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Some F-Band Optical Oscillator Strengths in Additively Colored Alkali Halides*†

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The number of magnetic centers in alkali halides additively colored in the alkali vapor has been determined by measuring the static magnetic susceptibility. Curie's law is followed, and the measurements have been extended to liquid helium temperatures to increase the sensitivity. The number of centers is compared with the optical absorption of the F band. Under the assumptions that the F-center electron wave function depends parametrically upon the coordinates of the surrounding nuclei, the electric dipole matrix element is independent of the nuclear coordinates, and the electric field at the electron is the Lorentz local field, the optical oscillator strengths for a Lorentz, Gaussian, and observed line shape are determined. The average for a series of experiments yield $f_L=0.66$ for KCl, $f_L=0.71$ for KBr, and $f_L=0.46$ for KI. Values for CsBr and NaCl are discussed.

INTRODUCTION

THE correlation of the magnetic and optical properties of F centers form the basis of this research. Excellent review articles concerning much of the experimental and theoretical work on color centers are given by Pohl[1] and more recently by Seitz.[2] The primary absorption in an alkali-halide crystal additively colored with the alkali vapor is the F band, and, following the model of deBoer,[3] may be regarded as an electron trapped at a halide vacancy. The characteristic optical absorption band is associated with the electronic transition of the F-center electron from its ground state to the first excited state, and the temperature dependent paramagnetism is due to the magnetic moment of the F-center electron. The doubly degenerate state of the F-center electron cannot be lifted by the crystalline field alone, and thus the magnetic susceptibility follows the Curie-law temperature dependence until either interaction between F centers or hyperfine coupling with the surrounding ions becomes important. Since these effects are important only at very low temperatures for available concentrations, one has an accurate measurement of the number of F centers. As will be discussed later, simple models of the F-band absorption indicate that the number of F centers and the amount of optical absorption are highly dependent on the electronic transition probability and also on the local field. A systematic study of this correlation when combined with the absorption energy, g value, and electron-spin—nuclear-spin coupling should assist in the analysis of the F-center wave functions.

Kleinschrod[4] initiated studies of this nature by determining the number of absorption centers from the optical absorption data and the Smakula dispersion formula, and then correlated the above with the chemical determination of the number of excess potassium atoms in the additively colored crystal. Jensen[5] correlated the change in magnetic susceptibility with the optical absorption of ultraviolet irradiated KBr. Scott *et al.*[6] indicated that the magnetic susceptibility at room temperature depended both on the concentration of F centers and other centers on the long-wavelength side of the F band. Hutchinson[7] in his paramagnetic resonance absorption experiments found a g value of approximately the free-electron value for the F center. Heer and Rauch[8] extended the static magnetic susceptibility measurements to helium temperature range and found that the F-center paramagnetic susceptibility followed Curie's law down to 1°K.

THEORETICAL ASPECTS

The absorption spectrum of the F-center electron, as observed, consists of a strong main band, the F band, which is attributed to an electronic transition from the ground state "a" to the excited state "b." The absorption cross section of the F center, under the assumptions that the wave function of the F-center electron depends parametrically upon the coordinates of the surrounding nuclei, i.e., the Born-Oppenheimer approximation, and the "Condon" approximation that the electric dipole matrix element is independent of the nuclear coordinates, can be written using the notation of Lax[9] as

$$\sigma(\nu)=\frac{8\pi^3\nu}{3ch}\left[\frac{n}{\epsilon}\left(\frac{E_e}{E}\right)^2\right]|\mathbf{M}_{ab}|^2 g(\nu), \qquad (1)$$

where ν is the frequency of the incident radiation, n the index of refraction for frequency ν, ϵ the dielectric constant, E_e the effective field seen by the electron, E the microscopic field, $\mathbf{M}_{ab}$ the electric dipole matrix element. For sufficiently narrow lines $g(\nu)$ is the experi-

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[1] R. W. Pohl, Proc. Phys. Soc. (London) 49, 3 (1939).

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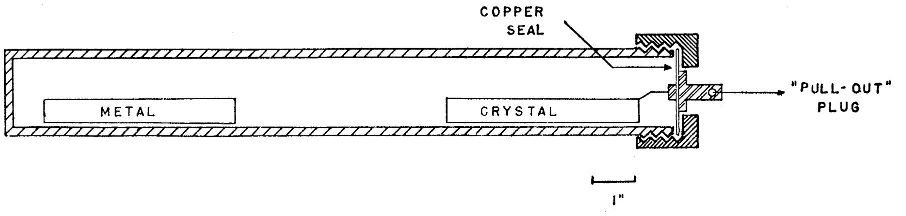

FIG. 1. Cross section of chamber for additive coloration of alkali-halide crystals.

mental line shape. The absorption coefficient $\alpha(\nu)$ is usually defined as $N\sigma(\nu)$, where N is the number of absorbers per cm^3. Since under the above assumptions $\int g(\nu)d\nu=1$, and in general

$$\begin{bmatrix}\text{area under the}\\ \text{absorption curve}\end{bmatrix}=\int\alpha(\nu)d\nu=\int N\sigma(\nu)d\nu,$$

the area under the absorption curve is intimately related to the matrix element of the electric dipole transition and the effective field seen by the electron. Lax[9] has shown that the area under the absorption curve is related to the matrix element of the electric dipole transition under even less restrictive conditions, and has applied these calculations to more complex models. The oscillator strength[10] f_{ab} is defined as

$$f_{ab}=(8\pi^2m/3he^2)\nu_{ab}|\mathbf{M}_{ab}|^2. \qquad (2)$$

If one further assumes that the field seen by the electron is the Lorentz local field, then $(n/\epsilon)(E_e/E)^2=(n^2+2)^2/9n$ and the absorption coefficient to the approximation given in Eq. (1) can be written as

$$\alpha(\nu)=[(\pi e^2/mc)(n^2+2)^2/9n]g(\nu)Nf, \qquad (3)$$

or

$$Nf=[9n/(n^2+2)^2](mc/\pi e^2)\begin{Bmatrix}\text{area under the}\\ \text{absorption curve}\end{Bmatrix}, \qquad (4a)$$

$$Nf=0.821\times10^{17}[n/(n^2+2)^2]\times\begin{Bmatrix}\text{area under absorption}\\ \text{curve in cm}^{-1}\times\text{ev}\end{Bmatrix}\text{ cm}^{-3}. \qquad (4b)$$

The "area under the absorption curve" $=(\pi/2)\alpha_{\max}\Delta\nu_{\frac{1}{2}}$ for a Lorentz line shape; and $(\pi/4\ln2)^{\frac{1}{2}}\alpha_{\max}\Delta\nu_{\frac{1}{2}}$ for a Gaussian line shape, where $\alpha_{\max}$ is the maximum absorption coefficient of the F band in cm^{-1} and $\Delta\nu_{\frac{1}{2}}$ the band width at half-maximum in electron volts. It may be noted that the Lorentz line shape yields the same oscillator strengths as the Smakula dispersion formula.[11]

There are two magnetic sublevels of the F-center electron due to its spin. The paramagnetic resonance experiments[12] have shown a hyperfine interaction with the surrounding ions which causes a small splitting of these levels. Korringa and Daunt[13] have computed the magnetic properties of F centers based on the molecular orbital model used so successfully by Kip and co-workers in explaining the microwave resonance experiments. Since these interactions are small, one obtains the usual Curie-law temperature dependence above 1°K, and the number of centers is related to the susceptibility by

$$\chi=(Ng^2\beta^2/4kT)[1-\tfrac{1}{3}(g\beta H/2kT)^2+\cdots], \qquad (5)$$

where g represents the g value, β the Bohr magneton, k the Boltzman constant, and H the external magnetic field. The correction to the g value due to spin-orbit coupling is quite small and will be neglected in our calculations. Saturation effects due to the second term in 5 may be kept small by using small values of H/T, and corrections only become important below 1.5°K for fields of less than 10 000 gauss. Deviations from Curie's law can occur due to the overlap of the wave functions of two adjacent F centers, the interaction giving rise to an anti-parallel state of lower energy. Estimates based upon hydrogen-like wave functions indicate that the overlap will not be important for concentrations of less than 10^{20} F centers per cm^3 at temperatures above 1°K.

EXPERIMENTAL TECHNIQUES

Preparation of the Additively Colored Crystals

Alkali halides colored by the addition of a stoichiometric excess of alkali metal possess a relatively stable coloration, and if the quench from high temperatures is sufficiently rapid the coloration will be primarily that due to the F center. Aggregate centers such as the R center, M center, and colloid center begin to develop during the quench, and will be frozen in if the quenching process is too slow. Since the models for the R and M centers suggested by Seitz[2] indicate that they may also be paramagnetic, and since the colloid band contributes to optical absorption in the same region as the F band, it is highly desirable that their concentration be minimized. Since other interests in the low-temperature properties required large samples, we developed the following techniques for the coloration of large samples.

A bomb which is suitable for independent control of the temperature of the vapor and of the crystal, and also suitable for rapid quench of the crystal, is shown in Fig. 1. The shell of the bomb is made from

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[10] See, for example, E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1935).

[11] A. Smakula, Z. Physik **59**, 603 (1930).

[12] Kip, Kittel, Levy, and Portis, Phys. Rev. **91**, 1066 (1953).

[13] J. Korringa and J. G. Daunt, Phys. Rev. **102**, 92 (1956).

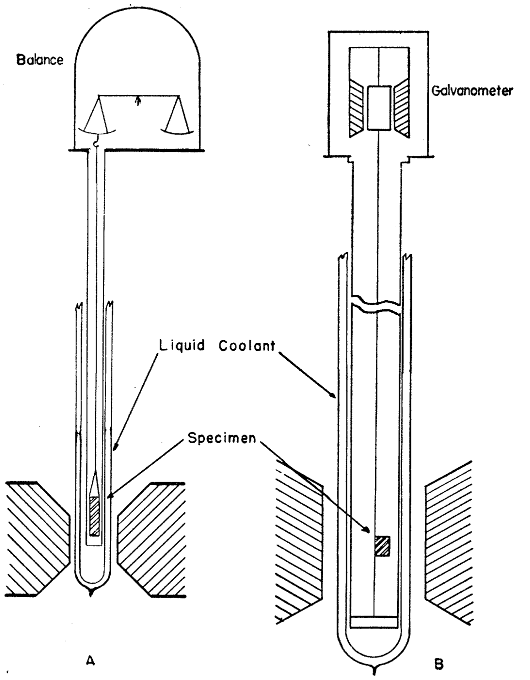

FIG. 2. (A) Schematic diagram of Gouy balance, and (B) schematic diagram of torsion balance for the measurement of magnetic susceptibility.

Inconel. An Inconel metal boat contains the alkali metal and is so adjusted in the bomb that its temperature is controlled by one furnace. The second metal boat contains the alkali-halide crystals, and their temperature is controlled by a second furnace. The end is sealed with a thin copper disk in a manner such that the seal can be broken by a quick tug on the pull-out plug, and the sample immediately dropped into the quench solution. Acetic acid was found to provide the most rapid quench. Alkali-halide crystals in the form of rods 1.2 cm in diameter and 2.5 cm in length were colored by this method. All crystals with a side-band absorption larger than 10% of the F-band peak and an inhomogeneity of coloration greater than 25% were regarded as unsuitable for our experimental work.

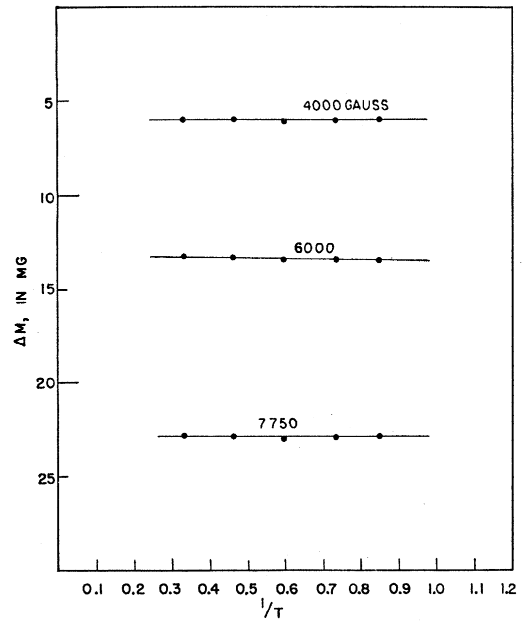

FIG. 3. The change in weight in milligrams of a pure KCl crystal upon application of a magnetic field as a function of $1/T(^\circ\mathrm{K})^{-1}$.

MAGNETIC MEASUREMENTS

The force[14] on a small element of volume of a weakly magnetic substance due to a magnetic field $\mathbf{H}$ may be written as

$$\text{force} = \mathbf{M}\cdot\text{grad}\mathbf{H}dv = \tfrac{1}{2}\chi\ \text{grad}\mathbf{H}^2 dv. \qquad (6)$$

$\mathbf{M}$ is the magnetic moment, and $\mathbf{M}=\chi\mathbf{H}$ for a nonferromagnetic substance. For a given experimental arrangement, the magnetic field distribution of the magnet remains constant, and the magnetic susceptibility is directly proportional to the force. The magnetic susceptibility of the colored alkali halides consists of the usual temperature independent diamagnetic susceptibility of the alkali halide, and a temperature dependent paramagnetic susceptibility due to the coloration. In the temperature region in which Curie's law is valid, i.e., the value of H/T is sufficiently small that the effects of saturation represented by the second term in Eq. (5) may be neglected, the magnetic susceptibility is given by

$$\chi = C/T + \chi_{\text{diamagnetic}}. \qquad (7)$$

Thus a knowledge of the high temperature diamagnetic susceptibility and the slope of the force *vs* temperature curve for a given magnetic field distribution yields a value for the Curie constant C. This may be combined with Eq. (5) for the number of magnetic spin systems. Using the free electron or observed g value, the number of centers/cm^3 is given by

$$N = 1.62\times 10^{+24}C \text{ center/cm}^3. \qquad (8)$$

Since in a strong magnetic field the magnetization M becomes constant, the sample may be examined for the presence of ferromagnetic impurities by measuring the Curie constant at a number of field strengths. If these values are consistent, then the effect of such impurities is small.

The force exerted on the sample by the applied magnetic field was measured by one of the two methods shown in Fig. 2. In each method the sample was surrounded by a small amount of helium gas for heat transfer to the liquid helium in the surrounding cryostat. The Gouy balance was used to measure the force

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[14] See, for example, L. F. Bates, *Modern Magnetism* (Cambridge University Press, Cambridge, 1953), third edition, for a discussion of the various techniques for measuring the magnetic susceptibility.

on the large specimens, and consisted of an Ainsworth Type BB chainweight balance modified for operation in a vacuum. The chain for restoring the balance is operated through vacuum seals. The magnetic field was produced by a conventional electromagnet and was controlled by the current to the field windings. Some results for the change in weight of a crystal of KCl received from Harshaw Chemical Company is shown in Fig. 3. The field at the magnet center is shown on each curve. The curves have negligible slope and indicate an impurity of less than 2×10^{16} magnetic ions per cm^3. Figure 4 shows the change in weight as a function of the reciprocal of the temperature for the additively colored sample of KCl labeled crystal No. 3 in Table I. This sample contains 7.5×10^{17} color centers per cm^3. Some of our experiments were repeated on small ($0.05\times 0.5\times 0.5$ cm) samples by the measurement of the torque on the sample in an inhomogeneous magnetic field. The restoring current through the galvanometer is a measure of the torque on the sample This latter method is more sensitive and has the additional avantage in that it may be combined with optical measurements.

TABLE I. Potassium chloride. N_m is the number of centers/cm^3 measured magnetically; α_{max} is the maximum absorption coefficient of F band; $\Delta\nu_{\frac{1}{2}}$ is the F-band width at half-maximum absorption; f_L, f_g, and f_{area} are the oscillator strengths determined for Lorentz, Gaussian, and observed line shapes.

Crystal	N_m cm^{-3}	α_{max} cm^{-1}	$\Delta\nu_{\frac{1}{2}}$ ev	f_L	f_g	f_{area}
1	9.5×10^{17}	169	0.35	0.67	0.45	
2	9.0×10^{17}	152	0.35	0.64	0.43	
3	7.5×10^{17}	131	0.35	0.66	0.45	0.51
4[a]	5.6×10^{17}	97	0.35	0.66	0.45	0.49
Average				0.66	0.45	0.50

[a] Measured by the torsion method.

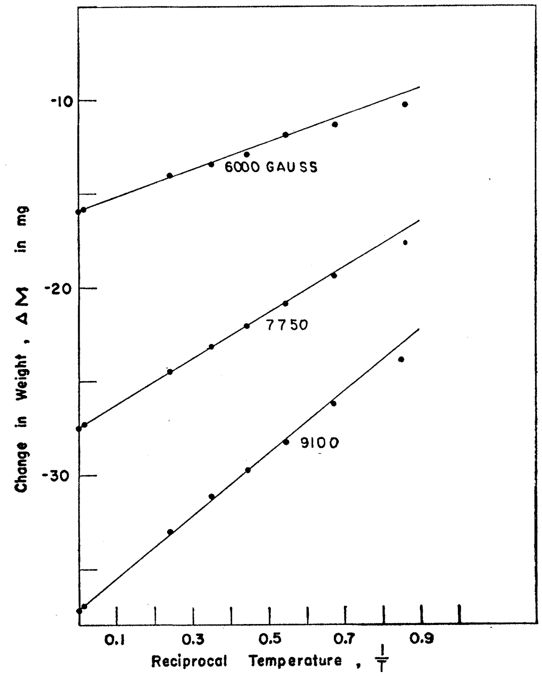

FIG. 4. Change in weight in milligrams of an additively colored KCl crystal upon application of a magnetic field as a function of $1/T(°K)^{-1}$.

OPTICAL MEASUREMENTS

After the magnetic susceptibility of the large samples was measured by the Gouy method, the specimen was cleaved into ten sections along the length of the crystal. Small crystals thin enough for optical absorption measurements were cleaved from each section. The optical absorption was measured with a Beckman Model DU spectrophotometer. If the optical absorption coefficient α varied by more than 25% throughout the length of the sample, the sample was rejected. The effective absorption coefficient for comparison with the magnetic measurements was taken as the average of the absorption coefficient for the individual sections. If the absorption coefficient is proportional to the number of absorbing centers, then this method of averaging is appropriate. Absorption curves are shown for each sample in the following discussion. Since optical imperfections were introduced in our cleaving process, the crystals were compared with uncolored cleaved crystals. This background was subtracted from the total absorption.

The averaging technique used for the large samples was not necessary for the small samples measured with the torsion balance. In this case the same cleaved chip could be used for both optical and magnetic measurements.

EXPERIMENTAL RESULTS AND DISCUSSION

All of the alkali-halide crystals examined in the following experiments were obtained from the Harshaw Chemical Company. The technique for quenching was

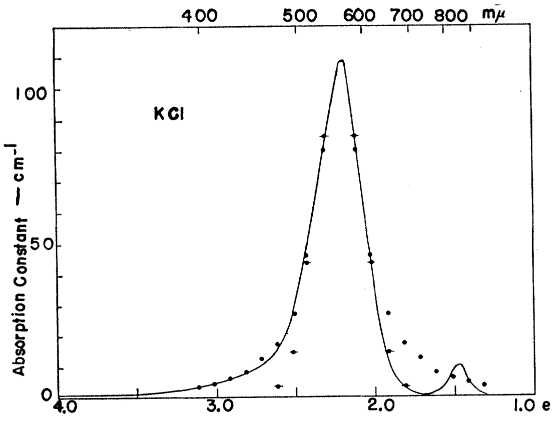

FIG. 5. The optical absorption curve for additively colored KCl. —Experimental absorption curve; ● Lorentz line shape; -●- Gaussian line shape.

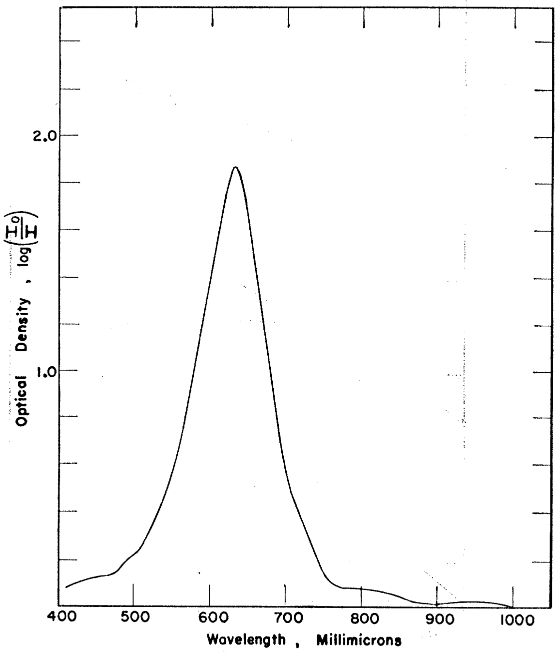

FIG. 6. Optical absorption curve for additively colored KBr.

described in a previous section; the earlier samples were quenched in carbon tetrachloride and the more recent samples in acetic acid. A control crystal was quenched in each case and experiments were made on only those crystals which showed favorable absorption spectra. The crystal, the number of centers measured magnetically, the optical absorption coefficient at room temperature, the band width at half-maximum, and the optical oscillator strengths determined for a Lorentz line shape, a Gaussian line shape, and from the area under the absorption curve are shown in the following tables for the various alkali-halide crystals. The absorption spectra of some of the selected crystals is shown in each case.

Potassium Chloride

Experimental results for KCl are tabulated in Table I.[15] The number of magnetic centers for the first three large samples was determined by the Gouy method, and the fourth sample was measured with the torsion balance. It may be noted that the optical oscillator strength for the Lorentz line shape as determined from the absorption coefficient, and the band width at half-maximum are in substantial agreement for the large and small samples, and over a concentration range of the order of two. An optical absorption curve is shown in Fig. 5. Circles indicating the shape of the absorption curve for the Lorentz and Gaussian line shapes are included for comparison purposes. As has been suggested previously,[16] the Lorentz line shape is more appropriate for a short region on the short-wavelength side, and the Gaussian line shape on the low-frequency side of the F band. A comparison with the area under the curve is included for samples 3 and 4.

The $f_L=0.66$ is considerably below Kleinschrod's[4] value of $f_L=0.81$ determined from a comparison of the excess potassium, and the more recent results of Silsbee[17] using microwave absorption techniques yielding $f_L=0.85$. Our value is more nearly in agreement with the work of Witt[18] on the change of density upon coloration by electrolysis. The work of Lin[19] on the expansion of x-irradiated KCl supports the results of Kleinschrod if the F-center coloration is a good measure of the number of vacancy pairs formed and the elementary assumption that the change in linear dimension is simply related to the number of pairs is valid.

The more obvious criticisms which may be applied to our static magnetic measurements regard the magnetic purity of our sample, and the contribution of color centers other than the F center to the susceptibility. The purity of an uncolored sample is shown in Fig. 2, the magnetic impurity being less than 2×10^{16} impurity atoms per cm^3. The susceptibility of samples which were subjected to the same coloration procedure as the samples reported above, but quenched too slowly for the formation of an appreciable number of F centers were measured. The magnetic susceptibility was comparable to the untreated sample and showed negligible temperature-dependent susceptibility. The samples usually showed a contribution due to the M center on

TABLE II. Potassium bromide.

Crystal	N_m cm^{-3}	$\alpha_{\max}$ cm^{-1}	$\Delta\nu_{\frac{1}{2}}$ ev	f_L	f_g	f_{area}
1	21.5×10^{17}	410	0.37	0.72	0.49	
2	4.97×10^{17}	88	0.38	0.70	0.47	
3	6.73×10^{17}	125	0.38	0.72	0.49	
4	10.5×10^{17}	184	0.38	0.69	0.47	0.52
Average				0.71	0.48	0.52

TABLE III. Potassium iodide.

Crystal	N_m cm^{-3}	$\alpha_{\max}$ cm^{-1}	$\Delta\nu_{\frac{1}{2}}$ ev	f_L	f_g	f_{area}
1	10.5×10^{17}	117	0.39	0.41	0.28	
2	7.2×10^{17}	102	0.38	0.51	0.35	
3[a]	5.0×10^{17}	66	0.37	0.46	0.31	
Average				0.46	0.31	

[a] Measured by the torsion method.

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[15] The maximum absorption $\alpha_{\max}$ is lower than indicated in the report by C. V. Heer and C. J. Rauch, *Conference on Low-Temperature Physics, Paris, 1955* (Centrenational de la Recherche Scientifique and UNESCO, Paris, 1956), p. 218, due to a numerical error, and due to a lack of correction for apparent absorption by surface imperfections. These corrections were made by measuring the absorption of uncolored cleaved crystals. This latter correction made the width at half-maximum more nearly the same.

[16] See discussions by C. C. Klick, Phys. Rev. **85**, 154 (1952); reference 9; R. V. Hesketh and E. E. Schneider, Phys. Rev. **95**, 837 (1954) and G. A. Russell and C. C. Klick, Phys. Rev. **101**, 1473 (1956); D. L. Dexter, Phys. Rev. **101**, 48 (1956).

[17] R. H. Silsbee, Bull. Am. Phys. Soc. Ser. II, **1**, 213 (1956).

[18] H. Witt, Nachr. Akad. Wiss. Göttingen **IIa**, 17 (1952).

[19] L. Lin, Phys. Rev. **102**, 968 (1956).

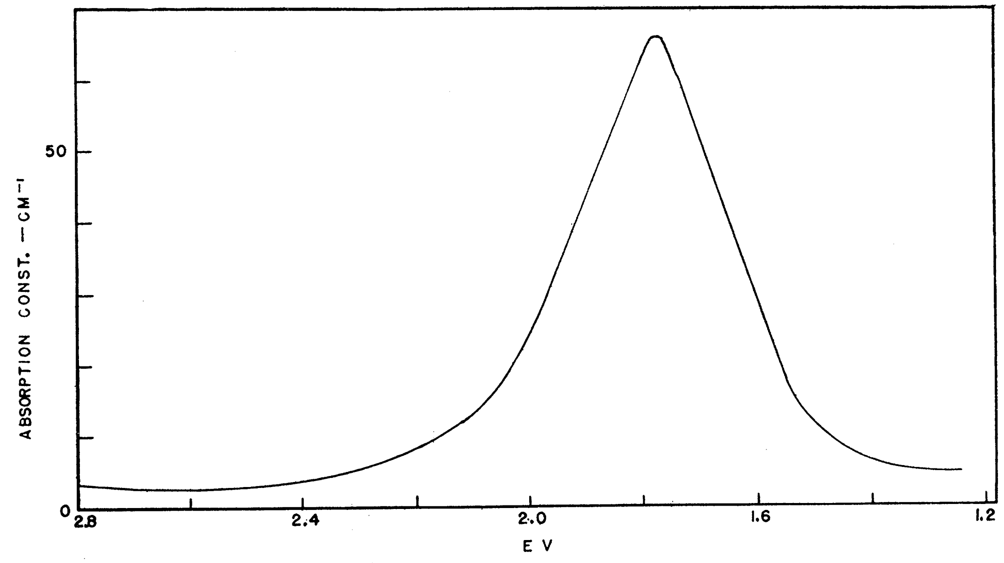

FIG. 7. Optical absorption curve for additively colored KI.

the long-wavelength side, the area under the absorption curve being of the order of 6% for the large crystal shown in Fig. 3, and 3% for the small crystal number 4. If the local field and the matrix element of the electronic transition is similar for the *M* and *F* centers, this would suggest an upward revision of the data by 3 to 6%. A very low value of the oscillator strength $f_L=0.2$ would be required for the *M* center to bring our value for the oscillator strength up to that of Kleinschrod. Experiments to more carefully assess the importance of the *M* center are in progress.

Potassium Bromide

Results for KBr are shown in Table II, and the absorption curve is shown in Fig. 6. The large absorption coefficient for crystal number 1 was estimated from the absorption at half-maximum. In contrast to KCl, the absorption curve is much more nearly Gaussian and possesses a very small amount of *M*-band coloration. The optical oscillator strengths are reasonably consistent over a wide range of concentration, and yield an average value of $f_L=0.71$. A value calculated from the area under the curve is shown for sample number 4. Since all samples have similar absorption curves, their f_{area} are consistent with this value.

Potassium Iodide

Potassium iodide was difficult to color with a great degree of homogeneity for large samples, and small samples deteriorated rapidly after coloration. The data for 3 crystals are shown in Table III and an absorption spectra in Fig. 7. The accuracy of the oscillator strength measurement is not great, and a value of $f_L\approx 0.46$ is suggested. The line shape is again more nearly Gaussian, but the area under the absorption curve is difficult to measure owing to the absorption of other types of coloration, such as the *R*, *M*, and colloid band on the long-wavelength side.

TABLE IV. Cesium bromide.

Crystal	N_m cm^{-3}	$\alpha_{\max}$ cm^{-1}	$\Delta\nu_{\frac{1}{2}}$ ev	f_L	f_g	f_{area}
1	4.3×10^{17}	55	0.32	0.38	0.26	

During the examination of KI, it was found that the coloration disappeared from the edge inward. Upon disappearance of the coloration the paper containers

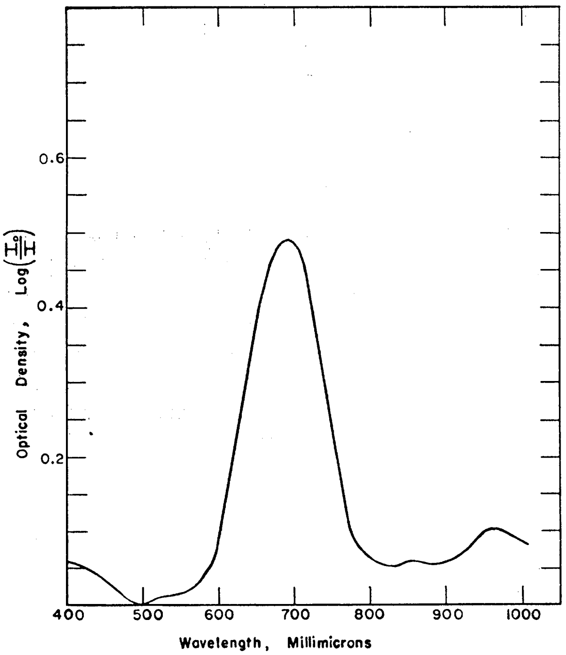

FIG. 8. Optical absorption curve for additively colored CsBr.

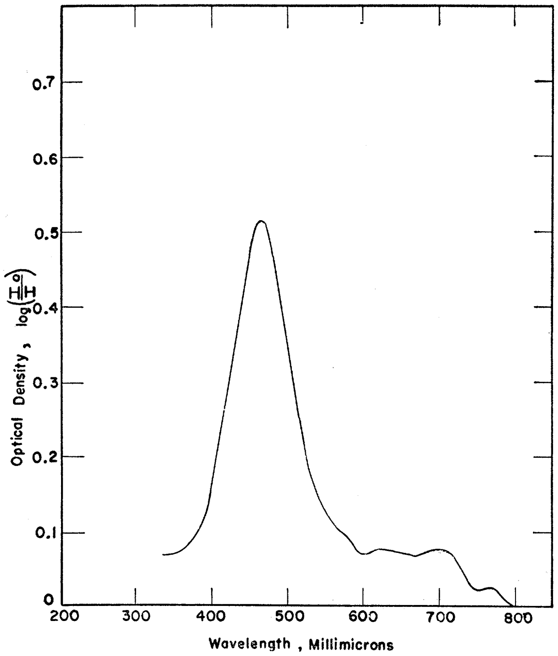

FIG. 9. Optical absorption curve for additively colored NaCl.

would become stained suggesting a chemical reaction with the paper. We examined the large samples for the change of susceptibility with time. No change was observed in a period of 40 hours. Part of the sample was cleaved and the optical absorption was measured. The remainder was stored for a period of one week, cleaved at the end of this time, and then measured optically. The optical measurements for these two specimens indicated no significant change. A small cleaved crystal would indicate a change in optical absorption coefficient of 30% in a period of twenty-four hours. The side band and colloid bands grew during this period.

Cesium Bromide

Cesium bromide was additively colored with potassium vapor. As in the KI crystals, the coloration deteriorated rapidly and measurements were completed in a period of twelve hours after quenching. CsBr crystals remain soft even at nitrogen temperatures and the large colored crystals could not be cleaved. The crystals were sawed, rather than cleaved into small chips, and a large correction for background due to poor surfaces was required for the optical measurements. The data for one of the most homogeneously colored crystals are shown in Table IV. The magnetic data are corrected for the nuclear spin contribution. An oscillator strength of $f_L \approx 0.38$ is indicated. The absorption curve is shown in Fig. 8. In conjunction with these measurements, the diamagnetic susceptibility of pure CsBr was found to be $\chi = -1.35\times 10^{-6}$ emu per cm^3.

TABLE V. Sodium chloride.

Crystal	N_m cm^{-3}	$\alpha_{\max}$ cm^{-1}	$\Delta\nu_{\frac{1}{2}}$ ev	f_L	f_0	f_{area}
1	0.96×10^{17}	12	0.58	0.7	0.47	

Sodium Chloride

Sodium chloride was additively colored with sodium vapor. These crystals were very difficult to prepare, and the absorption of the most homogeneously colored specimen is shown in Fig. 9. The M band as well as other absorption is large, the number of color centers and absorption other than the F band is larger than desirable, and the number of centers sufficiently small to make magnetic measurements difficult. The results for one crystal are shown in Table V. A value of $f_L \simeq 0.7$ is obtained which is in agreement with the value of Pick[20] determined from experiments on the quantum yield for the photochemical reaction of converting F centers to F' centers, but considerably below the value of Silsbee of $f_L = 0.87$ determined from microwave resonance experiments. The magnetic measurements on sodium chloride may be used as an example of the small amount of paramagnetic impurity introduced into the samples by the previously described coloring technique.

CONCLUSIONS AND ACKNOWLEDGMENTS

From the experimental observations and the calculations it appears that one may with some confidence correlate the number of centers measured magnetically with the optical absorption of the F band in additively colored alkali-halide crystals. In all samples the oscillator strengths determined from the area under the curve compared more favorably with the Gaussian line shape than with the Lorentz line shape. The simple model used for these calculations yields an integrated absorption which is dependent on the choice of the local field and the matrix element of the transition or the f value. The f values determined from our work on KI and CsBr, although of a preliminary nature, indicate values considerably smaller than those for KCl and KBr. Since the Lorentz local field is somewhat larger for KI than KCl, the index of refractions are 1.49 for KCl and 1.66 for KI, a considerably smaller matrix element for KI is suggested.

The large difference between the f values for KCl determined by the above techniques $f_L = 0.66$, and those determined by Kleinschrod, $f_L = 0.81$, cannot be explained satisfactorily at this time. Further work on the M band and crystals irradiated with x-rays is being continued.

The authors wish to thank Dr. J. G. Daunt and Dr. Jan Korringa for valuable discussions, and Mr. J. Gerstner for his assistance in the research work.

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[20] H. Pick, Ann. Physik **31**, 365 (1938).