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BEHAVIOR OF ALKALI HALIDES AS MATERIALS FOR OPTICAL COMPONENTS OF HIGH POWER LASERS

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Monographic Studies of Assembly

Behavior of alkali halides as materials for optical components of high power lasers*[1]

by

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1. Introduction

The development and perfecting of the construction of CO_2 lasers, of high power, operating in a system of pulses or continuous, has determined, in the recent years, a growing interest towards the properties of the transparent materials, in the field around 10.6 μm. These materials are necessary for the construction of the passive optical components of lasers (exit windows, prisms, lenses, mirrors). Together with the increase in the laser power, the power losses, which appear at the passage of the laser beam through these components, become larger and larger, thus capable to lead to their degradation, either through their destruction, or through the thermic de-focussing of the beam. Thus, there is the question of characterizing the transparent materials, from the point of view of their resistance at the passage of a laser beam with high power density, and perfecting some methods of obtaining materials with optimum values of physical

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parameters, which determine this resistance.

In the case of materials for lasers, operating under a continuous regime, the linear coefficient of absorption β, constitutes the essential parameter of the material characterization. For the lasers operating in the regime of pulses, besides the absorption coefficient, we must also take into consideration the power characteristic to the material destruction threshold P.

Besides all these parameters, in the comparison of the different materials used for the construction of the passive optical components of high power lasers, we also must take into account some other physical parameters, such as: the thermic conductibility, indicating the speed at which the heat is dissipated in the material, at the passage of the laser beam, the Young modulus, indicating the mechanical resistance of the material, the field of transmission, and the refraction index.

From table 1, which shows the values of these parameters for a series of transparent materials at 10.6 µm, it results that, one of the promising materials is KCl. This statement is also based on taking into account two additional factors: the relative facility with which we can obtain crystals of large dimensions, and the lowest cost of the materials considered.

All these arguments explain the special interest shown in the last few years by researchers from famous laboratories (Hughes Research Lab and Xonics, in the U.S.A., the Lebedev Institute of Physics, in the U.S.S.R., etc.), in the study of the processes which take place at the passage of the high power laser radiation through the KCl, as well as the efforts made for ~~the~~ obtaining crystals with the absorption coefficient closest to the ideal value.

Table 1

Physical parameters characteristic to transparent materials at 10.6 μm

Material	Diamond	Ge	CdTe	ZnSe	BaF_2	KCl
β (cm^{-1})	10^{-2}	$1.2 \cdot 10^{-3}$	$2 \cdot 10^{-3}$	$5 \cdot 10^{-3}$	$3 \cdot 10^{-5}$	$8 \cdot 10^{-5}$
Young Modulus (16^6psi)	100	15	5.3	10.3	sensitive to shocks	4.3
Transmission field (μm)	6.5 -	3-23	2-28	0.5-20	0.13 -12	0.3-20
Thermic conductibility (W/cm°C)	20	0.6	0.07	0.21	0.12	0.1
Index of refraction	2.41	4.0	2.67	2.40	1.4	1.46

In the following pages we undertake to quickly review the phenomena and the parameters characteristic to the passage of the high power laser beam through the transparent materials in the infra-red, then, describe the study materials and the results obtained on alkali halides, and especially on KCl. A great part of this paper presents original research, effectuated by authors in the last few years, part of them being published in different magazines or presented at specialized conferences.

2. Absorption of laser beam in materials

2.1. Presentation of phenomena

Where the laser beam passes through an optical component, a small fraction of the beam energy is deposited in the form of heat, in the material of the component. The heating of the laser windows through radiation absorption can lead to the destruction of the material (see following chapter), or to the beam de-focussing, through the appearance of thermic lenses (the modification of the window refraction index, from the center towards the side, due to its non-uniform heating, thus acting as a lens (1)). In this way, at high powers, only an absorption of 1% of the beam can lead to catastrophical effects. From here, it results that the critical factor which dictates the performance of a material for the passive optics of the high power laser is its linear coefficient of absorption β , corresponding to the laser wave length (2), measurement which is defined according to the well-known relation

$$\frac{I}{I_0} = (1 - R)\, e^{-\beta d}, \qquad (2.1)$$

where $\frac{I}{I_0}$ is the internal transmitancy of the crystal, and is defined as the ratio between the transmitted intensity and the incidental one,

R is the reflection coefficient of the crystal,

d is the thickness of the material crossed,

β is the absorption linear coefficient (in cm^{-1}).

Generally, the materials used in the infra-red technics, present also a residual absorption in the working domain, of two kinds: extrinsical and intrinsical (2).

The extrinsical mechanism of absorption is associated with atomic impurities or unwanted molecules (inclusions, gas bubbles, or with deviations from stoechiometry), and also network defects

(dislocations, inter-crystaline granites), as well as free surfaces which can affect the thermic equilibrium of the crystal. Since the achievement of pure crystals necessitate special conditions of preparation, the extrinsical absorption is, in general, responsible for the destruction of the components at high powers.

The intrinsical absorption is dictated by the absorption on the fundamental vibrations of the network, at the Reststrahl (ν_e) frequency, which is 6 to 8 times smaller than the frequency 943 cm^{-1} (10.6 μm), of the CO_2 laser, as well as than the electronic excitations ("Urbach Tail"). In the first case, the absorption is multiphonical, that is an absorbed photon generates two or more photons. It was established experimentally (3) and theoretically (4), that for $\nu > \nu_0$ the coefficient of multiphotonic absorption decreases almost exponentially with the increase in the frequency. If we extrapolate the measurements of the coefficients of intrinsical absorption, effectuated at low frequencies, the frequency 943 cm^{-1}, corresponding to the CO_2 laser, we obtain $\beta_{int} = 8.10^{-5}$ cm^{-1}, for KCl, and $\beta = 5.10^{-7}$ cm^{-1}, for KBr. This constitutes one of the main reasons why the alkali halides are considered among the most promising materials used in the construction of passive optical components of the CO_2 lasers. Nevertheless, the absorption, even in the best materials used at present, is dominated by impurities ($\beta > 10^{-2}$ cm^{-1}), and more recent, $\beta > 10^{-4} cm^{-1}$, in the specially prepared crystals.

The mechanism of absorption on electronic excitations of the connected electrones ("Urbach tail" (5)), appears in the materials with large "gaps", as, for example, in the case of alkali halides. In this case, the energy necessary to excite an electrone from the band of valence in the band of construction, is much higher than the frequency corresponding to the laser radiation. The contribution β_{int} to the intrinsical absorption from the electronic transitions, is obtained extrapolating, in the measurements effectuated, the excitations immediately under the band of conduction (6). These electronic transitions can ensure, together with the absorption on free carriers, the avalanche of ionization, which determines the intrinsical destruction of the transparent materials (7,8)

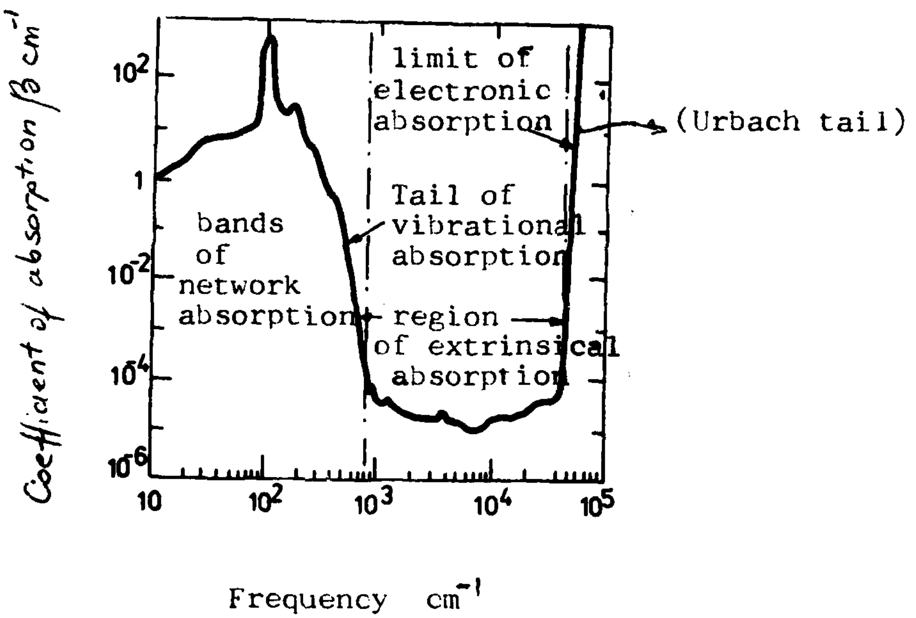

Fig 1. - Schematical spectrum of the coefficient of intrinsical absorption, function of frequency.

Figure 1 presents, schematically represented, the variation of the linear coefficient of absorption to the frequency, for a material which can be considered a good candidate for the infra-red laser optics. It is noticed that, between the absorption bands, on the fundamental vibrations of the network, and on the excitations of the connected electrones, it is drawn the curbe corresponding to the intrinsical absorption, which corresponds to $\beta < 10^{-4}$ cm^{-1}. Since fewer researches have been done in this field, the drawn shape of the curbe is, in fact, presumed (1,2,9). That is why, in the recent years, every effort was concentrated on the research of the mechanism of the extrinsical absorption, as well as on determining the values of β , at the corresponding laser frequency, in materials which can be candidates for the construction of the passive components of high power lasers.

2.2 Factors which determine the absorption

It is well known the fact that the impurities in alkali halides can introduce absorptions, both in the ultra-violet, and in the infra-red field of the probe spectrum. Most of these absorptions are associated with the oxygen, or with the anions containing oxygen, such as the ion OH^-. An important class of impurities absorbant in the infra-red is constituted by the poliatomic anions, which substitute the halide ions in the crystaline networks.

The monoatomic impurities can also absorb the infra-red radiation, both through the local modes of variation, and through the activated photons, other than the fundamental Reststrahl photons (10). However, the absorption due to the monoatomical impurities at 943 cm^{-1} is negligible, because both the phononic frequencies, and the local vibrational modes are much smaller than 943 cm^{-1}.

The infra-red absorption spectrum of a crystal which contains substitutional anionic impurities, consists in a number of narrow lines, which correspond to the internal vibration modes of the molecular poliatomic ions. In general, the internal vibrations of the ion impurities are relatively little affected by the crystal host. There are small shiftings in frequency, and increases in the width of the lines, which depend on the network. Due to a low simmetry of the ion at the interaction with the host network, the inactive modes in the infra-red can become weak absorbant, and the degenerated modes can be split in several components.

The frquencies measured for several ionic - molecular impurities in KCl crystals, are summarized in the table 2 (11-31).

Table 2

Absorption frequencies observed experimentally for several anionic - molecular impurities, in KCl crystals and concentrations estimated to produce an absorption coefficient $\beta = 10^{-4}$ cm^{-1} at the frequency 943 cm^{-1} of the CO_2 laser.

Ion	Frequency of absorption(cm^{-1})	Concentration in ppm for β (943cm^{-1}) $=10^{-4}$ cm^{-1}	Bibliography
SO_4^{-2}	630, 978, 1083, 1149, 1188	0,03	[11, 12, 13, 14]
CO_3^{-2}	680, 720, 883, 886, 1058, 1064, 1378, 1416, 1488, 1518	10	[15, 16, 14]
NO_3^-	842, 1062, 1396	10	[18, 19, 14]
NO_2^-	305, 1290, 1329	0,1	[20, 21, 22]
NCO_3^-	589, 672, 713, 850, 971, 1218, 1346, 1701, 3339	0,01	[23]
OH^-	3640	100	[24, 25]
NH_4^-	1405, 3100	10	[26]
CrO_4^{-2}	862, 870, 930, 941	0,1	[27, 28]
SeO_4^{-2}	834, 860, 909, 923	0,1	[20]
MnO_4^{-2}	899, 909, 914, 925	0,1	[31]

In the 3rd column, there are aligned the concentrations in ppm obtained through the extrapolation of the absorption lines, using the Lorentzian formulae of the absorption line, that is

$$\beta(\nu) = \frac{\text{const.}\,\Delta\nu}{(\nu-\nu_0)^2 + \left(\frac{1}{2}\Delta\nu\right)^2}, \qquad (2.2)$$

where $\beta(\nu)$ is the coefficient of absorption at frequency ν.

ν_0 is the freqency centered on the absorption line,

$\Delta\nu$ is the width of the line, at half its height.

For a concentration of the impurities of 0.1%, the coefficient of absorption at the center of a line of absorption $\beta(\nu_0)$ is comprised between 10^{2} and 10^{3} cm^{-1}, with $\Delta\nu$ varying from 4 to 50 cm^{-1}, variation which depends on the molecular species and the network.

The table presented above, though it summarized only those absorbant impurities which had been studied in the KCl crystals, it serves to illusrate the importance of the anionic molecular impurities in the radiation transmission at 943 cm^{-1} (10.6 µm). In connection with the anionic impurities which could be researched, in this sense we point out the SiO_4^{-4} ion, which has a powerful absorption at 1,000 cm^{-1} in glasses (32).

Also, besides the substitutional anionic impurities, there are also the molecular impurities, which could limit the laser transmission at 943 cm^{-1}. For example, the neutral molecules, such as H_2O or CO_2 can exist in the network interstice, in the crystal volume, or condensed, on the surface. Also, on the surface, some other organic molecules can be condensed, molecules which absorb at a frequency of 1020 cm^{-1}. The KCl crystals, doped with Pb^{2+} do not give absorption at the frequency of 943 cm^{-1}, but the Pb^{2+} complexes with the bivalent anions have absorptions in this place, in particular $PbCO_3$.

If we dope KCl with OH^- or CO_3^{2-} , we notice a small effect in the absorption at 943 cm^{-1}, when they are introduced separately, but when they are presented together, we notice a significant increase in the absorption, due to the posibility of the formation of the ion HCO_3^- , which absorbs strongly (33).

2.3. Experimental methods for measuring the linear absorption coefficient

As it results from the previously presented material, the crystals with a small coefficient of absorption, can be used in the laser technics for the construction of passive optical components.

The measurement of the small coefficient of absorption in solids, can be done through different technics:

- specroscopy with a double beam (34),
- calorimetry (35 - 42),
- spectral emission (43, 44).

In general, through the last two methods, we obtain a value closer to the real one, if we separate the absorption in volume, from the one on the surface, the quality of the crystal surfaces being determined in the process of absorption in the infra-red. This separation can be done through absorption measurements, in function of the crystal length. Obtaining a linear relation between the absorption and the length of the crystal, the coefficient of absorption in volume can be deducted from the gradient of the straight line, and the absorption on surface , by extrapolating the zero length of the probe. The good quality of the crystal surface, in the sense of obtaining a smaller absorption at surface, cannot be obtained only through the technique of abrasive polishing, which leads to the appearance of warm spots, through the local absorption of radiation, and scretches, which serve as spots where humidity attacks, or through the simple cleavage.

The spectroscopic method with double beam, in most cases, resorts to extrapolation, due to its low sensitivity. Generally, the linear coefficient of absorption can be measured spectroscopically, up to values of $\beta = 5 \cdot 10^{-3}$ cm^{-1}.

It is known that, the transmission of a material of a length L, with a coefficient of reflection R, a coefficient of absorption β , and the index of refraction n, is given by

$$T = \frac{(1 - R)^2 e^{\beta L}}{1 - R^2 e^{-2\beta L}}. \qquad (2.3)$$

where $R = \left(\frac{n - 1}{n + 1} \right)^2$.

This ecuation shows that, the detection of a small loss in volume, through conventional methods of transmission, implies the use of thick probes. Additionally, that the index of refraction n is not well known, in the region of the wave length of interest, the precision with which can be measured the coefficient of absorption, will be limited by the lack of knowledge of the losses through reflection at surface. Using the differential technique, in order to eliminate the effect of the losses through reflections at surface, the losses in volume can be measured directly. The measurements are done with the aid of a spectrophonometer with double beam. Thus, a thin probe of length L_{ref}, and transmission T_{ref}, is introduced in the beam of reference, and the thick probe which measures β, of length L_{probe}, and transmission T_{probe}, in the probe beam, we obtain the value T_{meas}, equal to

$$T_{ref} \times T_{meas} = T_{probe} \qquad (2.4)$$

from here

$$T_{meas} = e^{-\beta \Delta L} \frac{1 - Re^{-2\beta L_{ref}}}{1 - R^{2} e^{-2\beta L_{probe}}}, \qquad (2.5)$$

where

$$\Delta L = L_{probe} - L_{ref} .$$

For materials with small coefficients of reflection R, as in the case of alkali halides, $T_{meas} = e^{-\beta \Delta L}$. If $\beta \Delta L \leq 1$, the exponentials can be developed in a series, obtaining

$$\beta = \frac{1 - T_{meas}}{\Delta L} \cdot \frac{1 - R^{2}}{1 + R^{2}} \qquad (2.6)$$

With the purpose of detecting the impurities which could influence the absorption in the infra-red and ultra-violet fields, there were effectuated measurements on a series of KCl probes, which in the following will be noted thus:

1. - KCl crystals, previously grown at IFA,
2. - KCl crystals imported from the U.S.S.R.,
3. - KCl crystals imported from the Harshaw Co., U.S.A.,
4. - KCl crystals grown by the authors.

Using the spectrophonometry with double beam, we determined the level of impurities, as follows:

- From the position and the shape of the absorption lines, we identified the types of impurities present in the crystal.

- We estimated the value of the coefficient of absorption through the extrapolation of the tails of the absorption lines, using the Lorentzian shape of the absorption lines, and the relation (26).

Table 3 presents in a concentrated form, the results of these measurements.

Since we saw that the specrophonometric method with double beam is limited to values of $\beta > 10^{-3}$ cm^{-1}, for the determination of the coefficient of absorption in the interval values $10^{-5} < \beta < 10^{-3}$ cm^{-1}, the calorimetric method is used (36 - 43). In such an experiment, we use as source of radiation of 943 cm^{-1} (10.6 μ m), a CO_2 laser operating in a continuous regime, and the intensity absorbed in the crystal, following the irradiation. It goes without saying that, such an experiment can succeed only in the case when the crystal is well insullated thermically towards the exterior.

In this case, we start from the calorimetric relation

$$I_0 = I_R + I_T + P_a + P_p, \qquad (2.7)$$

where I_0 is the intensity of the incidental beam,

I_R is the intensity of the reflected beam,

I_T is the intensity of the beam transmitted, and has the significance from the relation (2.3),

P_a is the power absorbed in the material studied,

P_p is the power loses in the experiment chain.

If the absorptions are small ($10^{-5} < \beta < 10^{-3}$ cm^{-1}), P_a can become comparable to P_p , so that it is neccessary to measure separately P_a. P_p will enter the realm of errors of systematic measure of I_o and I_T. Thus, using the relation $P = I_0 - I_R - I_T$ obtained from the relation (2.7), with the above specifications, where P is the power absorbed effectively, and for I_o and I_R and I_T , relations well-known from the passage of light through a plan parallel blade, we obtain

$$P = I_T \frac{(1 + Re^{-\beta L})(1-e^{-\beta L})}{(1-R)e^{-\beta L}}, \qquad (2.8)$$

where the measurements have the significances known, and L is the thickness of the probe. In the case of small absorptions, $\beta L \ll$ L, the exponentials can be also developed in series. Introducing also the measurement $Q = \frac{P}{I_T}$, (the ratio between the intensity absorbed effectively, and the intensity transmitted), we obtain:

$$\beta = \frac{1}{L} Q \frac{1-R}{1+R}. \qquad (2.9)$$

For materials with a small coefficient of reflection, as in the case of alkali halides,

$$\beta = \frac{Q(1-2R)}{L} = \frac{P(1-2R)}{I_T \,.\, L}. \qquad (2.10)$$

Formulae (2.10) is used for the determination of the linear coefficient of absorption. For this, we must measure P and I_r. The experimental assembly comprises a CO_2 lase, operating a continuous regime, the power being generally between 40 and 70 W. We measure the heating of the crystal following the irradiation in a certain time interval. We determine thus the intensity absorbed effectively, according to the calorimetric ecuation

$$P = \frac{m\ c\ \Delta T}{t} \qquad (2.11)$$

where m is the crystal mass,
c is the caloric capacity of the material,
t is the time of irradiation,
ΔT is the growth of the crystal temperature.

This measurement can be determined in different ways: either with the aid of a thermocouple mounted in crystal, taking care that the laser beam does not fall directly on it, or, as we proceeded, with the help of a thermometer with resistance, which consists in a coil of very thin copper wire, placed monolayer on the edge of the crystal, in order to have a good thermic contact with the crystal. Thus, we measure the variation of the resistance of the copper wire, while the temperature of the crystal increases. The time t, the duration of the irradiation, is calculated using the ecuation of the heat conduction, presuming that the beam diameter is much smaller than the crystal diameter (condition which is also achieved experimentally). This time must be bigger than the time of propagation of the thermic wave from the center to the periphery of the crystal, and must be close to the time characteristic to the linear growth in the probe temperature, thus being able to eliminate the errors in the determination of the crystal heating. Obviously, during all these experiments, we must ensure a good thermic insulation of the probe from the exterior.
The experimental results we achieved, without extracting the absorption at surface, are presented in the table 3. From here, it can be noticed that real conclusive results are obtained if the volume absorption is separated from that of surface. Thus, crystals 1

Table 3.

Determination of the coefficient of linear absorption β using various methods and the destruction threshold for KCl crystals of different origins.

Nr. crt.	Tipul de cristale 1)	Determinări spectrofotometrice 2)		Determinări calorimetrice 5)	determinări de prag de distrugere în volum 6)	OBS
		Tipul de impurităţi 3)	β (cm^{-1}) calculat la 943 cm^{-1} 4)	β (cm^{-1}) determinat la 943 cm^{-1} folosind un laser CO_2 în continuu (60 W) 7)	P(W/cm^2)	
1	Cristale KCl crescute la IFA 9)	OH^-(UV) NO_2^- ($\nu = 1393$) NO_2 ($\nu = 1283$) SO_4^{-2} ($\nu = 1888$) ($\nu = 1150$) ($\nu = 1083$)	 $2.72 \cdot 10^{-3}$ $6 \cdot 10^{-3}$ $4,5 \cdot 10^{-3}$ $4,5 \cdot 10^{-3}$ $2,9 \cdot 10^{-3}$	$1,85 \cdot 10^{-2}$ (pe cristal clivat proaspăt) 8)	$1,1 \cdot 10^{7}$ (pe cristal clivat proaspăt). 8)	
2	Cristale KCl import din URSS 10)	nu s-au măsurat 12)		$1,6 \cdot 10^{-2}$ 15) (pe fereastră şlefuită)	$1,85 \cdot 10^{8}$ 8) (pe cristal clivat proaspăt)	
3	Cristale KCl import de la Harshaw 14)	Nu s-au identificat 16)	–	Nu s-au măsurat 16) $\approx 4 \cdot 10^{-3}$ în literatură [43] 17)	Nu s-a măsurat 16)	
4	Cristale KCl crescute de autori 11)	Nu s-au identificat 13)	–	$6 \cdot 10^{-3}$ 8) (pe cristal clivat proaspăt	$6,12 \cdot 10^{8}$ 8) (pe cristal proaspăt clivat.)	Cristale 18) crescute în condiţii speciale

Key: 1-type of crystal; 2- spectrophotometric determinations; 3- type of impurities; 4- calculated at; 5- calorimetric determinations; 6- destruction threshold determinations in volume; 7- determined at 943 cm^{-1} usina a CO_2; direct current laser (60 W); 8- for crystals newly split; 9- KCl crystals grown at IFA, 10- KCl crystals imported from USSR; 11- KCl crystals grown by the authors; 12- not measured; 13- not identified; 14- KCl crystals imported from Harshaw; 15- for polished face; 16- not measured; 17- in literature [43]; 18- crystals grown under special conditions and 2 present values of β , close enough, altough crystal 2 was grown under special conditions, required by its destination. It is nevertheless, noticed that the value of its destruction threshold is one order size larger than that of the crystal 1, but crystal 2 was a polished window, a little sweeped, which surfaces have introduced a strong absorption, while the quality of the freshly cleaved crystal surfaces is very good.

We laso tried an assembly to determine the linear coefficient of absorption, using as a source the CO_2 lase of high power, in pulse (45). In this case, β must be known, due to very different conditions under which it is achieved the absorption in the crystal, relative to the power density on the crystal; if in the continuous regime the power density is in the order of 10^2 - 10^3 W/cm^2, in pulse it is over 10^8 W/cm^2. As a preliminary result, we obtained for the crystal 1, a value of the linear coefficient of absorption approx. 5 times higher than in the case of the measurements effectuated using the continuous laser as a source. The elucidation of the behavior, presumed to be due to the appearance of some non-linear phenomena of densities of high power, neccessitates an accumulation of several absorption data, on several types of crystals, and under the conditions of a more careful experiment.

A variation of the calorimetric method is constituted by the barothermic method (46). In this case, the crystal heating, following the radiation absorption, is determined by measuring the growth in the pressure of a non-absorbant gas, which is placed in the same closed space with the probe. This experiment presents the advantage that it does not pose the problem of finding a spot to place the thermocouples.

Another method of measuremnt of the coefficient of absorption, is the method of spectral emission (43, 44). Theoretically, it consists in the determination of the probe spectral radiation, at a known and controllable temperature, relative to the radiation of the black body, situated at a known temperature. The difference between the probe radiation and that of the black body, is a measure of the probe spectral emission, and thus, of the absorption. Under special experimental conditions, (44), we can measure the emission (and thus, the absorption) with an approximation smaller than 6.10^{-9}, in relative units.

3. Damage threshold of material at laser pulses

3.1. Presentation of the phenomena

It is well known that the materials present a limit of the optical resistance to laser pulses of high power (47, 7-9). Thus, when the laser power is below a certain value of the threshold, the beam penetrates the transparent material, apparently without affecting it.

But, if the laser power exceeds this threshold value, the beam is absorbed, and effects appear, such as the expulsion of the material on the surface of the probe, and the formation of craters, the destruction of the interior of the probe, melting, vaporization, breaking. Under these conditions, the transmission of the beam through the crystal decreases very much.

The destruction of the high power components can be due to many causes:

1) The preferential absorption on the impurities (2), inclusions, pores, or cracks (7, 8, 48, 49), present in the material before the irradiation, producing a powerful local heat, can lead to melting and vaporization. The pressure created by the vaporized material produces shock waves, thus, the mechanical tension reaches to exceed the resistance of the material, and the latter one cracks . (47, 50-53) We can define (9) a measure F to describe the thermic destruction of a component, through the relation:

$$F = \frac{K}{\beta} (T_{destruction} - 300), \qquad (3.1)$$

Where K is the thermic conductivity,

β is the linear coefficient of absorption,

$T_{destruction}$ is the temperature of destruction, in Kelvin degrees.

Since alkali halides have small thermic conductivities, and a relatively small mechanical resistance, thermic destructions will be supposed, in the case when they are not sufficiently pure. For KCl, the temperature of destruction is 1074 K, and F = 549000 w (9).

2) Even if the materials are very pure and do not present other types of defects, there is a certain limit to which they can resist to high powers. This limit of intrinsical destruction of the crystals is dictated by the avalanche of ionization (47, 7-9, 49, 54-57). Thus, if the laser beam is focussed to the interior of a transparent material, the free electrons are accelerated in the intense electric field, associated with the radiation, sufficient to produce other electrones through successive collisions of ionization. If the rate of the electron production exceeds their loss through diffusion, recombination or collection, the avalanche takes place, which leads to a powerfully absorbent plasma. When this plasma is sufficiently dense, the losses of Joules heat through collisions in the electronic gas become so great, that there appears melting and vaporization of the material, in the respective region. Around this zone, the material is continuously destroyed by the shock waves produced by the thermic-generated tensions.

It was shown that in the materials with a large "gap", such as alkali halides, there is always an initial concentration of free electrons ($N \gtrsim 10^{8}$ electrones/cm^3), originating from the multiphonic absorption, interband absorption, or from the ionization of the donnor levels. These electrons, following the collision with longitudinal optical photons (polaronic coupling), they have a component of the speed in phase with the direction of the electric field, so that there can be a medium energy absorption from the electric field. In this way, the time between the two collisions is also very short ($\tau \gtrsim 10^{-15}$ s), much shorter than the duration of a laser pulse ($t \gtrsim 10^{-7}$ s). This is otherwise a condition for the formation of the avalanche.

In the paper (55), it is shown that the energy absorption rate from the electric field by an electron, is given by the relation

$$\frac{dW}{dt} = \frac{e^2}{m(1+\omega^2\tau^2)}, \qquad (3.2)$$

where e and m are respectively the charge and the mass of the electrone,

ω is the frequency of the laser radiation,

τ is the time between the collisions, electron-longitudinal optic phonon ($\tau \approx 10^{-15}$ s, in the majority of alkali halides, and $\omega\tau < 1$, for the frequency of the CO_2 laser).

On the other hand, as it was shown in the paper (8), the growth rate of the density of N electrons, at the time of the formation of the avalanche, is given by the relation

$$\frac{\partial N}{\partial t} = \eta(E)N - \left(\frac{\partial N}{\partial t}\right)_{\text{losses}} \qquad (3.3)$$

where η (E) is the probability on the time unit in order that an electron produces an ionized collision, and in function of the intensity of the electric field E(t); $N(t=0) \neq 0$, which means that the first free electrons must be created through another mechanism. The last expression represents the loss of electrons through diffusion, outside the region where the discharge takes place, through the recombination or the collection, at the imperfections of the crystalline network. The destruction through an avalanche can take place when $N > 10^{18}$ electrones/cm^3, thus, there is a growth, with a factor of 10^{10}, taking into account that the initial density of the electrons is approximately 10^8 electrons/cm^3.

Thus, the ionization avalanche limits the maximum density of the power flux, which the solid can support before its transformation into a hot plasma.

As it results from the relation (3.2), $N(t)$ grows as some electrons are accelerated to a higher energy than the "gap" energy. These electrons can create other free electrons through ionized collisions, but the probability on electron and the time unit of producing an ionization is small. Even at an initial density of 10^8 electrons/cm^3, since the focal volume where the avalanche is produced is small (approximately 10^{-5} cm^3), there will be few free carriers. This fact justifies the static fluctuations observed at the appearance of the destruction (58). Thus, there is a so-called chance that an electron reaches the energy neccessary to produce other ionizations.

As it was shown in the paper (59), the probability that the destruction appears after n consecutive pulses of a certain power, is given by

$$P_n = (1 - P_1)^{n-1} P_1, \tag{3.4}$$

where P_1 is the probability that the destruction appears from a single pulse, and it is approximately given by the relation

$$P_1 \approx \exp\left(-\frac{K}{E}\right), \tag{3.5}$$

where K is a determinant constant of the specific properties of the material, and the experimental conditions, and E is the electric field associated with the luminuous wave.

At smaller powers than the destruction power threshold, there appears a distortion of the thermic-induced beam (1, 60). Thus, the laser beam, having the maximum density in the center, creates a gradient of temperature in the component, from the center towards its edge, which determines a corresponding variation of the component refraction index. Consequently, the window will act as a lens, distorting the laser beam. The same optical distortion can appear due to the variation in the pressure exercized by the beam, which can lead to the curbing of the component (61).

It is noticed that, in general, the destruction threshold of the surfaces is smaller than that of the volume destruction (7, 9, 48, 62-65). The first cause of this phenomenon is the fact that the surfaces of the components are scratched, or may contain impurities or pores, due to polarization or wear.(48) In general, the dimensions of these defects are smaller than the wave length, since the surface of the components has been specially prepared to achieve optic qualities.

Around the edges of these imperfections, there appears a concentration of the electric field. On the other hand, there is also a tendency of the impurities to concentrate around the cracks, so that in the respective zones it increases the radiation absorption. Thus, the destruction threshold on the surface is 2 to 5 times smaller than that of the volume destruction.

Consequently, it is necessary to obtain cleaner sufaces. It was noticed (66), that throuth their bombardment with Argon ions, followed by the deposit in vacuum of a protecting thin layer, the surface destruction threshold can be considerably improved.

In this case, the surface can also be destroyed due to the formation of a dense plasma, following the appearance of the ionization avalanche.

In general, surface destructions have the shape of craters (fig. 2), out of which the material was expulsed. The cracks appear along the crystalo-graphic directions $\langle 100 \rangle$.

On the other hand, it was noticed that the destruction threshold on the entry surface of the laser beam is greater than the one on the exit surface. In the paper (65), it was suggested that this phenomenon is due to the Fresnel reflections, which appear at the propagation of light through a dialecric. Thus, at the normal incidence, a part of the light is transmitted, and a part is reflected, as it results from the Fresnel relations. At the entrance surface, the reflected wave is dephased with 180°, compared to the incidental one, and the transmitted wave is linked to the incidental one, through the relation:

$$I_{entrance} = \frac{4}{(n+1)^2} I_0 , \qquad (3.6)$$

where n is the refraction index of the dielectric medium. At the exit surface, the reflected wave is no longer dephased, so that the intensity of light on the exit surface is linked to the incidental intensity, through the relation

$$I_{exit} = \left(\frac{4n}{(n+1)^2} \right)^2 I_0 \qquad (3.7)$$

Thus, the ratio between the light intensity of the interior of the medium, at the exit surface, and the light intensity of the interior of the entrance surface, is

$$\frac{I_{exit}}{I_{entrance}} = \left(\frac{2n}{(n+1)} \right)^2 , \qquad (3.8)$$

For $n_{KCl} = 1.46$, $\frac{I_{exit}}{I_{entrance}} = 1.3$

Fig.2 - Aspect of the destruction caused by the beam laser (E=60mJ) on the surface of a KCl crystal.

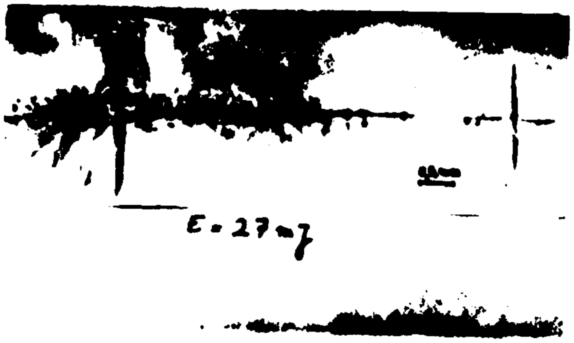

Fig.3 - Aspect of the destruction caused by the laser beam (E=27mJ) inside a KCl crystal, which contains unwanted impurities.

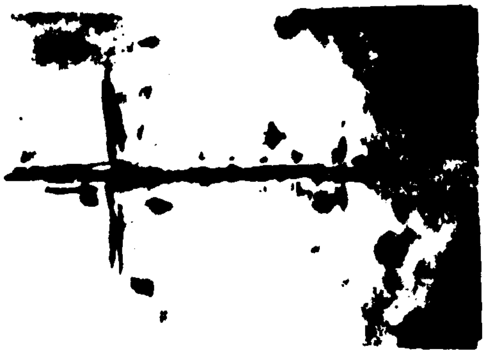

Fig.4 - Aspect of the destruction caused by the laser beam (E=176mJ), inside a KCl crystal, grown under special conditions, in order to avoid the unwanted impurities.

Thus, as a consequence of the Fresnel reflection, the plasma formed on the entrance surface, reflects the beam, thus determining a growth in the value of the destruction threshold.

3.2. Microscopic studies of damages and determination of the damage threshold

The microscopic study of the destruction in alkali halides crystals is important both from the point of view of determining the resistance to high power laser radiation, and that of the mechanism responsible for the production of these destructions. Thus, several groups of authors established (8, 35, 49, 67-71), that, irrespective of the type of material used, the destruction threshold represents the power of the radiation which leaves in the probe a trace of destruction, visible under the optical microscope. On the other hand, by studying under the microscope the shape of the destruction, for alkali halides, a distinction can be made between the destructions caused by the presence of the inclusions in the material, and the intrinsical ones (8, 49, 56). As expected, the first ones will appear at a smaller power.

We have achieved the irradiation of crystals under experimental conditions, which were presented in the precedent papers (72, 73). We must mention though, that in order to obtain reproductive values of the destruction power threshold of the crystals, several conditions (7, 8, 35) must be met simultenuously:

a) Experiments must be done with pulse CO_2 lasers which operate in the fundamental TEM_{00} mode. Only in such a way can we ensure the uniform distribution of energy on the beam surface, and avoid the appearance of the hot spots, where the energy is several times greater. It was also noticed that the destruction thresholds, determined with the multimodal beams, on the same types of crystals, were approximately 4 times smaller than the ones obtained by using the TEM_{00} mode (7).

b) In the second place, the experimental arrangement must be such, as to eliminate the possibility of the appearance of the self-focussing phenomenon. In the opposite case, the destruction will appear at a density of smaller power than the real value of the threshold. Self-focussing appears in the dielectric mediums, since the refraction index presents a weak dependence on the intensity transmitted. This dependence is described by the relation

$$n = n_0 + n_2 |E|^2. \qquad (3.9)$$

where $|E|^2$ is the intensity of the elctric field, associated with the luminuous beam.

n_0 is the value of the medium refraction index, in the absence of self-focussing,

$n_2 > 0$, thus at a larger electric field, the dielectric medium acts in the sense of the growth of the energy of interaction. One of the parameters which characterizes self-focussing is the critical power for self-focussing, and it represents the power at which the collimated beam with the Gaussian profile, is self-focussed in compensation with its dispersion, due to the difraction. This value is approximately 10^8 W, for alkali halides, in the case of the CO_2 laser (λ = 10.6 μm).

In order to reach the power density which causes the destruction, without producing self-focussing, we work at smaller powers ($\simeq 10^6$ W), but focussed on the probe with the aid of lenses (8).

c) The third condition to be fulfilled in order to obtain experimentally an exact value of the power of the destruction threshold, is to determine, as accurately as possible, the diameter of the laser beam, in the lens focus, in order to have a real value of the destruction threshold density (35). In our experiments, we used the method described in (74). In the optical microscope, with the aid of a micrometric scale, attached to the ocular, we measured the dimensions of the traces that the laser beam of different energies leaves on a developed film, placed in the lens focus. Thus, it was found that the area of the focal spot, corresponding to the small energies of the laser, thus values close to the threshold ones, is very close to the value calculated according to the formula:

$$A = \frac{\pi d^2}{4}$$

where $d = \theta f$, θ is the beam divergence, and f - the lens focal distance. In our experiments, the diameter of the focal spot was approximately $10\lambda \approx 10^{-2}\,\text{cm}^{-1}$.

The values of the density of the destruction power threshold, determined by us on KCl crystals, obtained from different sources, are given in the table 3. The measurements were done on a single pulse. When the power density reaches the threshold value, a sparkle appears, due to the appearance of plasma, which produces the destruction. If we compare (table 3) the measured values of the absorption coefficient β with those of the destruction threshold P, for the different KCl crystals, we notice , as expected, that at the crystals of high purity, which have a small β value, the destruction threshold is big.

We stated at the begining of this chapter that the microscopic study of the general aspect of the destructions caused inside the crystals by the laser beam, can give information about the mechanism according to which the destruction was produced. As examples, we will use two of the probes: the crystal 1 which presents impurities, with absorption bands around the frequency 943 cm^{-1} (10.6 μm), (see table 3), and the crystal 4, where the lowest value of the linear coefficient of absorption was measured.

In figure 3, taken from the crystal 1, we see that the destruction is formed of small spherical zones, from where the microclivages start.

Such a destruction is of thermic nature. The spherical zones are micromeltings in the places where, due to the presence of impurities, the beam is preferentially absorbed (8, 73). The microclivages are due to the pressure excercized by the shock wave, generated thermically.

In the figure 4, which represents a destruction characteristic to a purer crystal (crystal 4), the destruction is cross-shaped (8), and is due mainly to the ionization avalanche, previously discussed.

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