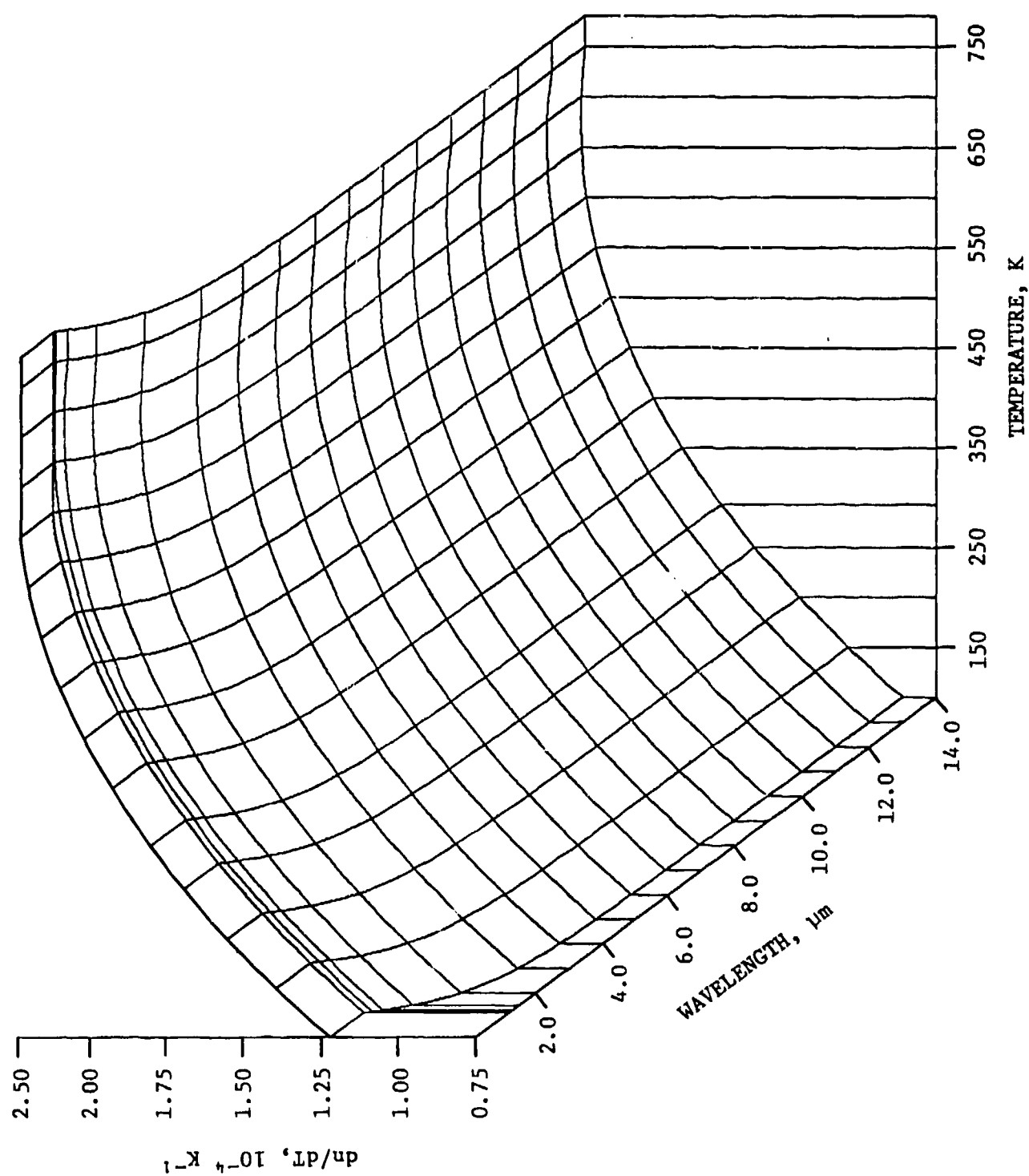
a. Electron Excitation

In the transparent region, the absorption is usually low. As the photon frequency is increased approaching the high frequency edge, a threshold point is reached where absorption is abruptly increased to several orders of magnitude within a narrow frequency range as a result of excitation of electrons from valence band to conduction band across the energy gap. Although the threshold of continuous optical absorption facilitates a means for the determination of energy gap, difficulties are encountered because the process of such excitation in silicon and germanium is indirect; the participation of phonons in the indirect process must be considered. A considerable amount of work was dedicated in this particular problem for the determination of accurate energy gap as a function of temperature and pressure.

b. Carrier Absorption

In the insulators, optical absorption due to carriers is negligible. While in the metals, optical absorption due to free electrons is extremely high; moderate variation of the free electron concentration is not sensitively reflected in the change of absorption level. Only in the semiconductors, optical absorption is very much influenced by the carrier concentration. Since the

FIGURE 8. RECOMMENDED n - λ - T DIAGRAM OF SILICON

FIGURE 9. RECOMMENDED dn/dT - λ - T DIAGRAM OF SILICON

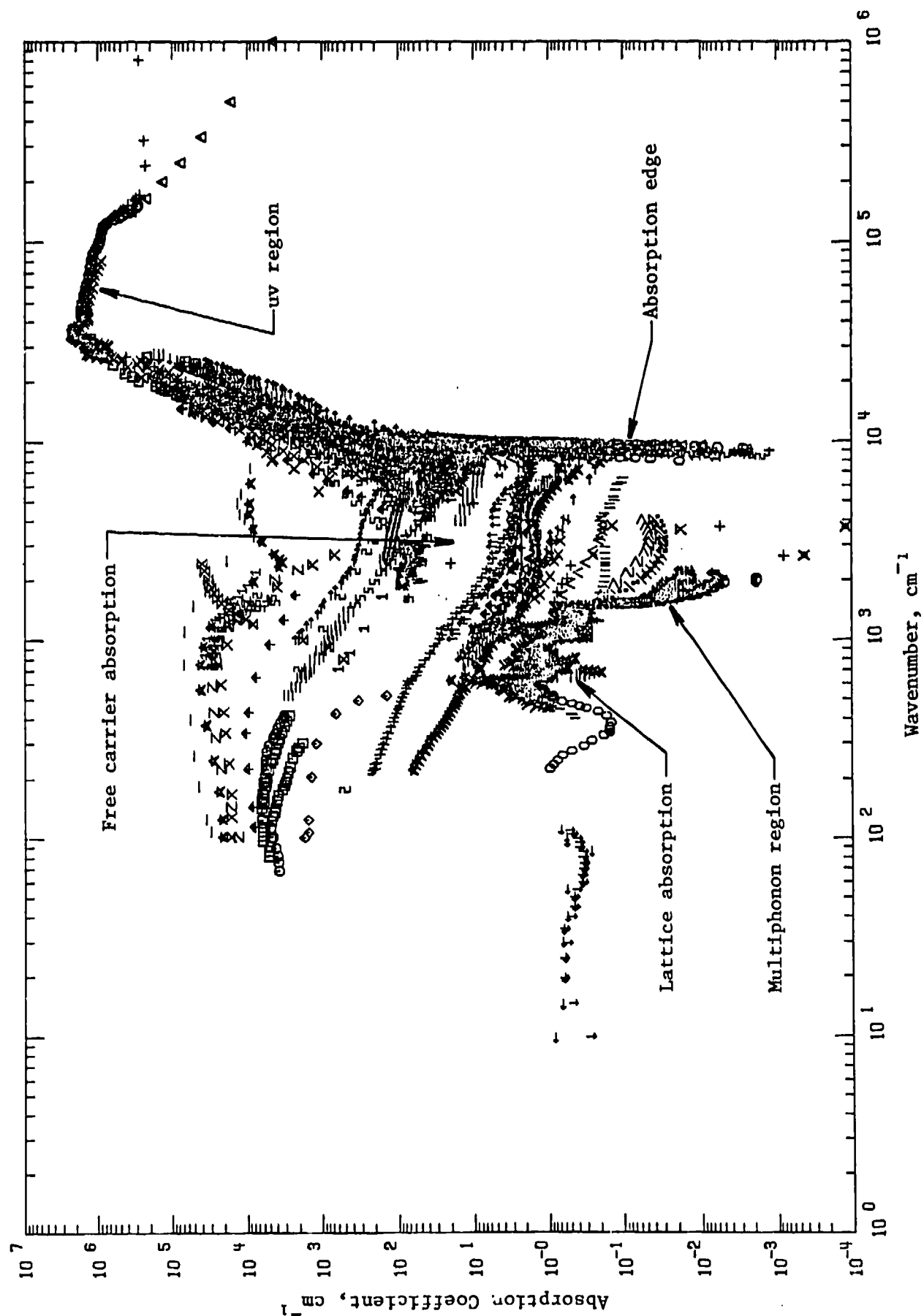


Figure 10. Absorption Coefficient of Silicon

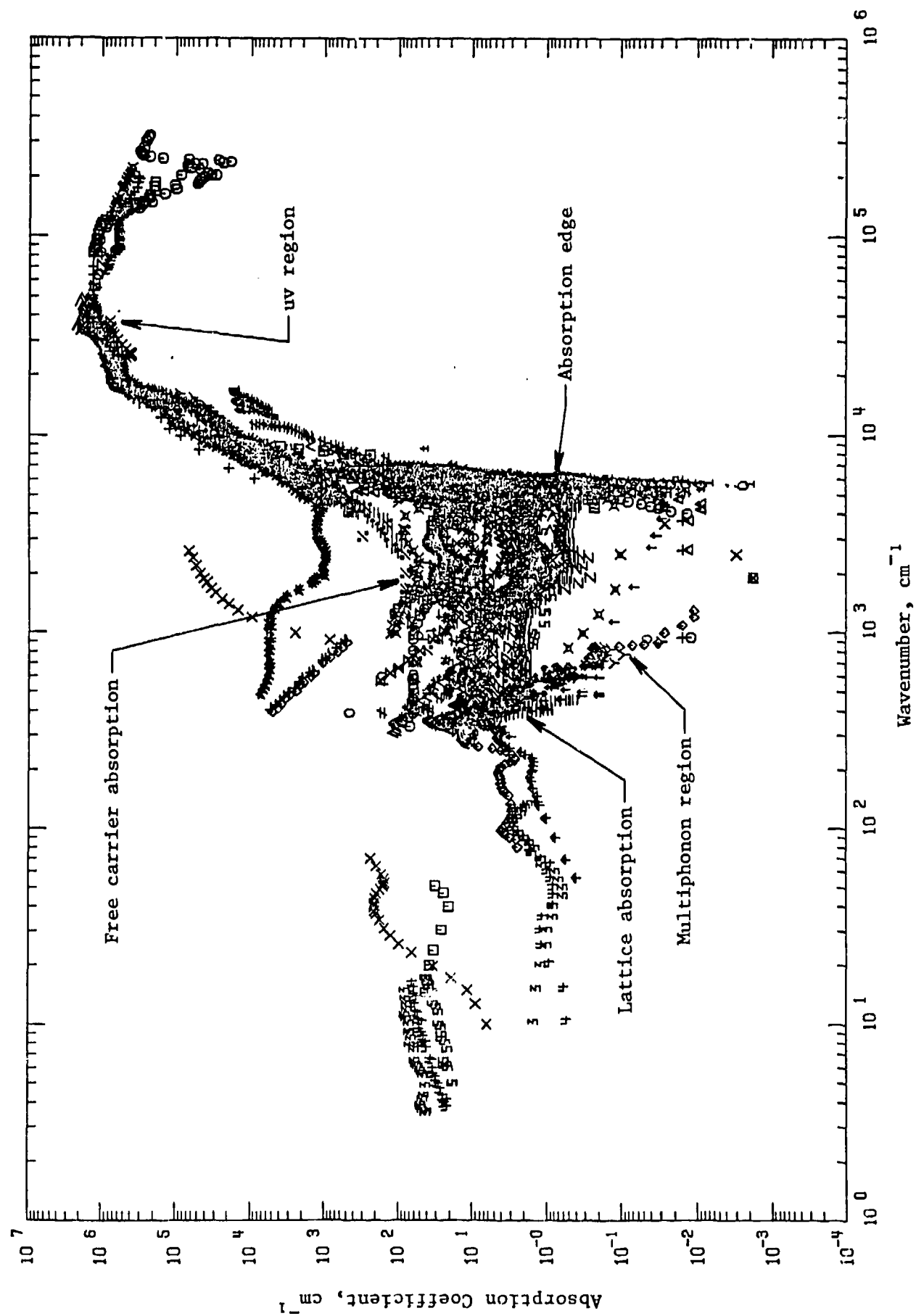


Figure 11. Absorption Coefficient of Germanium

type and concentration of carrier can be introduced in a controllable manner, the effects of carriers on the optical absorption can be studied. By use of the law of mass action, one can introduce donors so that the carriers in the sample is predominantly electrons. Absorption due to free electrons as a function of wavelength, temperature and other parameters can be differentiated. Likewise, introduction of acceptors makes sample of hole predominating; thus absorption due to holes can be observed. At high temperatures, the optical absorption of a low impurity germanium sample behaves closely to that of an intrinsic sample because the thermally excited electron-hole pairs predominate carriers from impurities. For heavily doped silicon and germanium, the carriers are essentially thermally ionized from impurity atoms. There is a temperature region called exhaustion region in which all the available impurity atoms are thermally ionized. Since the impurity concentration is far more than the carrier concentration that are thermally excited from valence band to conduction band of the host crystal, the carrier concentration remains as a constant in the exhaustion region. The existence of such a region offers a practical way to investigate the effect on the absorption of parameters other than the carrier concentration.

c. Lattice Vibration Absorption

In a pure specimen, the radiation due to lattice vibration is conspicuous at low temperatures while that due to thermally excited carriers is predominant at high temperatures. In a doped specimen, the absorption due to carriers is predominant even at low temperatures. It is therefore clear that absorption spectrum due to lattice vibration can only be observed when a highly purified specimen is used.

There have been a number of recent studies on how the infrared absorption drops off as the frequency becomes much greater than the fundamental lattice frequencies. These studies have been mainly carried out for laser window materials. In the highly purified samples, the absorption coefficient exhibits an exponential fall-off over two to four decades to the lowest values of α that can be measured. Whether this exponential absorption tail is characteristic of all other classes of materials is not known.

All the absorption mechanisms will be discussed in the technical report which is currently in preparation. The corresponding data of the mechanisms will be analyzed so that either the most probable values or the best available data for the specified conditions are recommended.

5.4. Studies on the Refractive Index of III-V Compounds

The work on the refractive index of III-V compounds is currently in the process of data extraction.

6. PUBLICATIONS

H. H. Li, 'Refractive Index of Alkali Halides and Its Wavelength and Temperature Derivatives,' J. Phys. Chem. Ref. Data, 5(2), 329-528, 1976.

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H. H. Li, 'Absorption Coefficient of Alkali Halides (Part I),' CINDAS Technical Report 54, submitted to AFOSR in March 1979.

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H. H. Li, 'The Infrared Absorption Coefficient of Alkali Halides,' International Journal of Thermophysics, 1(1), 97-134, 1980.

H. H. Li, 'Absorption Coefficient of Alkaline Earth Halides,' CINDAS Technical Report 57, submitted to AFOSR in April 1980. This report was submitted to J. Phys. Chem. Ref. Data for publication. It is now in the process of review.