PHYSICAL REVIEW VOLUME 127, NUMBER 5 SEPTEMBER 1, 1962

Color Centers in Additively Colored CsBr*

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(Received April 30, 1962)

Color centers have been studied in CsBr single crystals additively colored with potassium or cesium. More than ten absorption bands were produced, most of which had previously been found in crystals colored by ionizing radiation. All bands produced by irradiation at or above liquid nitrogen temperature appear to arise from trapped electrons. The most prominent bands in the additively colored crystals are the F band at 642 mμ (liquid nitrogen temperature) and a band at 243 mμ which appears to be due to an impurity. Optically bleaching the F band at 200°K produces bands analogous to the M and R bands in other alkali halides while bleaching at liquid nitrogen temperature yields a broad band identified as the F' band. Evidence that the asymmetry of the F band arises from several excited states of the F center is presented.

INTRODUCTION

COLOR centers have been studied in many alkali halides but primarily those of the NaCl structure. It is of interest to extend these studies to alkali halides of the CsCl structure to see if the different lattice structure has any effect on the color centers. Such a study can be aided by the large body of knowledge in existence on color centers in NaCl-type salts and also may provide a test for some of the color center models currently used. Smakula and Avakian,[1] and Rabin and Schulman[2] have reported on color centers in irradiated cesium halides and the latter workers have also examined the F band in additively colored CsBr. Avakian and Smakula observed at least ten absorption bands in irradiated CsBr, the two most prominent bands being at 241 mμ and 646 mμ at liquid nitrogen temperature (LNT). The latter band, corresponding to the F band in other alkali halides, is asymmetric, Rabin and Schulman having decomposed it into three components at liquid helium temperature. In CsCl they find the corresponding band also has three components.[2] The present work was undertaken to determine which of the bands of reference 1 result from trapped electrons and to study further the properties of some of these bands.

Recently two more papers on color centers in cesium halides have appeared.[3,4] The work of Rabin and Schulman deals primarily with color centers produced by ionizing radiation, including the result of irradiating at liquid helium temperature. The work of Hughes and Allard describes the electron spin resonance spectra of the F center and a hole center in CsCl.

EXPERIMENTAL

Harshaw and Korth single crystals were used. These crystals showed traces of thallium absorption bands[5] ($\alpha\sim0.1$ cm^{-1}, $\lambda=260$ mμ, and 212 mμ at LNT) and evidence of hydroxyl ions (see later). They also contained from 100 to 1000 ppm of Na, 100 ppm of Rb, and 50 to 100 ppm of K. Ag, Cu, Fe, Mg, and Si were also detected spectrographically. The absorption at wavelengths less than 260 mμ was not characteristic of pure crystals. There was structureless absorption to at least 220 mμ, the limit of reliable measurement.

The crystals were colored for 12 h at 500°C with potassium or cesium vapor whose equilibrium temperature was between 350 and 450°C. All bands except a colloid-like band appeared to be independent of the two metals used for coloring. After polishing, the 1 to 3 mm thick crystals were heated in air to 500°C for 1 to 2 min on a platinum-clad steel block and quenched on a copper block. During and after this procedure the only light allowed on the crystals was from a 15 W lamp covered with several layers of blue cellophane transmitting between 350 and 500 mμ, and beyond 1000 mμ. Optical absorption measurements were made on a Beckman DU spectrophotometer. Using an oscillator strength of[6] 0.26 and assuming a Gaussian shape for the entire F band gave color center densities between 3×10^{16} and 1.6×10^{17} cm^{-3}.

RESULTS

Figure 1 shows the absorption spectrum of an additively colored crystal about a half hour after quenching.

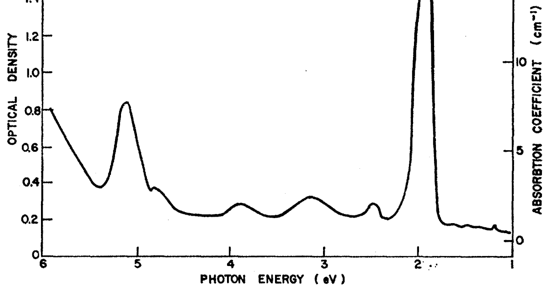

FIG. 1. Absorption spectrum of quenched additively colored CsBr taken at liquid nitrogen temperature.

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[*] Contribution No. **1110**. Work was performed at the Ames Laboratory of the U. S. Atomic Energy Commission.

[1] P. Avakian and A. Smakula, Phys. Rev. **120**, 2007 (1960).

[2] H. Rabin and J. H. Schulman, Phys. Rev Letters **4**, 280 (1960).

[3] H. Rabin and J. H. Schulman, Phys. Rev. **125**, 1584 (1962).

[4] F. Hughes and J. G. Allard, Phys. Rev. **125**, 173 (1962).

[5] M. Forró, Z. Physik **58**, 613 (1929).

[6] C. J. Rauch and C. V. Heer, Phys. Rev. **105**, 914 (1957).

TABLE I. Spectral positions and widths of absorption bands in CsBr.

Additively colored[a]		Irradiated[b]		
Present work		Reference 1		Reference 3
Wavelength (λ) of maximum (mμ)	Full width (W) at half maximum (eV)	λ (mμ)	W (eV)	λ (mμ)
				205[c]
224	0.13			
239	0.15			235[d]
243	0.25	241	0.25	245[d]
258	0.1			
266	0.1	270	0.3	
				280
322	0.1	315	0.5	320
390	0.3	390	0.35	395
				450[d]
481–500	0.1	480		480[d]
				570[e]
646	0.22	646	0.20	645
700				
761		780		
821		840		
910	0.07	930	0.1	
1045	0.03	1055	0.04	1050

[a] Measured at LNT.
[b] Irradiated and measured at LNT.
[c] Outside the wavelength region of the present investigation.
[d] Not found in all crystals by Rabin and Schulman.
[e] Observed in the present investigation, but included in the 646-mμ band.

Nine of the ten bands observed by Avakian and Smakula[1] in irradiated CsBr are seen here. (Their λ_9 was a weak band on the long-wavelength side of the F band, suggesting that it is a band similar to the R bands in other alkali halides.) Table I lists the characteristics of these bands and compares them with the bands of references 1 and 3. Other characteristics of these bands are described below.

224-mμ band This band does not occur in all crystals but was found in some Harshaw crystals which exhibited a step at 225 mμ in the absorption spectrum before coloration. The absorption at 224 mμ was increased by the coloring process (not just the quenching), and the band was several times higher than the original step. Since it did not occur in all crystals, this band is probably due to an impurity. "Not occurring in all crystals" means that the bands were found in all samples cut from the same block of crystal, but *not* in samples cut from other blocks. Two Harshaw crystals, purchased at different times, gave different ultraviolet absorption bands after additive coloration. These bands usually occurred in different samples from one block despite different thermal histories and coloration levels.

239-mμ band In some crystals grown in the laboratory from Dow CsBr a band at 239 mμ occurred upon additive coloration and quenching. These crystals contained about ten times as much thallium as the Harshaw or Korth crystals and were probably less pure with respect to other impurities. This band is due to an impurity but probably not thallium since additive coloration to low levels left the height of the 260-mμ thallium band unchanged.

243-mμ band This band appears prominently in all crystals, but has a different shape in different crystals due to overlap with the Tl band and probably the band at 239 mμ. If a colored crystal is cooled slowly from the temperature of coloration only the 243-mμ band and a "colloid" band occur. Irradiation in the 243-mμ band at room temperature bleaches it slowly giving rise to the F band. Subsequent optical bleaching of the F band leads to the normal products of such a bleach; the 243-mμ band is not restored. Irradiation at LNT bleaches the 243-mμ band very slowly (3 h with 2-mm slits on spectrophotometer removed 12%), but no new bands produced by this bleach could be found. When, instead of being optically bleached, the slowly cooled crystal was reheated and quenched, a large F band occurred and the 243-mμ band was smaller. In the thinner samples quenching removed the 243-mμ band completely.

This band might be the U band (due to substitutional H^- ions) because its behavior closely resembles that of the U band in other alkali halides. Etzel and Patterson[7] showed that the U band was always present if crystals containing OH^- ions were additively colored. No OH^- absorption could be resolved in these crystals with the present equipment but the absorption in the tail of the fundamental absorption band was somewhat high (Figs. 1 and 7 of reference 1 show a slight shoulder at 225 mμ and 240 mμ, respectively, the former in the region where OH^- absorption would be expected). However the introduction of hydrogen into the colored crystals by heating them at 500°C in hydrogen at several atmospheres pressure until the blue color disappeared failed to yield any new absorption in the ultraviolet, so the identification of the 243-mμ band as the U band cannot be made with certainty. The band is somewhat more removed from the absorption edge than is the U band in other alkali halides.

Two different crystals showed a band at 243 mμ having twice the normal width. There was a trace of a shoulder on the short wavelength side of this band in these crystals. In these crystals an unusually large 239-mμ band probably occurred simultaneously with the 243-mμ band.

258- and 266-mμ bands These small bands do not occur in all crystals and are probably due to impurities. Tl gives a band at 260 mμ in uncolored crystals, which remains after coloration. No band at 280 mμ was seen. The width of the *270-mμ* band of reference 1 (Table I) may be attributed to the overlap with the 258-mμ band.

322- and 390-mμ band. These are not present in all crystals and are very weak in all cases. There is a considerable difference in the widths reported for the 322-mμ band (Table I).

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[7] H. W. Etzel and D. A. Patterson, Phys. Rev. **112**, 1112 (1958). See also F. Kerkhoff, Z. Physik **158**, 595 (1960).

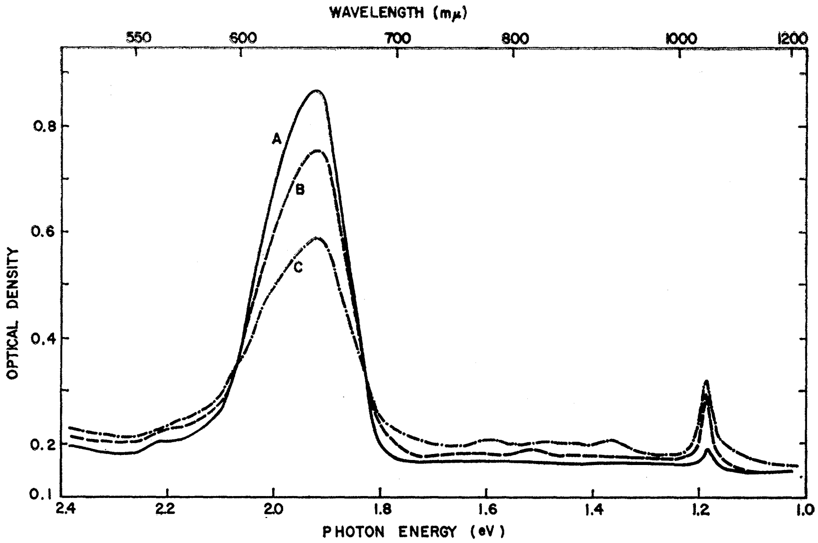

FIG. 2. Result of bleaching at dry ice temperature with F-band light. Spectra measured at liquid nitrogen temperature. A. No bleach. B. 5-min bleach. C. 60-min bleach. Bleached at 645 mμ with 0.1-mm slits on spectrophotometer.

500-mμ band This band was found in all crystals but with varying degrees of prominence. It could be bleached by 500-mμ light at room temperature but not at LNT. At both temperatures the 500-mμ light reduced the F band, but this could be due to a tail on the F band extending to 500 mμ. The 500-mμ band occurred upon slow cooling of the crystal and was made more prominent by quenching. There was no correlation between the height of the 500-mμ band and that of the F band. In some crystals this band peaked at 480 mμ instead of 500 mμ. This band was present in all crystals and may be caused by a genuine color center, although there are impurities common to all samples. Further study, especially of the dependence of this band on the thermal history of the sample, is needed.

642-mμ band This band will be called the F band because of its similarity to the F band in other alkali halides. Rabin and Schulman[2,3] have presented arguments for identifying this band as the F band. They have shown that this band possesses structure and could consist of three components. Analogous decomposition of this band can be made at LNT and the relative areas of the components are about the same as in reference 3 for liquid helium temperature (65:30:5 for the 1.905, 2.01, and 2.08 eV components at LNT). The shape of the F band at about 15°K agrees with that obtained by Rabin and Schulman. At dry ice temperature (DIT) and room temperature (RT) the sub-bands overlap so severely that such a decomposition is useless. The high-energy component[8] lies at the position of the K' band that Drickamer *et al.*[9,10] find in CsBr under pressure. They find that as the pressure is increased the F band diminishes and the K' band grows. Illumination in either the F or K' band bleaches both bands. In other alkali halides such as KI, the K' band appears at pressures sufficient to make the CsCl structure more stable. The "K' component" of the F band appears to be a property of the CsCl lattice, not of the Cs nearest neighbors to the F center.

The F band in a freshly quenched crystal decays slowly in the dark. A quenched crystal subsequently heated in the dark to 90°C for 18 h showed a smaller F band and an increased "colloid" band. The F band can be optically bleached rapidly at RT, the ultimate product being a "colloid" band although thermally unstable R- and M-like bands can be detected. (However, see the section on the "colloid" band.) At DIT the optical bleach yields the R- and M-like bands (see

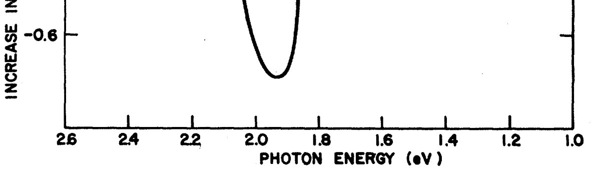

FIG. 3. Result of bleaching at liquid nitrogen temperature with F band light. The optical density after the bleach minus the optical density before the bleach is plotted. The plot shows the F' band created and the F band lost.

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[8] Rabin and Schulman[3] list this component separately. See Table I. There is a possibility that this component is not part of the F band in CsBr. This is discussed further in the present paper and in reference 3.

[9] W. G. Maisch and H. G. Drickamer, J. Chem. Phys. Solids **5**, 328 (1958).

[10] R. A. Eppler and H. G. Drickamer, J. Chem. Phys. **32**, 1418 (1960).

Fig. 2) while a bleach at LNT gives a broad band analogous to the F' band (Fig. 3).

Partially bleaching the F band in an oriented crystal with polarized light produced no 100, 110, or 111 dichroism in the F band. This bleach took place at LNT with light in the long wavelength component of the F band.

1045-mμ band This band formed upon optical bleaching of the F band at DIT-RT (Fig. 2), but disappeared in several hours in the dark at RT. It was occasionally found in unbleached crystals, but this was attributed to inadvertent optical bleaching of the F band. It appears to correspond to the M band in other alkali halides. The full width at half maximum is about 0.026 eV at LNT and about 0.017 eV at about 15°K. Although the band is narrow, it still shifts considerably to higher energy with decreasing temperature (see Table II). Because of its narrowness, accurate determination of the M band shape is difficult, but it appears not to be Gaussian, being skewed somewhat toward long wavelengths, and perhaps having a long wavelength tail. The band bleaches optically at DIT, partially restoring the F band and also reducing the band at 821 mμ (when present) formed by the F bleach. It cannot be bleached at LNT. Figure 2 also shows that as the M band grows, the asymmetry of the F band grows. There appears to be a second and possibly a third transition of the M center[11,12] at 2.0 and 2.08 eV or a new center formed by F-light bleach.

821-mμ band This band grows slowly upon DIT F bleach after a prominent M band has formed. It is unstable at RT and contributes to the restoration of the F band.

700, 761, and 905 mμ bands These small bands are produced by prolonged F light bleach at DIT, are also unstable at RT, and are smaller than the 821-mμ band. The band at 700 mμ occasionally formed upon F light bleaching at RT and LNT. It appeared similar to the 615-*mμ* band in CsCl (Fig. 3 of reference 3). Conditions for its occurrence were not determined.

F' band Irradiation in the F band at LNT produced the changes shown in Fig. 3. The broad band peaking at about 775 mμ but extending from 480 mμ to above 1200 mμ appears analogous to the F' band in other alkali halides. None of the bands on the red side of the F band produced by F light at DIT occur upon LNT F irradiation. The F' band bleaches with 820 mμ light at LNT and in the dark at DIT, the F band being restored in each case. If an α band occurred during F' production, its peak was at $\lambda < 215$ mμ.

TABLE II. Spectral position and width of the M band in CsBr.

Approximate temperature (°K)	Wavelength of maximum (mμ)	Full width at half maximum (eV)
15	1037	0.017[a]
90	1045	0.026–0.034
200	1057	0.055
300	~1080	~0.08

[a] Two measurements only.

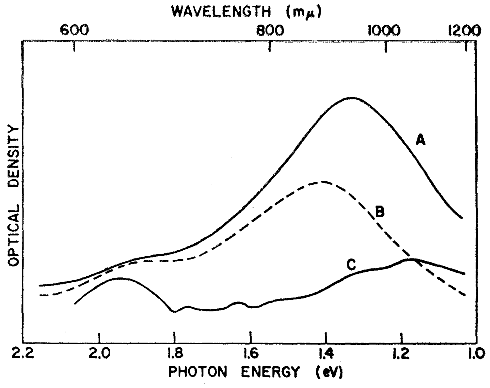

FIG. 4. Absorption due to "colloidal" centers, measured at liquid nitrogen temperature. The optical density scale is different for each crystal. A. Crystal colored with potassium and "slowly" cooled from coloration temperature (no quench). B. Potassium colored crystal, quenched, then bleached in the F band at room temperature. C. Crystal colored with cesium, treated as the crystal of curve B.

"Colloid" band When the crystals are cooled slowly to room temperature after coloration or are exposed to F light for long periods of time at room temperature, the only prominent bands are the 243-mμ band, a weak F band, and a broad band with a maximum between 850 and 990 mμ (potassium colored crystals). (See Fig. 4.) This latter band is relatively stable against optical bleach. In crystals colored with cesium this "colloid" band peaked at 1050 mμ and did not form as quickly upon RT F-light irradiation as in potassium colored crystals. The weak bands at 700, 761, 821, and 905 mμ were produced during such a bleach of a cesium-colored crystal. If the crystals are kept in the dark for a long time (weeks) after RT optical production of the "colloid" band, the F band grows at the expense of the "colloid" band, about one-third of the original F band being restored. Because of this restoration of the F band, the "colloid" band is probably not a true colloid band but arises from small aggregates of F centers and is analogous to the R' band of Scott and Bupp.[13]

DISCUSSION

The present work shows that nearly all bands produced by irradiation at or above LNT arose from trapped electrons. This can be seen from Table I where nearly all bands which occur in all irradiated crystals are seen to occur in at least some additively colored crystals. The exceptions are the 205-mμ band and possibly the 280-mμ band. This latter band may be the 266- or 270-mμ band in the other columns of Table I. With these possible exceptions no V bands were found

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[11] T. Tomiki, J. Phys. Soc. Japan **15**, 488 (1960).

[12] F. Okamoto, Phys. Rev. **124**, 1090 (1961 .

[13] A. B. Scott and L. P. Bupp, Phys. Rev. **79**, 341 (1950).

at LNT by Avakian and Smakula,[1] and by Rabin and Schulman.[2,3] Jacobs *et al.*[14] have additively colored CsBr with bromine and found a band at 275 mμ which may be a V band. This band does not correspond to a prominent band in irradiated CsBr reported by others, although it lies at the position of a large band produced by x-irradiation by Jacobs and co-workers. Data taken at low temperature on bromine-colored CsBr after proper quenching are needed to relate this band to V centers. Fairly large bands at 435 and 490 mμ and a smaller band at 340 mμ were produced by Rabin and Schulman[3] by irradiation at liquid helium temperature. These bands do not occur in additively colored crystals and could arise from trapped holes.

An important feature of the F band is its substructure. Rabin and Schulman cite evidence for the structure's being due to several transitions of one center. Additional evidence in favor of this view is the fact that bleaching the F band at LNT gives an F' band and a reduced F band with the same shape as before. Bleaching the F' band restores the F band with its original shape. This eliminates the possibility that part of the F-band structure arises from another center whose concentration is proportional to the F center concentration, e.g., the F_2^+ center,[15] unless vacancies or F centers are mobile at LNT. Production of the F band with the same structure by irradiation at liquid helium temperature also speaks against this reason for the structure.

The accepted model for an F center in other alkali halides is an electron trapped at an anion vacancy. The F band arises from transitions from a $1s$ state to the degenerate $2p$-like states. The possibility that in the cesium halides the structure is due to $1s$-$2p$, $1s$-$3p$, $1s$-$4p$ (or $1s$-continuum) transitions is unlikely because the oscillator strengths should diminish for the higher energy transitions and this is not observed. In CsCl the three sub-bands have about equal areas.[3] In CsBr the second component has about half the area of the first, but this could be assumed to be two unresolved bands of equal area, making three components of equal area as in CsCl. The structure could then arise from a transition to a $2p$ state split into three levels. (This decomposition of the F band in CsBr ignores the 580 mμ component (Drickamer's K' band). Because this component is so small and is overlapped by the larger components of the F band it is impossible at present to know whether the 580-mμ absorption arises from the F center.)

If all the F centers were sufficiently perturbed the degeneracy of the p-like first excited states would be lifted and structure in the F band would be expected. This splitting could lead to more than one luminescent emission band from the center and the emission should be polarized upon excitation with polarized light. A perturbation with sufficiently low symmetry to split the p state into three levels in all crystals of the CsCl structure is puzzling.

The experiment with polarized light appears to eliminate two possibilities for such a perturbation: the proximity of a dislocation to all F centers and the proximity of impurities to all F centers to form centers like the A center.[16] Spectrographic analysis revealed enough sodium and potassium in all crystals to associate several ions of these impurities with each F center at all levels of coloration. The work of Drickamer also tends to eliminate impurities as the cause of the K' part of the structure. He observed the K' band in Harshaw KI and KBr, both presumably purer than CsBr, as soon as they assumed the CsCl structure.

The splitting could be due to a local distortion (ionic displacement) about the F centers. The low activation energy for anion vacancy motion in CsBr and CsI[17] would tend to make distortions about F centers more likely in these salts than in most NaCl-type alkali halides. If this distortion is not static at LNT no dichroism would be expected upon partial bleaching with polarized light. There would also be no anisotropy in the F center electron spin resonance. (None was observed in CsCl at LNT[4] but this may have been due to not having a good single crystal.) The fact that the F center in CsCl exhibits a g shift comparable to that of nearly spherically symmetric F centers in NaCl-type alkali halides[4] remains to be explained, since a distortion strong enough to split the $2p$ levels as much as they appear would also mix considerable amounts of higher angular momentum components into the $1s$ ground state.

A peculiar aspect of the F-center structure is its relationship with the K' band of Drickamer *et al.*[9,10] Drickamer *et al.* found the areas of the F and K' bands to be in the ratio of about 10:1 at 20 000 atm and 1:10 at 50 000 atm. (The compression was not perfectly hydrostatic and it is difficult to assess the importance of this in the changes they observed.) Such a dramatic change in oscillator strength needs explanation and data between 1 and 20 000 atm are needed to relate the K' band to the structure of the F band. If indeed the F band in CsBr has three components of equal area then the small additional part of the F band, related to the K' band, may have nothing to do with the F center.

The M band is one of the narrowest color center bands known. At about 15°K its full width at half maximum is only 0.017 eV. This corresponds to the energy of one longitudinal optical phonon, 0.016 eV in CsBr (estimating the restrahl peak to be at 120μ). This narrowness is a characteristic of M bands observed in other alkali halides. In NaCl, Klick and Compton[18] found the M band width to be 0.09 eV at 77°K and

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[14] G. Jacobs, L. Fiermans, and F. Van de Wiele, Physica **27**, 144 (1961).

[15] A. Gold, Phys. Rev. **123**, 1965 (1961).

[16] F. Lüty, Z. Physik **165**, 17 (1961).

[17] D. W. Lynch, Phys. Rev. **118**, 468 (1960).

[18] W. D. Compton and C. C. Klick, Phys. Rev. **112**, 1620 (1958).

estimated the ground-state configuration coordinate frequency to be 1.7×10^{12} cps or about one-fifth the longitudinal optical mode frequency. In CsBr the corresponding ground-state frequency (0.77×10^{12} sec^{-1}) is about one-fifth the longitudinal optical mode frequency (4.1×10^{12} sec^{-1}) which is much lower in CsBr than in NaCl.

The appropriate model for the *M* center appears to be two *F* centers.[19] It is surprising that the *M* band in CsBr, if it too arises from two adjacent *F* centers, does not exhibit the structure of the *F* band. Other aggregate bands, especially the *R*, do not develop to a great extent in CsBr; the bands located between the *F* and *M* bands are weak at all times. No "negative absorption"[3] occurred in the region between the *F* and *M* bands. Irradiation seems necessary in order for luminescence to be stimulated in this wavelength region.

ACKNOWLEDGMENTS

The author wishes to thank R. W. Major who aided in the early stages of the experimental work and Dr. Ronald Fuchs for several helpful discussions.

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[19] C. Z. van Doorn and Y. Haven, Philips Research Repts. **11**, 479 (1956), and C. Z. van Dorn, Philips Research Repts. **12**, 309 (1957).

PHYSICAL REVIEW VOLUME 127, NUMBER 5 SEPTEMBER 1, 1962

Direct Optical Detection of the Ground-State Population Changes of Neodymium in Ethylsulfate Crystals*

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(Received April 16, 1962)

A method is described for optically detecting population changes in paramagnetic salts and applied to the ground state of neodymium ethylsulfate and neodymium in lanthanum ethylsulfate. By observing the transmitted right or left circularly polarized light, the population of *one* of the ground Zeeman levels is selectively monitored. The ground-state population is changed by paramagnetic resonance and thermal relaxation processes. Knowing the ground state, the circular polarization of the light, and the selection rules, information about the excited states can be obtained. The spin-lattice relaxation time between Zeeman sublevels of the ground state is also obtained. After pulsing the microwave resonance transition, the decay of the transmitted light is observed; the decay curve is related to the spin-lattice relaxation curve. Relaxation-time measurements in the temperature range of 1.87 to 2.16°K were made in both the pure and dilute salt. The values ranged from 17 msec for the lower temperature to 11 msec for the higher temperature. The slope of the relaxation time versus the temperature T indicates a temperature dependence for the relaxation time of $T^{-2.5}$ in this temperature range for the 3% neodymium in lanthanum ethylsulfate and $T^{-1.7}$ for the neodymium ethylsulfate crystal. The crystal c axis was oriented parallel to the magnetic field of 4780 Oe; the microwave frequency was 23.6×10^{9} cps.

INTRODUCTION

A VARIETY of experiments have been discussed or performed whose purpose has been to measure optically population changes or spin relaxation between ground Zeeman sublevels.

Wesemeyer and Daniels[1,2] have examined the effect of paramagnetic resonance saturation on the Faraday effect in neodymium ethylsulfate. They examined the spectral region about 200 Å away from strong absorption lines using the 5461 Å line of mercury as did Becquerel, DeHaas, and van den Handel[3] in their work on the Faraday rotation of neodymium ethylsulfate. Daniels and Wesenmeyer found that when the crystal absorbed microwave energy in paramagnetic resonance and the microwave power was sufficient to cause appreciable saturation, the magnetic moment and the Faraday rotation were reduced, as predicted by Kastler.[4]

More recently, Daniels and Rieckhoff[5] have utilized

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* The research reported herein was supported in part by the Army Research Office, Durham, North Carolina.

† This paper forms a part of the dissertation submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Physics at the University of California, Los Angeles, California, January, 1962.

‡ On leave as Fulbright Research Scholar (1961–62) at the Institut de Physique, Université de Strasbourg, Strasbourg, France.

[1] H. Wesemeyer and J. M. Daniels, Z. Physik **152**, 591 (1958).

[2] J. M. Daniels and H. Wesemeyer, Can. J. Phys. **36**, 405 (1958).

[3] J. Becquerel, W. J. DeHaas, and J. van den Handel, Physica **5**, 753 (1938).

[4] A. Kastler, Compt. rend. **232**, 953 (1951).

[5] J. M. Daniels and K. E. Rieckhoff, Can. J. Phys. **38**, 604 (1960).