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Dichroism of the F and M Bands in KCl†

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The dichroism of the F and M absorption bands that can be produced by optical bleaching of the F band with polarized light at 87°K is critically dependent on the concentrations of F and M centers. The changes in half-widths and peak positions of the F and M bands that accompany the development of dichroism suggest that the F-band dichroism is due to a spatial association of F and M centers. It is probable that an excited F center can transfer energy to a neighboring M center. Dichroism of the F and M bands can be produced at room temperature by optical bleaching of either band with polarized radiation.

INTRODUCTION

DICHROISM of the M band in KCl after partial bleaching with polarized light has been observed by Ueta.[1] This behavior of the M band is convincing evidence that the M center possesses lower than cubic symmetry. Seitz[2] has suggested the model of the M center that is illustrated in Fig. 1.

The dichroic F band reported by van Doorn and Haven[3] is surprising since there is good evidence that the F center possesses cubic symmetry. Van Doorn and Haven used additively colored crystals containing both F and M centers. After partial bleaching of the F band with polarized light at liquid-nitrogen temperature, the F and M bands were dichroic to polarized light. This behavior was observed only if M centers were present in the crystal. It has been suggested that the observed dichroism of the F band is apparent and due to another absorption band that coincides in peak wavelength and half width with the F band. There has also been speculation that this absorption band is due to an optical transition of the M center from the ground state to an excited state of higher energy than that responsible for the M band.

Lambe and Compton[4] have made a study of the luminescence excited by the absorption of polarized radiation by F, M, R_1, and R_2 centers. A striking observation is that absorption of polarized radiation by the F band produces polarized luminescence from M centers if both F and M centers are present in the crystal. These authors have suggested that an excited F center can transfer its energy to a neighboring M center.

The purpose of the present work was to investigate the development of dichroism in the F and M bands of KCl during bleaching of the F band.

EXPERIMENTAL PROCEDURE

Crystals of KCl were obtained from the Harshaw Chemical Company. They were colored additively in potassium vapor. Samples with dimensions of approximately 6 mm×12 mm×1 mm were cleaved and heated to 430°C in air for several minutes. The samples were cooled rapidly to room temperature and mounted in a cryostat similar to that described by Dutton and Maurer.[5] These operations were performed in the dark.

The crystal samples were then exposed at room temperature to unpolarized radiation which partially bleached the F band and developed the M band. The 546-mμ line of a General Electric Company Type A4 mercury arc and a Wratten 77A filter was used for this purpose.

The optical absorption was measured as a function of wavelength with a Beckman Type B spectrophotometer after cooling with liquid nitrogen. A crystal temperature of 87°K was obtained with liquid nitrogen in the cryostat. This temperature was constant throughout the experiments. Smakula's equation with an oscillator strength of unity was used to calculate the initial concentrations of F and M centers.[6]

The crystals were next irradiated at liquid-nitrogen temperature with 546-mμ radiation that had been linearly polarized with a Polaroid Corporation HN32 polarizer. The radiation was incident normally upon a face of the crystal that was parallel to a (100) plane. It was polarized in the [011] direction. The intensity of the polarized radiation was constant throughout the experiments.

After exposure to the polarized radiation, the optical

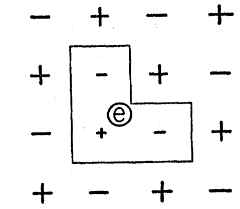

FIG. 1. The Seitz model of the M center. An electron is trapped at a vacancy aggregate consisting of two negative ion vacancies and a positive ion vacancy.

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[1] M. Ueta, J. Phys. Soc. Japan 7, 107 (1952).

[2] F. Seitz, Revs. Modern Phys. 26, 7 (1954).

[3] C. F. van Doorn and Y. Haven, Phys. Rev. **100**, 753 (1955).

[4] J. Lambe and W. D. Compton, Phys. Rev. **106**, 684 (1957).

[5] D. Dutton and R. J. Maurer, Phys. Rev. **90**, 126 (1953).

[6] A. Smakula, Z. Physik **59**, 603 (1930); [see F. Seitz, *Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940), first edition, p. 662].

TABLE I. Concentrations of *F* and *M* centers in KCl crystals.

Crystal	N_F (cm^{-3})	N_M (cm^{-3})
1	1.04×10^{17}	0.73×10^{15}
2	0.99×10^{17}	0.31×10^{16}
3	0.98×10^{17}	0.54×10^{16}
4	0.89×10^{17}	0.74×10^{16}
5	0.92×10^{17}	0.96×10^{16}
6	0.70×10^{17}	0.43×10^{16}
7	0.76×10^{16}	0.43×10^{15}

absorption of the crystals was remeasured using radiation polarized in [011] and [$0\bar{1}1$] directions.

RESULTS

During the exposure of the crystals at room temperature to the unpolarized radiation of wavelength 546 mμ, the magnitude of the *F* band decreased continuously. The R_1 and R_2 bands showed a continuous growth. The *M* band grew during the early stages of exposure but later decreased in magnitude. The ratio of the peak heights of the *M* and *F* bands showed a similar behavior as a function of the exposure time. Table I contains the concentrations of *F* and *M* centers present in the crystals after the bleaching of the *F* band with unpolarized radiation at room temperature. An attempt was made to keep the *F*-center concentration constant and to produce a range of *M*-center concentrations.

The positions of the maxima and the shapes of the *F* and *M* bands change during bleaching of the *F* band at room temperature.[7] Table II shows the position of the maxima and half-maxima of the *F* and *M* absorption bands at 87°K. $\epsilon_m{}^F$ and $\epsilon_m{}^M$ are the photon energies at which the maxima of the *F* and *M* bands occur. ϵ_h and ϵ_l are the photon energies at which the absorption has decreased to half-maximum on the high- and low-energy sides of a band, respectively. During the early stage of bleaching of the *F* band, as the *M* band increases, there is a relative increase in absorption on the low-energy side of the *F* band and a shift of the *F*-band maximum to lower energy. At a later stage in the bleaching of the *F* band, as the *M* band is decreasing, there is a relative increase in absorption on the high-energy side of the *F* band and the maximum of the *F*-band absorption shifts to a photon energy higher than that of the original *F*-band maximum. As the *M* band develops, its maximum shifts to lower energies, and there is a relative increase in absorption on the low-energy side of the band.

Figure 2 shows the effect of bleaching the *F* band of crystal 4 with polarized radiation at liquid nitrogen temperature. The room temperature exposure of this crystal to unpolarized 546-mμ radiation was insufficient to create the maximum number of *M* centers. Curve *A* shows the optical absorption at 87°K after the room temperature bleach of the *F* band. Curves *B* and *C* are the optical absorption for radiation polarized along the [011] and [$0\bar{1}1$] directions, respectively, after the exposure of the crystal at 87°K to radiation of 546-mμ wavelength polarized in the [011] direction. The upper curve *D*, which will be called the dichroic spectrum, is the difference between curves *B* and *C*.

The exposure of the *F* band to polarized radiation decreased the total number of *F* centers and produced a dichroic absorption in the spectral region of the *F*-band absorption. As curve *D* shows, the maximum of this dichroic spectrum does not coincide with the maximum of the *F*-band absorption but lies at a slightly longer wavelength. This "*F* peak" of the dichroic spectrum moved to shorter wavelength as it developed. Appreciable dichroism is observed in the region of the *K* band on the short-wavelength side of the *F* band. The dichroism of the *M* band in the spectral region of the *M* band is of opposite sign to that of the *F*-band region. The maximum of the dichroic spectrum in the *M*-band region coincides with that of the original *M*-band absorption after 3 hours of exposure to polarized light, but, in the earlier stages of bleaching of the *F* band, the maximum of the dichroic spectrum occurs at a smaller energy than that of the original *M* band. It is not possible to decide from the data of Fig. 2 if the total number of *M* centers has altered during the low-temperature irradiation of the *F* band. Auxiliary experiments were performed in which crystals containing *F* and *M* centers were irradiated with unpolarized 546-mμ radiation at liquid-nitrogen temperature. The magnitude of the *M* band was not altered by this procedure. In view of these and Ueta's data, there is

TABLE II. Location of the *F*- and *M*-band maxima and half-maxima at 87°K. ϵ_m is the photon energy of the band maximum; ϵ_h and ϵ_l are the photon energies of the band half-maxima.

Crystal	$\epsilon_m{}^F$ (ev)	$\epsilon_h{}^F$ (ev)	$\epsilon_l{}^F$ (ev)	$\epsilon_m{}^M$ (ev)	$\epsilon_h{}^M$ (ev)	$\epsilon_l{}^M$ (ev)
1	2.283	2.420	2.177	1.530	1.565	1.495
2	2.266	2.415	2.153	1.530	1.569	1.491
3	2.266	2.413	2.157	1.528	1.570	1.485
4	2.280	2.413	2.165	1.528	1.570	1.483
5	2.280	2.413	2.155			
6	2.290	2.440	2.150			

TABLE III. Growth of dichroism in crystal 2 at 87°K. ΔF is the peak height of the *F* dichroic spectrum; ΔM is the peak height of the *M* dichroic spectrum; ϵ_m', ϵ_h', and ϵ_l' are the locations of the maximum and the half-maxima of the *F* dichroic spectrum; ϵ_m'', ϵ_h'', and ϵ_l'' are the locations of the maximum and the half-maxima of the *M* dichroic spectrum.

Exposure time (hours)	ΔF	ΔM	ϵ_m' (ev)	ϵ_h' (ev)	ϵ_l' (ev)	ϵ_m'' (ev)	ϵ_h'' (ev)	ϵ_l'' (ev)
	(optical density)							
0.5	0.035	0.045	2.16_5	2.23_0	2.10_0	1.51_4	1.54_8	1.46_5
1.5	0.067	0.110	2.21_0	2.28_0	2.11_5	1.52_0	1.56_4	1.47_8
3.0	0.090	0.159	2.21_0	2.29_0	2.12_5	1.52_4	1.56_9	1.48_0
13.5	0.140	0.300	2.25_0	2.34_0	2.13_5	1.52_8	1.56_9	1.48_2

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[7] J. D. Konitzer and J. J. Markham, Phys. Rev. **107**, 685 (1957).

little doubt that the irradiation of the *F* band with polarized light has produced a nonuniform distribution of the original *M* centers among the possible directions of their orientation in the crystal.

The general increase in absorption for photon energies less than 2.0 ev indicates that some *F'* centers have been generated. It was observed that, the smaller the number of *M* centers present, the larger was the number of *F'* centers produced by the low-temperature irradiation of the *F* centers.

Table III shows the development of dichroism as a function of time of exposure of the *F* band to polarized radiation. The data were taken with crystal 2 at 87°K. ΔF and ΔM are the peak magnitudes of the dichroic spectrum in the region of the *F* and *M* bands, respectively. ϵ_m is the photon energy at which a maximum occurs in the dichroic spectrum. ϵ_h and ϵ_l are the photon energies at which the dichroic bands fall to half-maximum magnitude. During the growth of dichroism the dichroic bands, in the *F*- and *M*-band spectral regions, shift from lower energies toward the *F* and *M* absorption bands. The dichroic band in the *M*-band spectral region coincides in peak position with *M* band after prolonged exposure, but the peak position of the dichroic band in the *F*-band spectral region remains at a lower energy than that of the peak of the *F* band.

Figure 3 shows data obtained from crystal 2. Curve *A* is the optical absorption at 87°K after the room temperature bleach of the *F* band with polarized light. Curves *B* and *C* are the optical absorption as measured at 87°K with radiation polarized in the [011] and $[0\bar{1}1]$ directions, respectively, after optical bleaching at 87°K with 546-mμ radiation polarized in the [011] direction. A notable development of the *F'* band has occurred. Curve *D*, the dichroic spectrum, is the difference between curves *B* and *C*. In addition to curve *D*, the dichroic spectrum is illustrated at three intermediate stages of its development in order to show the progressive shift of the *F*- and *M*-band maxima to higher photon energies during the bleaching of the *F* band with polarized radiation.

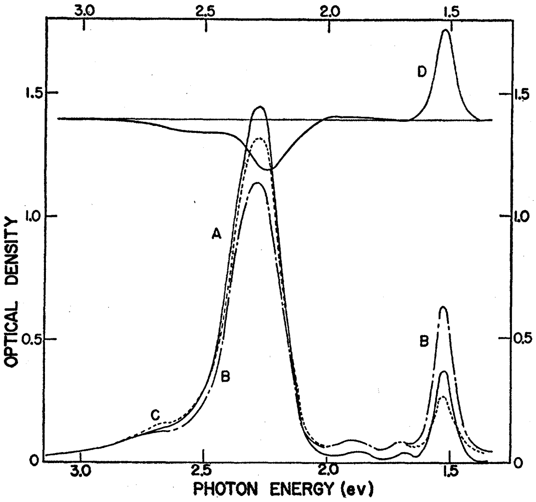

FIG. 2. Dichroism in KCl (crystal 4) after exposure of the *F* band to polarized radiation for five hours at 87°K. (A) Absorption before irradiation. (B) Absorption after exposure as measured with radiation polarized in the [011] direction. (C) Absorption after exposure with radiation polarized in the $[0\bar{1}1]$ direction. (D) Difference between (B) and (C) or dichroic spectrum. Crystal thickness 0.53 mm.

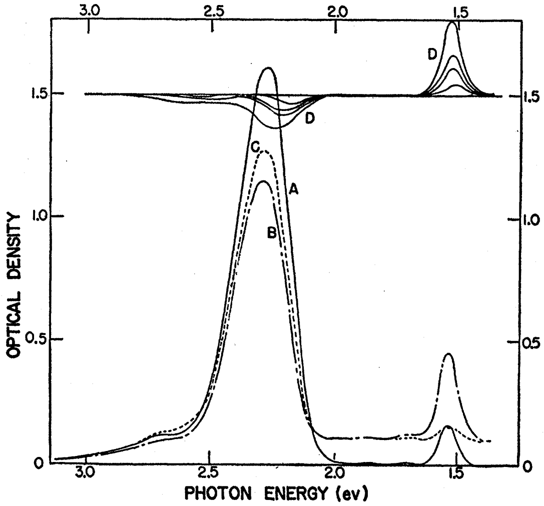

FIG. 3. Dichroism in KCl (crystal 2) after exposure of the *F* band to polarized radiation at 87°K. (A) Absorption before irradiation. (B) After irradiation as measured with radiation polarized in the [011] direction. (C) After irradiation as measured with radiation polarized in the $[0\bar{1}1]$ direction. (D) Difference between (B) and (C), the dichroic spectrum. The dichroic spectrum is also illustrated at three intermediate stages of its development. Crystal thickness 0.53 cm.

Figure 4 shows the behavior of the quantity $\Delta M/M_0$ as a function of time of exposure to polarized radiation at liquid-nitrogen temperature. ΔM is the peak height of the dichroic spectrum in the *M*-band spectral region. M_0 is the peak height of the original *M* band. The dichroism of the *M* band develops very rapidly during the initial irradiation of the *F* band with polarized light at low temperature but saturation occurs eventually. The value of $\Delta M/M_0$ for a given exposure time is critically dependent on the concentration of *M* centers. It increases with increasing concentration of *M* centers if the *F*-center concentration is constant as shown by the data of Fig. 4. The properties of the crystals used to obtain the data of Fig. 4 are summarized in Table I.

If the maximum development of the *M* band by exposure of the *F* band to radiation at room temperature is exceeded, the dichroism is reduced as shown by the relative position of the curve for Crystal 6 of Fig. 4.

Crystal 1 was cooled from 430°C to liquid-nitrogen temperature without exposure to 546 mμ radiation at room temperature. Despite the omission of the room temperature bleach of the *F* band, there was a small but detectable concentration of *M* centers. The original concentrations of *F* and *M* centers were 1.0×10^{17} cm^{-3}

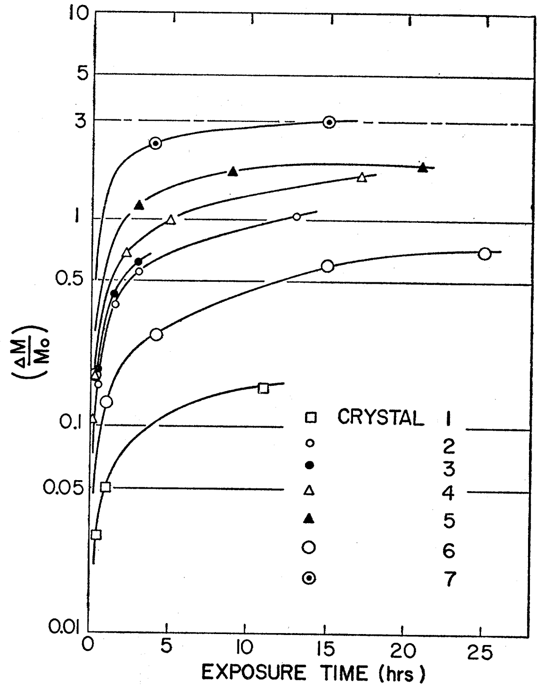

FIG. 4. Growth of dichroism in the *M* band.

and 0.7×10^{15} cm^{-3}, respectively. As Fig. 4 shows, the dichroism developed in this crystal was smaller than that occurring in the other crystals. There was a considerable destruction of *F* centers and generation of *F′* centers during the exposure to polarized radiation at low temperature.

Crystal 7 contained about 10^{16} *F* centers/cm^3. The exposure of the crystal at room temperature was chosen so as to develop the maximum concentration of *M* centers. The maximum concentration of *M* centers that could be obtained in such a crystal was only one twentieth of the maximum concentration obtainable in a crystal containing 10^{17} *F* centers/cm^3.

The ratio of the dichroism of the *F* band to that of the *M* band (expressed as the ratio $\Delta F/\Delta M$, where ΔF is defined in a manner similar to ΔM) is shown in Fig. 5 as a function of time of exposure at liquid nitrogen temperature to polarized 546-mμ radiation. $\Delta F/\Delta M$ decreases from a value near unity to an asymptotic value of 0.5 as the time of exposure increases. The behavior of crystal 1 was exceptional. $\Delta F/\Delta M$ for this crystal, which contained the smallest concentration of *M* centers, remained near unity during the entire time of exposure.

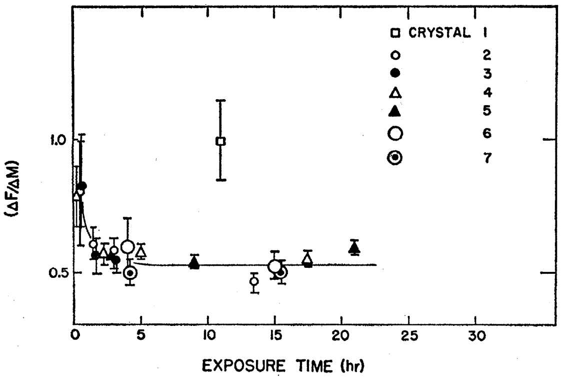

FIG. 5. The ratio $\Delta F/\Delta M$ *versus* time of exposure.

The thermal stability of the dichroism was studied by warming the crystals to temperatures above 87°K and remeasuring the optical absorption after recooling to 87°K. Approximately a tenth of the dichroic spectrum in both the *F*- and *M*-band spectral regions disappeared on warming to 130°K, if the original irradiation of the *F* band with polarized light did not exceed a few hours. If the irradiation of the *F* band with polarized light exceeded 10 hours, this partial disappearance of the dichroism did not occur. A substantial part of the dichroism persisted in all cases to 400°K where the *M* band itself began to disappear.

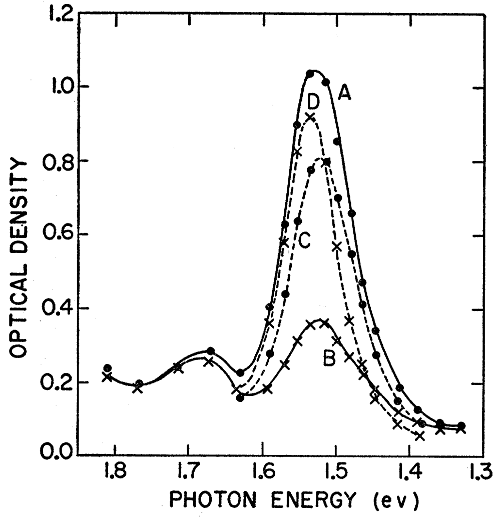

FIG. 6. The change in dichroism resulting from a nine-hour exposure at 87°K to unpolarized 546-mμ radiation. *A* and *B*, before exposure. *C* and *D*, after exposure. *A* and *D* were measured with radiation polarized in the [011] direction. *B* and *C* were measured with radiation polarized in the [$0\bar{1}1$] direction. Original dichroism produced by [011] radiation.

The optical stability of the dichroism was tested by irradiating the crystals with unpolarized 546-mμ light at 87°K. Two stages in the disappearance of the dichroism could be distinguished. The difference between the absorption maxima for the two perpendicular directions of polarization became smaller but the two absorption bands were displaced with respect to each other. A much longer exposure to unpolarized radiation brought the absorption bands, as measured with the two directions of polarization, into coincidence. Figure 6 shows the change in the *M* absorption bands produced by a 9-hour exposure of the *F* band to 546-mμ unpolarized radiation. The *M* absorption band for radiation polarized in the [011] direction is displaced in the direction of smaller photon energy relative to the *M* absorption band as measured with radiation polarized in the [0$\bar{1}$1] direction. In the *F*-band region the relative position of the absorption bands, as measured with these two directions of polarization, was the reverse of that shown for the *M* band.

Dichroism of the *F* and *M* bands was obtained on irradiation at room temperature of crystals containing *F* and *M* centers with polarized 546-mμ radiation. Dichroism of the *F* and *M* bands was observed also on irradiation with polarized light in the *M* band at room temperature. The dichroism observed in these two types of experiments was similar and possessed important features that differed from the dichroism produced at liquid nitrogen temperature. Figures 7 and 8 illustrate the dichroism produced at room temperature.

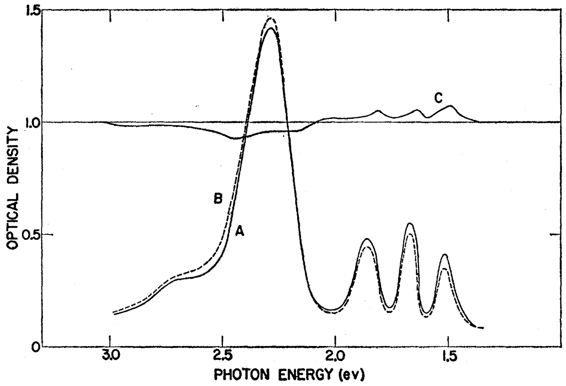

FIG. 7. Dichroism produced at room temperature by exposure to 546-mμ radiation polarized in the [011] direction. Absorption measured at 87°K. (A) Measured with radiation polarized in the [011] direction. (B) Measured with radiation polarized in the [0$\bar{1}$1] direction. (C) Difference between (A) and (B). Crystal thickness 0.056 cm.

The magnitude of the dichroism produced at room temperature was always small relative to that produced at low temperature. The dichroic spectrum in the spectral region of the *F* band was broader than at low temperature and there was a remarkable relative increase of dichroism on the high-energy side of the *F* band. As a result, the room temperature "*F*-band" dichroism had its peak magnitude at a shorter wavelength than the peak of the *F* band. Dichroism of the same sign as that of the *M* band was observed in the R_1 and R_2 bands.

An unsuccessful search was made for the development of ultraviolet absorption bands during the room temperature development of the *M* band and bleaching of the *F* band with unpolarized radiation in an effort to discover a high-energy optical transition of the *M* center.

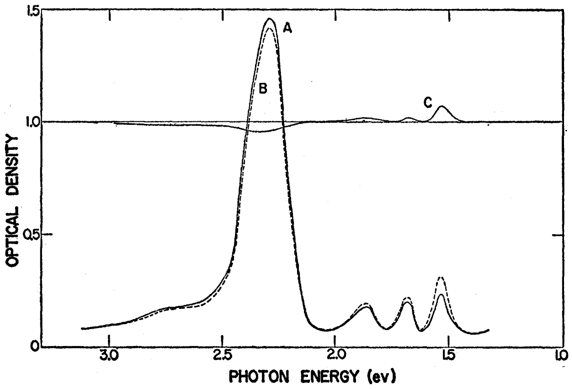

FIG. 8. Dichroism produced at room temperature by exposure to 830-mμ radiation polarized in the [011] direction. Absorption measured at 87°K. (A) Measured with radiation polarized in the [011] direction. (B) Measured with radiation polarized in the [0$\bar{1}$1] direction. (C) Difference between (A) and (B). Crystal thickness 0.042 cm.

DISCUSSION

Inui, Uemura, and Toyozawa,[8] who assumed Seitz's model of the *M* center, have concluded that the *M* band is the result of a dipole transition. The six possible orientations of the dipole axis along the face diagonals of the crystal lattice are illustrated in Fig. 9. The optical transition probability of the *M* center for polarized radiation is proportional to $\cos^2\theta$, where θ is the angle between the dipole axis and the direction of polarization. If the *M* centers are oriented with equal concentration, n_0, in each of the six possible directions, the absorption coefficient for unpolarized radiation is

$$K_0 = 2\alpha n_0, \qquad (1)$$

where α is a constant. After irradiation of the crystal with radiation polarized in the [011] direction (direction 1 of Fig. 9), the absorption coefficient for radiation polarized in this direction is

$$K_{\parallel} = \alpha(n_1 + n_3), \qquad (2)$$

where n_1 is the concentration of *M* centers with axes in direction 1 and n_3 is the concentration of *M* centers with axes in each of the directions 3, 4, 5, and 6. The absorption coefficient for radiation polarized in the [0$\bar{1}$1] direction (direction 2 of Fig. 9) is

$$K_{\perp} = \alpha(n_2 + n_3), \qquad (3)$$

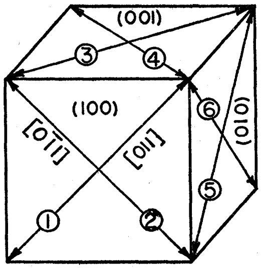

FIG. 9. Possible orientations of the *M*-center axis in KCl.

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[8] Inui, Uemura, and Toyozawa, Progr. Theoret. Phys. (Japan) 8, 355 (1952).

and

$$K_{\parallel}-K_{\perp}=\alpha(n_1-n_2). \qquad (4)$$

Since it was found experimentally that the total number of M centers was not changed during the low-temperature irradiation of the F band, one has

$$\frac{n_0-n_3}{n_0}=\frac{K_{\parallel}+K_{\perp}-2K_0}{K_0}. \qquad (5)$$

It was also found experimentally that $[K_{\parallel}+K_{\perp}-2K_0]$ was always positive so that n_3 is less than n_0. The concentration of M centers oriented in the directions 3, 4, 5, and 6 of Fig. 9 decreased therefore during the exposure of the F band to radiation polarized in direction 1.

If we assume that during exposure of the F band to polarized radiation, the concentrations of M centers that are oriented in a given direction change in the following simple manner:

$$\frac{dn_1}{dt}=-\left[\frac{dn_2}{dt}+4\frac{dn_3}{dt}\right]; \quad \frac{dn_2}{dt}=-\beta_2 t; \quad \frac{dn_3}{dt}=-\beta_3 t, \qquad (6)$$

we obtain

$$\frac{K_{\parallel}-K_{\perp}}{K_0}=3-[\exp(-\beta_2 t)+2\exp(-\beta_3 t)], \qquad (7)$$

where $(K_{\parallel}-K_{\perp})/K_0$ is the quantity $\Delta M/M_0$ that is shown in Fig. 4.

If the dichroism in the region of the F band is due to an optical transition of the M center, the ratio $\Delta F/\Delta M$ should remain constant during the exposure of the F band to polarized radiation. This conclusion follows from Eq. (4) since, according to this assumption, both ΔF and ΔM are proportional to (n_1-n_2). Since Fig. 5 shows that $\Delta F/\Delta M$ decreases during exposure, another explanation of the dichroism in the F-band region must be found.

The assumption that the reorientation of the M-center axes is described by Eqs. (6) is inconsistent, also, with the assumption that the polarized radiation of wavelength 546 mμ is directly absorbed by the M center. The quantity $\Delta M/M_0$ depends, in this case, only upon the constants β_2 and β_3 in the manner described by Eq. (7). $\Delta M/M_0$ should be independent of the concentration of M centers and should saturate at the value 3 for all crystals. As Fig. 4 shows, this behavior is not observed. $\Delta M/M_0$ increases with increasing M-center concentration and decreases with increasing F- or R-center concentration.

The suggestion of Lambe and Compton that an exchange of energy is possible between F and M centers is in better agreement with the data. M centers lying near F centers may distort the F centers and permit them to exhibit dichroic absorption. A slight shift of the absorption band of these special F centers with respect to the absorption band of normal F centers is plausible.

A relative increase in absorption is observed on the low-energy side of the F and M bands during optical bleaching of the F band with unpolarized light at room temperature. The development of dichroism on the low-energy side of both bands is a prominent feature of the early stages of the growth of dichroism. These observations suggest that M centers that are in the immediate neighborhood of F centers are reoriented during the early stages of bleaching while M centers that are located at a greater distance from F centers participate in the later stages of the process.

The fact that $\Delta M/M_0$ saturates at a value less than 3, as predicted by Eq. (7), can be accounted for by assuming that a concentration n_s of M centers with axes in a given direction interacts with F centers. The fraction of these M centers is n_s/n_0 and the saturation value of $\Delta M/M_0$ is $3n_s/n_0$. It is necessary, however, to assume that n_s is not proportional to n_0 in order to explain the dependence of $\Delta M/M_0$ on the original concentration of M centers. It appears that there must be more than one M center situated in the immediate vicinity of an F center if an energy transfer from F to M centers is to occur. The fact that the ratio $\Delta F/\Delta M$ is less than unity also leads to the conclusion that the number of F centers affected by the reorientation of M-center axes is less than the number of reoriented M centers. In this connection it is interesting that the exceptional crystal, for which $\Delta F/\Delta M$ was unity throughout the exposure, had a small concentration of M centers that were not produced by the bleaching of F centers at room temperature. The spatial relationship of M to F centers in this crystal may have been quite different from that existing in the other crystals.

Why the crystal containing the smallest concentration of F centers (10^{16} cm^3) should have exhibited the largest saturation value of $\Delta M/M_0$ is not clear.

The dichroism that was observed on the high-energy side of the F band in the spectral region of the K band raises difficulty for the interpretation of the K band as due to an optical transition of the F center. The peak height of the dichroic spectrum in the region of the K band was about one-fourth the peak height of the dichroic spectrum in the region of the F band. This ratio is much larger than the ratio of the peak height of the K band to that of the F band, which was found to be about 0.05 by measurements with unpolarized light.

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