

Light Scattering in Alkali Halide Single Crystals

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Light scattering by crystals of potassium chloride obtained from several commercial suppliers was measured. All of the experimental observations can be explained by postulating that the scattering comes from dislocations which are surrounded by scattering centers having an anisotropic polarizability ellipsoid. The effective length of the scattering units is calculated from theory and experiment to be about 3×10^{-4} cm, and the effective radius is less than 2.5×10^{-6} cm. Also, no correlation between the scattering power of a crystal and the growth rates of either the *F* band or *M* band under ionizing radiation was found. This suggests that the bad regions associated with the light scattering do not affect the coloration properties of a crystal.

INTRODUCTION

MUCH can be learned about certain types of lattice imperfections in transparent solids from observations of light scattering. From the intensity and wavelength dependence of the scattering, estimates of the size, shape, and concentration of the scattering units within the material may be made. In general, the problem is complicated by the presence of a variety of scattering units. This is particularly true for alkali halides. For example, the possible scatterers in KCl single crystals are precipitates, vacancy-impurity clouds around dislocation lines, grain boundaries, cavities, etc.¹⁻⁵ Therefore, in these crystals the evaluation of the scattering data may be somewhat more difficult than for solids containing only one type of scattering unit. However, from careful experiments and a comparison of all the available experimental observations with theory it is possible in principle to identify the type or types of scattering units which are dominant as well as their effective sizes, shapes, and concentrations.

The present work has a multiple purpose: (1) to determine (by measurements of the wavelength dependence, angular dependence, crystal orientation dependence, and the depolarization ratios of the light scattering) the character of the dominant scattering units within the crystals; (2) to find out what defects compose the scattering centers within each scattering unit, e.g., vacancies around an edge dislocation would form a cylindrically-shaped scattering unit with vacancies as the scattering centers; and (3) to investigate the extent to which the "bad regions" (which are the scattering units) contribute to the formation of defects under ionizing radiation.

EXPERIMENTAL PROCEDURE

The crystal ingots used in this research were purchased from the Harshaw Chemical Company, the

* Oak Ridge National Laboratory is operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

¹ S. P. F. Humphreys-Owen, Proc. Phys. Soc. (London) **B68**, 325 (1955).

² K. G. Bansigir and E. E. Schneider, J. Appl. Phys. **33**, 383 (1962).

³ O. Theimer, C. A. Plint, and W. A. Sibley, Ann. Phys. (N. Y.) **9**, 475 (1960).

⁴ O. Theimer, and C. A. Plint, Ann. Phys. (N. Y.) **3**, 408 (1958).

⁵ C. A. Plint and W. A. Sibley, J. Appl. Phys. **33**, 3167 (1962).

Isomet Corporation, and the Optovac Corporation. In the case of the angular dependence investigations, it was found best to use cylindrically-shaped single crystals 2.5 cm high and about 5 cm in diameter with the cylinder axis a [001] crystallographic direction. The [010] and [100] directions in the cylindrical samples were determined by x-ray photography. Crystals of almost any shape could be used for determining the wavelength dependence of the scattering and the minimum usable size was around 5 cm³. Table I shows the crystal designation and Rayleigh ratio, $R_u = R^2/i/I_0V$ where R is the distance of the observer from the scattering units, i is the intensity of the scattered light, V is the scattering volume, and I_0 is the intensity of the incident light, for the specimens investigated. The Rayleigh ratios were measured with incident light of wavelength, $\lambda = 546 \text{ m}\mu$ and scattering angle, $2\theta = 90^\circ$.

The intensity of the scattered light was measured using an EMI 6256S multiplier phototube which could be rotated around a cylindrical quartz holder containing the sample and the index liquid, benzene. The incident light beam originated from a 300-W concentrated arc lamp and passed through a Bausch and Lomb $\frac{1}{2}$ -m monochromator before entering the crystal as a parallel beam of rectangular cross section. The intensity of the incident beam as a function of wavelength was found by using the same multiplier phototube as for the scattered light and by attenuating it with Cary optical-filter assemblies. The intensity of the light scattering for $2\theta = 90^\circ$ was about 10^{-7} of the incident beam intensity. The volume of the sample "observed" by the detector was about 6 mm³. The normal precautions were taken to avoid stray light in the system³ and as an additional check the wavelength dependence, dissymmetry and depolarization ratio of the scattered light were measured for benzene and for lead silicate glass.⁶ It was found that inserting a 2.5 cm diameter glass bottle containing distilled water into the incident beam did not give the Rayleigh-law wavelength dependence. The scattered intensity of the water was comparable with that for the KCl sample which gave the lowest scattering power; therefore, there is

⁶ This sample was kindly furnished by E. Lell of Bausch and Lomb. The composition of this glass by weight percent is SiO₂-42%, PbO-48%, K₂O-7.1%, TiO₂-1.5%, ZrO₂-0.3%, As₂O₃-0.3%.

TABLE I. Rayleigh's ratios, dissymmetries, and depolarization ratios of several KCl single crystals

Sample	Crystal ^a orientation angle Φ deg.	Rayleigh's ratio ^{b,c} $R_u \times 10^6 \text{ cm}^{-1}$	Dislocation density $\Lambda \times 10^{-5} \text{ cm}^{-2}$	Dissym- metry ^d z	Depolarization ratios ^{b,e}		
					ρ_u	ρ_v	ρ_h
Benzene		16.5		0.98	0.420	0.265	
EDF-1-2	0	28.1		0.99	0.066	0.029	
	45	28.0		0.99	0.067	0.030	
H:40	0	7.1	0.71		0.524		
H:100	0	6.1			0.483		
H:105C	0	13.1		2.4	0.337	0.147	0.544
	45			2.0	0.283	0.031	0.112
H:107C	0	15.0	6.4	2.6	0.217	0.070	
	45			2.8	0.187	0.036	
I:100	0	1.8					
I:101	0	1.6	2.0-4.0				
I:102C	0	5.0		2.1	0.444		
	45			2.8	0.537	0.063	
I:103C	0	8.6	4.3	2.9	0.578		
	45			3.5	0.461	0.068	
I:104C	0	18.0		2.0	0.625	0.214	0.388
	45			2.4	0.630	0.114	0.178
O:100	0	4.4	4.3		0.320		
O:103C	0	1.3	3.3	2.9			
	45			2.9			
O:104C	0	3.1		2.8	0.083		
	45			3.3	0.136		

^a Defined on p. 7.

^b All these measurements were made for $\lambda = 546 \text{ m}\mu$; $2\theta = 90^\circ$.

^c These numbers are normalized to that for pure benzene which is taken as the average of those given in Ref. 13.

^d Defined on p. 5.

^e See p. 8 for a definition of these symbols.

some doubt as to the accuracy of the wavelength dependence for this sample, O:103C. In this connection it should be mentioned that the mismatch in the indices of refraction between water and benzene (or glass) was much greater than that between the crystals and benzene; thus, a relatively higher surface scattering was to be expected for the water.

The polarization measurements were made at $\lambda = 546 \text{ m}\mu$ with Polaroid Corporation HN32 plastic laminated polarizers. A consideration of the reproducibility of the depolarization results and all the possible errors and corrections inherent in these measurements, as enumerated by Goldstein⁷ and reviewed by Stacey,⁸ indicated that the observations are accurate to only about 20%.

All the angular dependence data for the scattering power, $P = i_{2\theta}/I_0V$ which are presented were corrected with the factor $\sin 2\theta$, which accounts for the fact that the scattering volume "seen" by the detector depends on the scattering angle as $1/\sin 2\theta$. Also, the Thompson factor $(1 + \cos^2 2\theta)$ has been removed. The scattering power of benzene when depicted in this manner should be constant as a function of scattering angle; therefore, the dissymmetry, $i_{45^\circ}/i_{135^\circ}$, should be unity.

The optical absorption measurements were made on crystalline plates of thickness 0.1 cm or less which were cleaved from the samples used for the light scattering observations. The irradiations were performed in a $4.1 \times 10^6 \text{ R/h}$ Co⁶⁰ gamma source with the crystalline plates placed in light-tight holders, which

were equipped with slides to permit measurement of the absorption spectra of the specimens. Further experimental details of the color center aspects of this work have already been published.⁹ In order to determine if a correlation between the dislocation density, Λ , and the Rayleigh ratio of the various crystals could be made the crystals were etched by using an acetic acid etchant.¹⁰

RESULTS

Light Scattering

A large variation of the wavelength dependence and magnitude of the scattering power is observed for the different alkali halides, and even among the different KCl crystals. Figure 1 pictures the wavelength dependence when the incident light beam is along a $\langle 100 \rangle$ crystallographic direction and $2\theta = 90^\circ$ for two crystals of KCl and one sample each of KBr and LiF, all purchased from Harshaw. Although the KBr sample has a higher scattering power than any of the other specimens, the LiF crystal has a scattering power between those for the two KCl samples shown. Figure 2 shows the wavelength dependence for several samples of KCl obtained from different suppliers. No correlation of the purchase date with the light-scattering intensity was found. It is clear from Figs. 1 and 2 that it is not possible, at this time, to find the effect of the host matrix, i.e., KCl, LiF, or KBr, on the scattering units. Also, the scattering is apparently by centers which are

⁷ M. Goldstein, *J. Appl. Phys.* **30**, 493 (1959).

⁸ K. A. Stacey, *Light Scattering in Physical Chemistry* (Academic Press Inc., New York, 1956).

⁹ W. A. Sibley and E. Sonder, *Phys. Rev.* **128**, 540 (1962).

¹⁰ M. Sakamoto and S. Kobayashi, *J. Phys. Soc. Japan* **13**, 800 (1958).

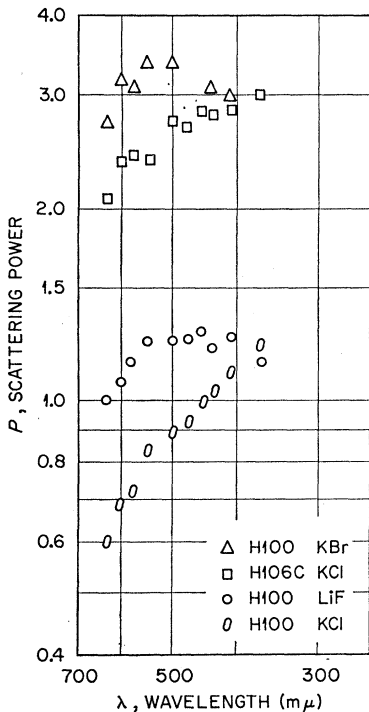


FIG. 1. Wavelength dependence of the scattering power of LiF, KCl, and KBr single crystals. The wavelength, λ , is the wavelength of the incident beam in air.

not intrinsic defects such as vacancies, but extrinsic imperfections such as impurities of various types. If this were not so, it would seem that the variations in scattering power among crystals from the same supplier and having the same approximate dislocation density should be less. An estimate of the absolute scattering power of these samples can be obtained by a comparison with the scattering from benzene shown in Fig. 2. The slope of the logarithm of the scattering power of benzene as a function of the logarithm of the wavelength is -4 , as would be expected for Rayleigh scattering.

Previously it has been observed that KCl crystals grown by Harshaw possessed a crystallographic orientation dependence of the light scattering.³ It is now possible, using an improved experimental apparatus and observing a smaller volume element within the crystals, to make a more complete study of this orientational dependence in crystals of different types and obtained from different sources. The angular dependence of the light-scattering for two different orientations of the crystals is shown in Fig. 3 for two KCl crystals and for benzene. The open-data points were taken with the crystals oriented so that the incident light beam was along a $\langle 100 \rangle$ crystalline direction. The full data points represent the angular dependence with the incident light beam along a $\langle 110 \rangle$ crystalline direction. This same procedure has been used throughout the paper so that full data points always represent an orientation, $\Phi=45^\circ$, with the incident beam along $\langle 110 \rangle$, and the open points are for the incident light along $\langle 100 \rangle$, $\Phi=0^\circ$. The interesting aspects of Fig. 3

are the height of the peak for sample H:105C and the fact that there is no evident orientational dependence for the sample O:104C. It should be mentioned that all the Harshaw and Isomet crystals, which could be used for investigating the angular dependence, had the orientation dependence; however, none of the Optovac specimens had a marked orientation dependence. The shape and height of this orientation peak was observed to vary from sample to sample, and Fig. 4 illustrates that even within one sample, I:104C, there can be a large variation depending upon which volume element of the crystal the detector observes. Sample I:104C was not a typical sample since the variations in the peak were not usually so pronounced, and it was found, when this sample was cleaved into two equal halves, that a rather large polycrystalline section had grown

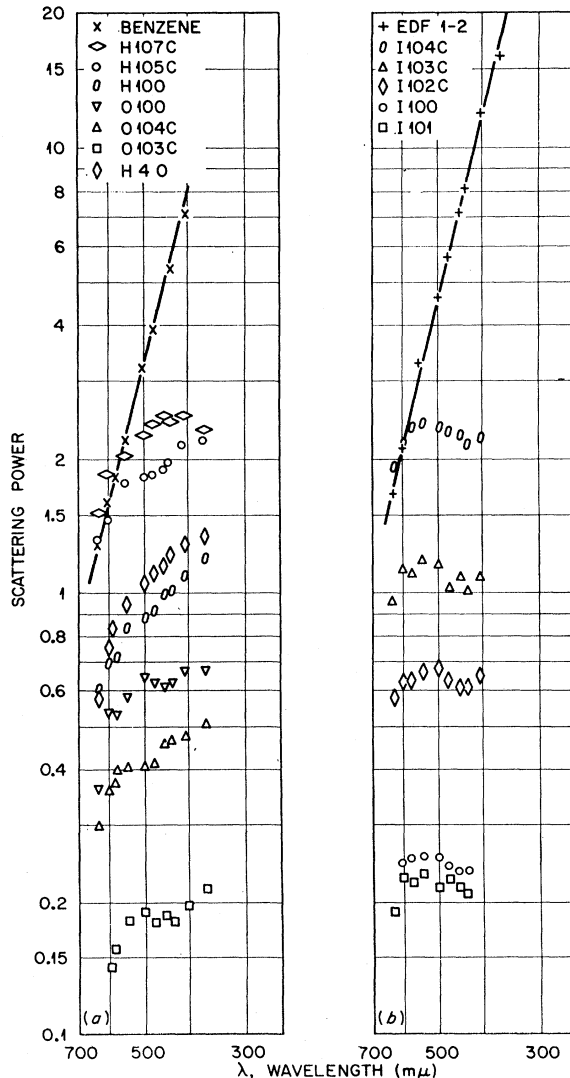


FIG. 2. Wavelength dependence of the scattering power of benzene, lead silicate glass (EDF-1-2), and KCl single crystals produced by different commercial companies.

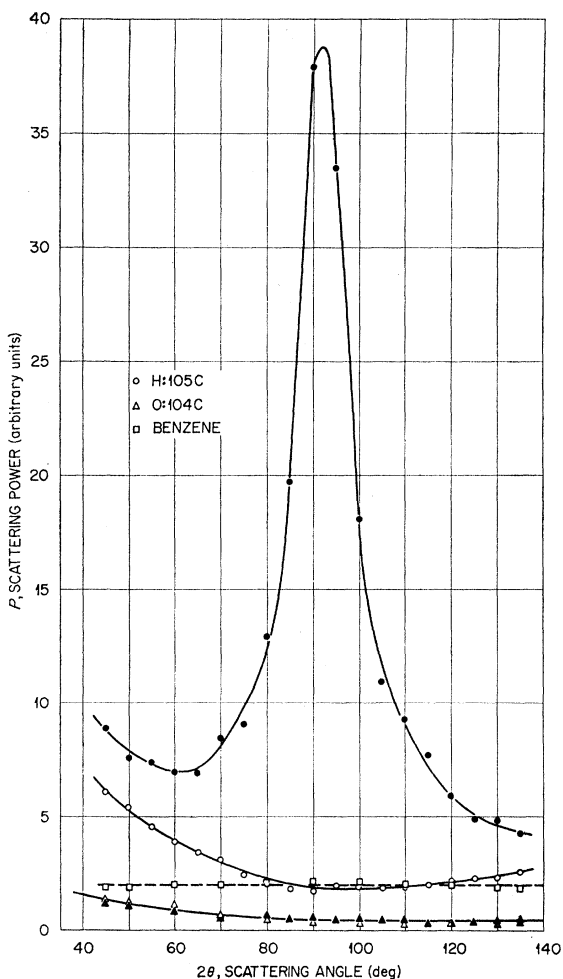


FIG. 3. Angular dependence of the scattering power of benzene, and KCl crystals H:105C and O:104C. The orientation $\Phi=45^\circ$ is represented by the full symbols, and $\Phi=0^\circ$ by the open symbols.

into the middle of the specimen. The well-formed peak in Fig. 4 was found for the good part of the crystal and the peak shape became progressively worse as more of the polycrystalline section came into the observed volume element. The orientational dependence was also measured, as shown by Fig. 5, for Harshaw LiF and Harshaw KBr, although a complete investigation of these samples was not attempted.

The depolarization and dissymmetry measurements are summarized in Table I. As has been mentioned the scattering pattern for the orientation $\Phi=45^\circ$ has a large peak at $2\theta=90^\circ$. If Φ varies by a few degrees then the peak shifts by twice this amount,³ and since the crystals could only be oriented to within $\pm 2^\circ$ of $\Phi=45^\circ$ the intensity measurements taken from a specific angle depend on the position and shape of the scattering peak. Therefore, the accuracy of the dissymmetry and depolarization measurements for the orientation $\Phi=45^\circ$ is not very good except where special care was taken to orient the crystal more precisely, as for sample

H:105C. The depolarization ratios are defined in the following manner:

$$\rho_u = H_u/V_u, \quad \rho_v = \frac{H_v}{V_v}, \quad \rho_h = \frac{V_h}{H_h},$$

where the intensities of the horizontal and vertical components of the light scattered at $2\theta=90^\circ$ are denoted by H and V , respectively, and the subscripts u , h , and v denote unpolarized, horizontally polarized, and vertically polarized incident light. In every crystal it was observed that, within experimental error, the relation $H_v=V_h$ was obeyed. A system of optically isotropic spherical scattering units irrespective of size should give $\rho_v=0$; it is clear from the table, that in all the crystals investigated, this case does not obtain.

Other Measurements

In an effort to determine if a correlation could be found between the "bad regions" associated with the light scattering and other properties of the crystals, several other measurements were made. Since the scattering power of the various samples is observed to vary over an order of magnitude, it should be possible to determine if the number of bad regions or their composition, which is proportional to the scattering

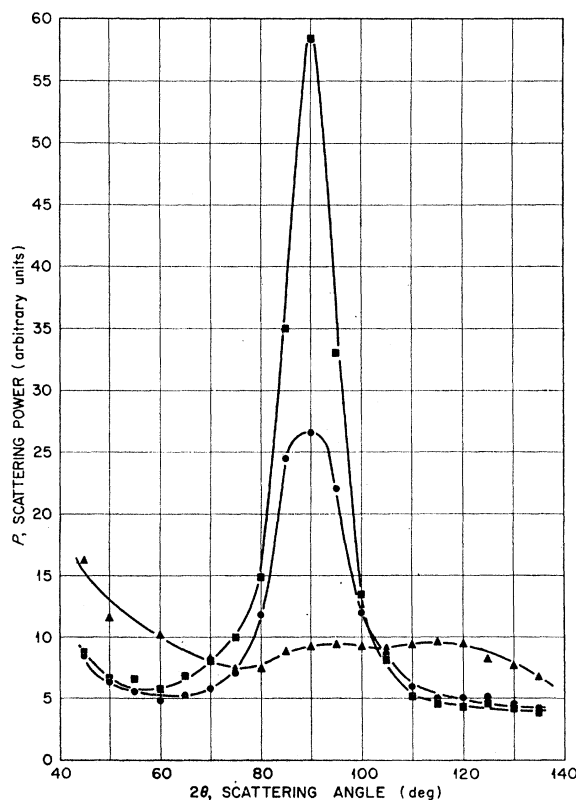


FIG. 4. Angular dependence of the scattering power of one KCl crystal I:104C as a function of position within the sample.

power, influence the radiation coloration rate for such entities as the F and M centers.

The radiation growth curves for F centers produced by gamma rays in some of the crystals investigated are shown in Fig. 6. A comparison of these curves with the scattering power of the same specimens (see Fig. 2) makes it clear that the light-scattering "bad regions" within the crystals do not contribute to the production of F centers after first-stage coloration is completed. The first-stage coloration was not investigated in detail and there is no evidence as to whether a correlation can be found in this case or not. Similarly no correlation was found between the M center growth curves and the light-scattering power.

It was also possible, by making optical measurements of the so-called OH^- band in KCl (204μ), to show that this impurity was not responsible for the light scattering in the Harshaw crystals investigated. A comparison of R_u and the dislocation densities, Λ , of the samples suggests that qualitatively there may be a connection between them. A quantitative correlation between R_u and Λ for the various specimens would be expected only if the scattering were due to vacancy clouds around dislocations and if all of the crystals had the same concentration of vacancies available to form these clouds.

THEORY OF LIGHT SCATTERING BY DISLOCATIONS

The light-scattering by a triad of mutually perpendicular cylindrical bad regions of length Z^* with the scattering centers having a radial distribution function $\rho(r)$ has been calculated by Lester.¹¹ It was found, for $\rho(r) = (V/2\pi r^* Z^*) \exp(-r/r^*)/r$, where V is the scattering volume and r^* is the effective radius of the cylinder, that the scattered intensity i can be written as

$$i = I_0 \frac{2\pi^4 n^4}{R^2 \lambda^4} V^2 \left[\frac{1}{1 + r^{*2}(A^2 + B^2)} \right] (1 + \cos^2 2\theta) \times \sum_{j=i}^3 \langle (\alpha^2)_{av} \rangle_j \left[\frac{\sin(C_j Z^*/2)}{\frac{1}{2} C_j Z^*/2} \right]^2. \quad (1)$$

Where j is an index which ranges over the three possible dislocation orientations, ξ , for the triad in a crystal, and $|\mu| = (4\pi n/\lambda) \sin\theta$; n is the index of refraction,

$$\begin{aligned} A &= |\mu| \cos(\theta - \Phi), \\ B &= -|\mu| \sin(\theta - \Phi) \cos\xi, \\ C &= -|\mu| \sin(\theta - \Phi) \sin\xi, \end{aligned}$$

$\langle \alpha^2 \rangle_{av}$ is the average effective square of the polarizability per unit volume for the scattering system. The

¹¹ D. M. Lester, Thesis, University of Oklahoma, 1960 (unpublished).

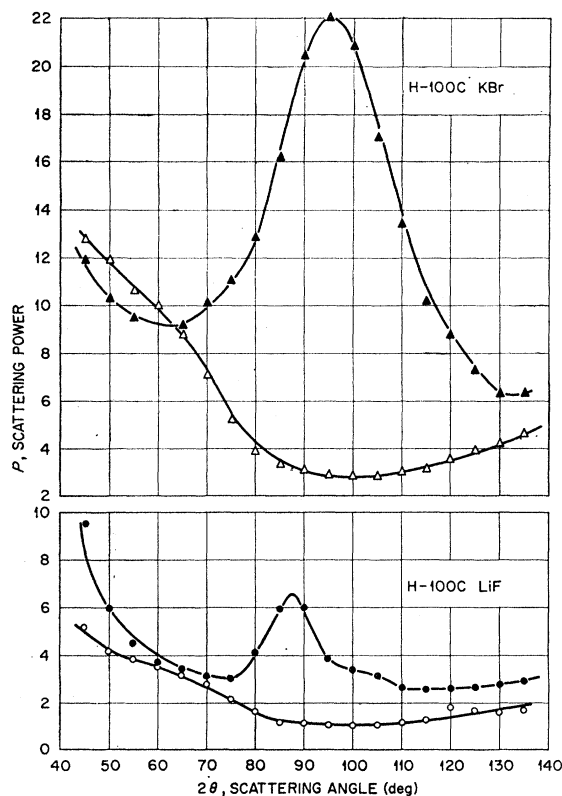


FIG. 5. Angular dependence of the scattering power for two samples of Harshaw LiF and KBr.

angles θ , ξ , Φ are most easily defined as follows: 2θ is the scattering angle, i.e., the angle between the direction of the incident light beam and the direction of observation; ξ is the inclination of the cylindrical bad region axis, Z' , to the vertical, and Φ is the orientation angle of the projection of this bad region axis into the XY plane.

If the polarizability for the scattering system is isotropic, Eq. (1) indicates that, for a triad of mutually perpendicular cylinders oriented along X , Y , and Z , respectively, when $\Phi = 45^\circ$, $Z^* = 3 \times 10^{-4}$ cm, and $r^* \leq 2.5 \times 10^{-6}$ cm, the scattering pattern of i versus 2θ should have a peak at $2\theta = 90^\circ$ with half-width of about 15° and a relative height of twice the scattering power for $2\theta = 45^\circ$. The dissymmetry z should be one. The case $\Phi = 0^\circ$ has no scattering peak and also has a dissymmetry of one. Moreover, the scattering for $2\theta = 45^\circ$ is the same for the cases $\Phi = 0^\circ$ and $\Phi = 45^\circ$. The theory for this special isotropic case has been discussed at length in Refs. 3, 4, and 11, and it can also be shown that Eq. (1) agrees very well with the expression found by van de Hulst¹² for scattering by cylinders.

When the scattering centers do not possess spherical symmetry and the polarizability is not isotropic, then the scattering must be pictured as a consequence of

¹² H. C. van de Hulst, *Light Scattering by Small Particles* (John Wiley & Sons, Inc., New York, 1957).

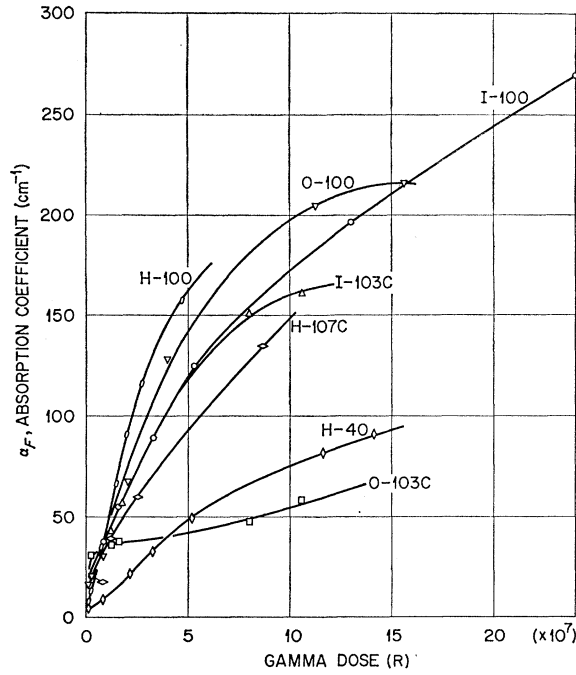


FIG. 6. *F*-center coloring curves for some of the KCl crystals used in this investigation.

the incident light wave inducing an oscillating electric moment in a molecule which is optically unsymmetrical. We will consider only the case where the centers possess an axis of rotational symmetry so that their polarizability ellipsoids are spheroids ($\alpha_{xx'} = \alpha_{yy'} \neq \alpha_{zz'}$). If the principal axes of the polarizability ellipsoids are x' , y' , and z' , and the position of the scattering center relative to the x , y , z system is fixed by the two angles φ and η , where η is the angle between z and z' and φ is the angle between the x axis and the projection of the z' axis into the x - y plane. Therefore, the relation between the components, α_{ij}' and α_{ij} , of the polarizability tensors for the primed and unprimed coordinate systems respectively can be written

$$\begin{aligned} \alpha_{zz} &= \alpha_{zz'} \cos^2 \eta + \alpha_{xx'} \sin^2 \eta, \\ \alpha_{yz} &= (\alpha_{zz'} - \alpha_{xx'}) \sin \eta \cos \eta \cos \varphi, \\ \alpha_{xz} &= (\alpha_{zz'} - \alpha_{xx'}) \sin \eta \cos \eta \sin \varphi, \\ \alpha_{xx} &= \alpha_{zz'} \sin^2 \eta \sin^2 \varphi + \alpha_{xx'} (\cos^2 \varphi + \cos^2 \eta \sin^2 \varphi), \\ \alpha_{yy} &= \alpha_{zz'} \sin^2 \eta \cos^2 \varphi + \alpha_{xx'} (\sin^2 \varphi + \cos^2 \eta \cos^2 \varphi). \end{aligned} \quad (2)$$

In the evaluation of the effect that an anisotropic polarizability has on the scattering pattern, i versus 2θ , it must be remembered that any analysis of the light-scattering pattern can only determine a pair distribution function or average projection of the scattering units. Therefore, when a scattering unit has several different orientations in the lattice, the light-scattering pattern is characteristic of a superimposition of the orientations. For the special case of dislocation-defect-

cloud scattering, this observation is important since for the (100) face of a crystal the line formed by the edge of the extra-half planes of the edge dislocations and the crystal face may be in any of four directions $[011]$, $[0\bar{1}1]$, $[01\bar{1}]$, and $[0\bar{1}\bar{1}]$ and the defect cloud may have a particular orientation with respect to these lines. It is, therefore, necessary to use an effective polarizability ellipsoid representative of the average projection of the scattering units to evaluate the scattering pattern. This can be done by averaging with the proper weighting function over φ . It would be expected that the scattering centers in the cloud around the edge of the extra-half plane of the dislocation would be at least partially oriented because of the elastic and electric fields associated with this line. Therefore, it is necessary that the weighting function over φ show this effect. When an arbitrary weighting function of $\cos^4 \varphi$ representing an effective defect distribution for the case $\Phi = 0^\circ$ is used, then Eq. (2) may be written

$$\begin{aligned} \langle \alpha_{zz}^2 \rangle_{av} &= (\alpha_{zz'} \cos^2 \eta + \alpha_{xx'} \sin^2 \eta)^2, \\ \langle \alpha_{zy}^2 \rangle_{av} &= \frac{1}{2} (\alpha_{zz'} - \alpha_{xx'})^2 \sin^2 \eta \cos^2 \eta, \\ \langle \alpha_{xy}^2 \rangle_{av} &= \frac{1}{24} (\alpha_{zz'} - \alpha_{xx'})^2 \sin^4 \eta. \end{aligned} \quad (3)$$

If a weighting function of $\sin^4 \varphi$ is employed, characteristic of the effective defect distribution for dislocation lines along the Z axis and the case $\Phi = 45^\circ$, then it is found that,

$$\begin{aligned} \langle \alpha_{zz}^2 \rangle_{av} &= (\alpha_{zz'} \cos^2 \eta + \alpha_{xx'} \sin^2 \eta)^2, \\ \langle \alpha_{zy}^2 \rangle_{av} &= \frac{1}{2} (\alpha_{zz'} - \alpha_{xx'})^2 \sin^2 \eta \cos^2 \eta, \\ \langle \alpha_{xy}^2 \rangle_{av} &= \frac{5}{24} (\alpha_{zz'} - \alpha_{xx'})^2 \sin^4 \eta. \end{aligned} \quad (4)$$

When the scattered intensity, i , is evaluated as a function of scattering angle, for the triad of mutually perpendicular cylinders mentioned earlier, it is found that the intensity peak occurring at $2\theta = 90^\circ$ depends markedly on the orientation of the scattering-center polarizability ellipsoids and on the ratio of $\alpha_{zz'}/\alpha_{xx'}$. The dissymmetry is not necessarily unity for this case, and the scattered intensity for $2\theta = 45^\circ$ when $\Phi = 0^\circ$ is not equal to that for $2\theta = 45^\circ$ when $\Phi = 45^\circ$ as was the case for an isotropic polarizability. Also, the scattering pattern is dependent upon the expression chosen for the weighting function. Therefore, the parameters that most affect the shape of the calculated scattering pattern are the weighting function, the ratio of $\alpha_{zz'}/\alpha_{xx'}$, the orientation, η , of the defect-scattering-center polarizability ellipsoid with respect to the dislocation line or bad region cylinder axis Z' , and the length of the bad region Z^* . The ratio $\alpha_{zz'}/\alpha_{xx'}$ and the parameter η can be determined from the depolarization measurements,

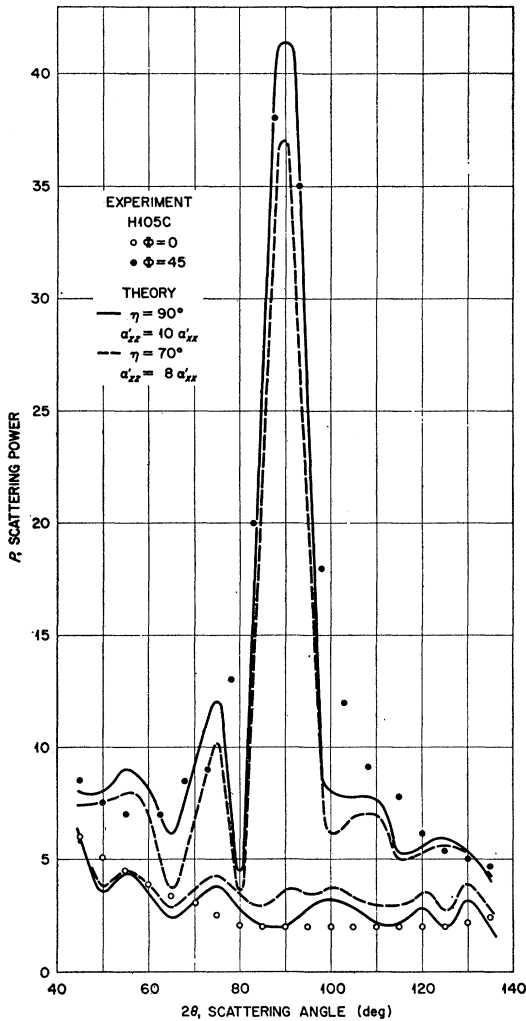


FIG. 7. Comparison of light scattering theory and the experimental data for sample H:105C. In the theory the effective dislocation length, Z^* , was taken as 3×10^{-4} cm.

since

$$\begin{aligned} H_v &\propto \sum_{j=1}^3 \langle \alpha_{yz}^2 \rangle_{av}, & V_h &\propto \sum_{j=1}^3 \langle \alpha_{xz}^2 \rangle_{av}, \\ V_v &\propto \sum_{j=1}^3 \langle \alpha_{zz}^2 \rangle_{av}; & H_h &\propto \sum_{j=1}^3 \langle \alpha_{xy}^2 \rangle_{av}. \end{aligned} \quad (5)$$

DISCUSSION

If it is assumed that the dominant light-scattering units in the crystals are dislocations surrounded by impurity clouds, then it is possible through the use of Eqs. (1), (3), and (4) to calculate the scattering pattern for given values of η and $\alpha_{zz}'/\alpha_{xx}'$ and for the two different orientations $\Phi=0^\circ$ and $\Phi=45^\circ$. Also, values for η and $\alpha_{zz}'/\alpha_{xx}'$ can be found independently from the depolarization data given in Table I. It must be remembered that the values for η and $\alpha_{zz}'/\alpha_{xx}'$ are those characteristic of the apparent scattering unit and

not just the individual scattering centers. This is especially important since the apparent scattering unit is a superimposition of the individual scattering units which have several different orientations. Consider only crystal H:105C as a typical example. It is found that the depolarization data give $\eta=70^\circ$ and $\alpha_{zz}'=6.3 \alpha_{xx}'$ for the orientation $\Phi=0^\circ$, and $\alpha_{zz}'=8.2 \alpha_{xx}'$, and $\eta=80^\circ$ for $\Phi=45^\circ$. These values for $\alpha_{zz}'/\alpha_{xx}'$ seem rather large, even for triatomic molecules which could be the impurities responsible for the observed scattering. However, since the impurities are in the electric field of an edge dislocation it is possible that the ratio $\alpha_{zz}'/\alpha_{xx}'$ is increased; particularly for the values of η that are observed. Figure 7 shows the fit of theory to experiment which can be obtained using Eqs. (1), (3), and (4) and the values shown in the figure for η and the ratio $\alpha_{zz}'/\alpha_{xx}'$ with $Z^*=3 \times 10^{-4}$ cm. The theory has been adjusted to fit one experimental data point: That for the orientation $\Phi=0$ and scattering angle $2\theta=45^\circ$. No other fitting was necessary. Of course, the experimental data points cannot give the resolution and fine structure which the theory predicts because there is a spread in the observation angle and because the dislocation networks are probably not perfectly aligned. However, the fit of theory to experiment is apparently rather good and gives credibility to the idea that the dominant scattering units within the crystals are dislocations surrounded by defect clouds.

The large variation in scattering intensity shown in Figs. 1 and 2 suggests that the individual scattering centers and their concentrations within the scattering units vary from sample to sample, and are most likely impurities. The infrared spectra of these and other crystals (some as long as 10 cm) were investigated and it was found that the Harshaw KCl samples contained traces of such impurities as BO_2^- . The Isomet samples showed traces (about 0.5 ppm) of OCN^- . If these impurities enter the crystalline lattice as linear molecules, then their anisotropic polarizability ellipsoid and their concentration would make them one possible source of observed light scattering.

Rayleigh's ratio for benzene scattering at $546 \text{ m}\mu$ is $16.5 \times 10^{-6} \text{ cm}^{-1}$,¹³ and a comparison of the scattering power of benzene with that of the individual samples investigated allows one to calculate a value for $NV^2 \langle \alpha^2 \rangle_{av}$ from the formula

$$\begin{aligned} P = \frac{i}{I_0 V} &= V^2 \left(\frac{2\pi^4 n^4}{R^2 \lambda^4} \right) N \left[\sum_{j=1}^3 \langle \alpha^2 \rangle_{avj} \left(\frac{\sin(C_j Z^*/2)}{C_j Z^*/2} \right)^2 \right] \\ &\times (1 + \cos^2 2\theta) \left[\frac{1}{1 + r^{*2}(A^2 + B^2)} \right], \quad (6) \end{aligned}$$

where N is the number of scattering units per unit volume and j ranges over the three possible disloca-

¹³ D. K. Carpenter and W. R. Krigbaum, J. Chem. Phys. 24, 1041 (1956).

tion orientations, ξ , in KCl with $\Phi=0$ or 45° . All other symbols are the same as those in Eq. (1). For Harshaw KCl samples the quantity $NV^2 \langle \alpha^2 \rangle_{av}$ varies from 0.80 to 1.70×10^{-26} cm³ and for Isomet KCl the variation is from 0.21 to 2.37×10^{-26} cm³. It is possible to calculate the polarizability per unit volume for the scattering units since we have experimental estimates for V and N . Consider the case which requires the largest value for $\langle \alpha^2 \rangle_{av}$, sample H:40. When $V=4.0 \times 10^{-15}$ cm³ and $N=1.9 \times 10^7$ cm⁻³ then $\langle \alpha^2 \rangle_{av}^{1/2}$ is 0.5×10^{-2} . If an upper limit of 10^{21} cm⁻³ is taken for the density of scattering centers within the bad region, then the effective defect polarizability, $\langle \alpha^2 \rangle_{av}^{1/2}$, is 5×10^{-24} cm³. These values seem high, but are not unreasonable since this would correspond to a total of only 7.6×10^{13} scattering defects per unit volume of crystal.

The Optovac KCl crystals do not show the characteristic orientation peaks in the scattering pattern that are observed for the Harshaw and Isomet samples; therefore, it is perhaps not feasible to apply the above analysis in this case. However, it should be mentioned that if the dislocation networks in these crystals are somewhat irregular and the polarizability ellipsoid is not markedly anisotropic, then the distinctive peak in the scattering pattern will not be apparent. Moreover, it was found, by etching, that the particular Optovac samples investigated had a high concentration (between 5 and 80 mm⁻³) of sub-boundaries. This would cause the dislocation networks to be very non-uniform and would reduce the possibility of seeing an orientational dependence of the light scattering. Thus, it is not possible to eliminate dislocations as the dominant scatterers in the Optovac specimens. This point is further illustrated by Fig. 4 which shows what happens to a scattering pattern as the dislocation networks become more and more irregular.

Previously,^{2,5} it was suggested that the relatively high forward scattering at $2\theta=45^\circ$, and, hence, the large dissymmetry for KCl crystals could be due to a contribution to the scattering by rather large precipitates. This approach allowed the use of an isotropic polarizability in Eq. (1) and the addition of the contributions to the scattering from the two different types of scattering units did fit the data that were then available. However, the present experimental observations suggest that only one type of scattering unit is responsible for the scattering. The scattering at $2\theta=90^\circ$ for $\Phi=45^\circ$, is about a factor 20 greater for some crystals than the scattering at $2\theta=90^\circ$ for $\Phi=0^\circ$ and if the isotropic polarizability case used before were applicable this factor should be two. Furthermore, several samples were taken to high temperatures (525 to 725°C) and then air cooled. As had been noted before,⁵ the scattering power of the heat treated samples

was quite different from that for the same specimens before heat treatment, but the high forward scattering and the large dissymmetry were still observed. This suggests that, unless the precipitates grew and dispersed in exactly the same manner as the defect cloud around the dislocations, only one type of scattering unit is responsible for all the scattering observed.

A comparison of the Rayleigh ratio values given in Table I with the rate of F -center coloration shown in Fig. 6 indicates that the scattering centers within the bad regions do not affect the growth rates for either F or M centers. The differences in the Rayleigh ratios given in Table I can be due to changes from sample to sample of either the dislocation density, Λ , or the type and concentration of impurity and, hence, $\langle \alpha^2 \rangle_{av}$, or the effective scattering volume, V . The latter is relatively well known from the analysis of the scattering pattern which gives $Z^* \approx 3 \times 10^{-4}$ cm and $r^* \leq 2.5 \times 10^{-6}$ cm. Thus, it is only possible to state at this time that the coloration rates in the specimens investigated are not due to the type of impurities surrounding the dislocations, and may not be affected by the number of light-scattering bad regions within the crystals.

SUMMARY

(1) It is possible to explain all of the experimental data thus far obtained in terms of light scattering from only one type of scattering unit. Dislocations, which are surrounded by scattering centers having an anisotropic polarizability ellipsoid, are most probably the scattering units responsible for the light scattering.

(2) The large variations in the observed scattering patterns suggest that the regularity of the dislocation networks is very good in only a few of the crystals investigated and indeed varies from place to place within a specific sample. Moreover, the height of the scattering peak and its variation among different samples for the orientation $\Phi=45^\circ$ suggests that the scattering centers are impurities or impurity complexes.

(3) There is no correlation between the scattering power of a crystal and the growth rates of either the F band or M band under ionizing radiation. This suggests that the bad regions associated with the light scattering do not affect the coloration properties of a crystal.

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