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Color Centers in Cesium Halide Single Crystals*

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Color centers have been investigated in the CsCl-type alkali halides. Cesium chloride, bromide, and iodide single crystals were grown from the melt and CsCl crystals also from solution. Coloration was produced by 130-kv x rays, 3.0-Mev electrons, and by electrolysis. In CsI coloration resulted from electrolysis only. The absorption of uncolored and colored crystals has been measured from 0.175 to 3.5 μ at 25°, −78°, and −190°C. After coloration all three crystals show one strong band in the visible (near infrared for CsI) and several weaker bands at shorter and longer wavelengths, which shift with temperature change. The spectral positions in mμ for a number of the bands at −190°C are:

	λ_1	λ_2	λ_3	λ_4	λ_5	λ_6	λ_7	λ_8	λ_9	λ_{10}
CsCl	227		270	370	430	579	715	780	855	980
CsBr	241	~270	~315	390	480	646	(780)	840	(~930)	1055
CsI	270	330		425	535	750			1050	1185

The strongest band (λ_6) behaves similarly to the F band in the NaCl-type alkali halides. The half-width of the band (0.20 to 0.23 ev at −190°C) and its increase with temperature, the shifting of the band maximum toward longer wavelengths upon warming to room temperature (by ~0.08 ev), and the conversion by bleaching with light into other bands support the assignment of this band as the F band. The spectral position of the band maximum approximately follows the Mollwo relation ($\lambda_{\max} = \text{const}\ d^n$, where d = interionic distance and $n = 2.5$).

Bleaching experiments suggest the assignment of the band λ_{10} as the M band and the bands between the F and M bands as R bands. The origin of the ultraviolet bands is still uncertain.

Up to now, color-center studies have been confined mainly to the face-centered cubic NaCl-type crystals.[1–6] The present work is extended to the simple cubic CsCl-type alkali halides CsCl, CsBr, and CsI which had been studied only cursorily. In these crystals one would expect relatively small modifications of color centers compared with those in the NaCl-type.

Cesium chloride has been colored previously with electrons,[7] radium rays,[8–10] and x rays.[11] Radium-irradiated crystals (grown from solution and from the melt) show absorption at 560 mμ.[12] Pohl[1] reported the maximum at 600 mμ and Jacobs[13] at 603.9±0.7 mμ.

The color centers in CsCl are very unstable. In daylight the color fades away in minutes.[11,14] Bleaching proceeds about 10 times faster than in KCl and about 1000 times faster than in NaCl.[9,10] Thermal bleaching of CsCl is also faster than in KCl and NaCl.[9,15] The bleaching properties resemble those of RbCl.[12,15] Crystals grown from the melt show deeper and more stable coloration than those grown from solution.[12]

Cesium bromide can be colored by irradiation with x rays at room temperature and at −190°C.[16] The absorption spectrum at −190°C shows the F band at 648 mμ and other bands at 475, 380, 320 (very weak), and 280 mμ. At room temperature the F-band maximum shifts to 675 mμ. While the F band and the near-ultraviolet bands could be bleached with light, that at 280 mμ was only slightly affected.[16] The stability of the coloration in alkali halides decreases as one proceeds from KBr to RbBr and on to CsBr.[9–11]

GROWTH OF CRYSTALS

Using c.p. grade starting materials, CsBr and CsI crystals were grown from the melt in a Vycor crucible by the Bridgman method (i.e., unidirectional cooling of the melt). Cesium chloride was grown by a double-growth technique[17] because of its transition at 469°C. The face-centered cubic modification was first grown from the melt, and then the crystal was transformed into the simple cubic phase by repeating the procedure. Cesium chloride crystals were also grown from solution with urea additive. These crystals, however, contained 0.01% NH_4Cl impurity.[17]

The melt-grown crystals of all three salts showed almost no absorption (Fig. 1) except for CsCl which showed a weak band (probably caused by some impurity) at 188 and a still weaker one at 243 mμ. For crystals of several millimeter thickness the absorption

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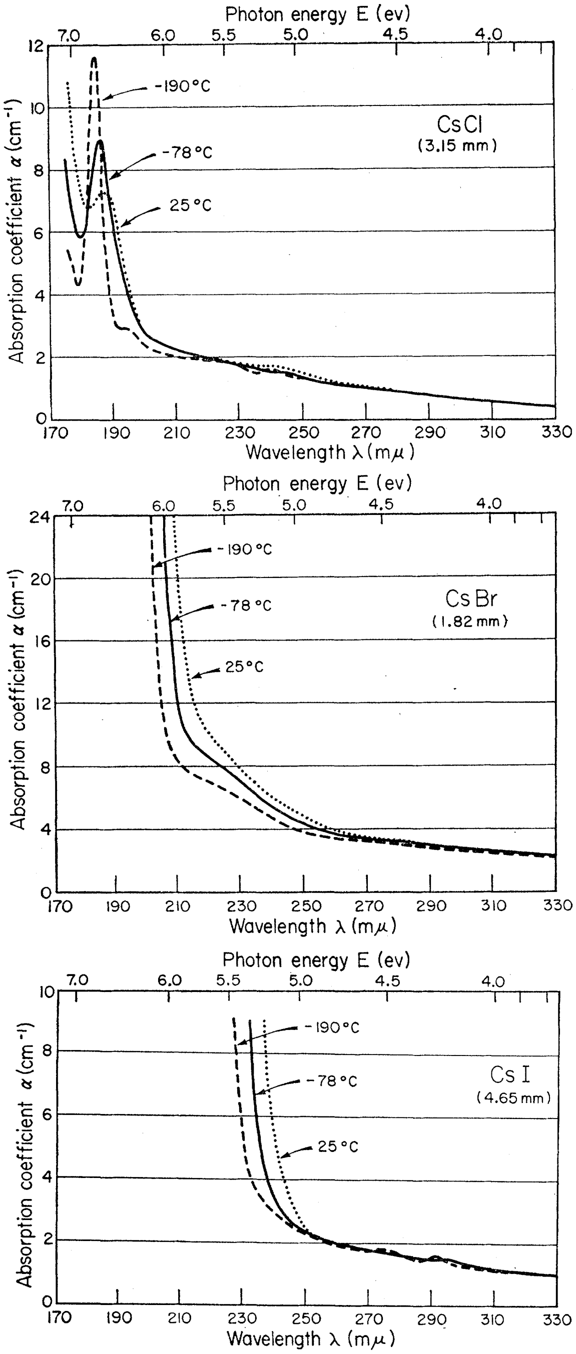

FIG. 1. Ultraviolet absorption of uncolored CsCl, CsBr, and CsI crystals grown from the melt.

edges for CsCl, CsBr, and CsI are around 175, 210, and 235 mμ, respectively. The present values for CsBr and CsI are in agreement with the older data.[18,19]

ABSORPTION MEASUREMENTS AND COLORATION OF CRYSTALS

Optical absorption was measured between 0.175 and 3.5μ with a Beckman DK-1 extended-range spectrophotometer.

Since color centers in cesium halides are not very stable at room temperature, the samples were usually irradiated and measured in a special cryostat at dry-ice and liquid-nitrogen temperatures. Hard x rays (130 kv, 9 ma) and 3.0-Mev electrons were used for the coloration.

Photochemical bleaching was carried out by illuminating the colored crystal with the spectrophotometer

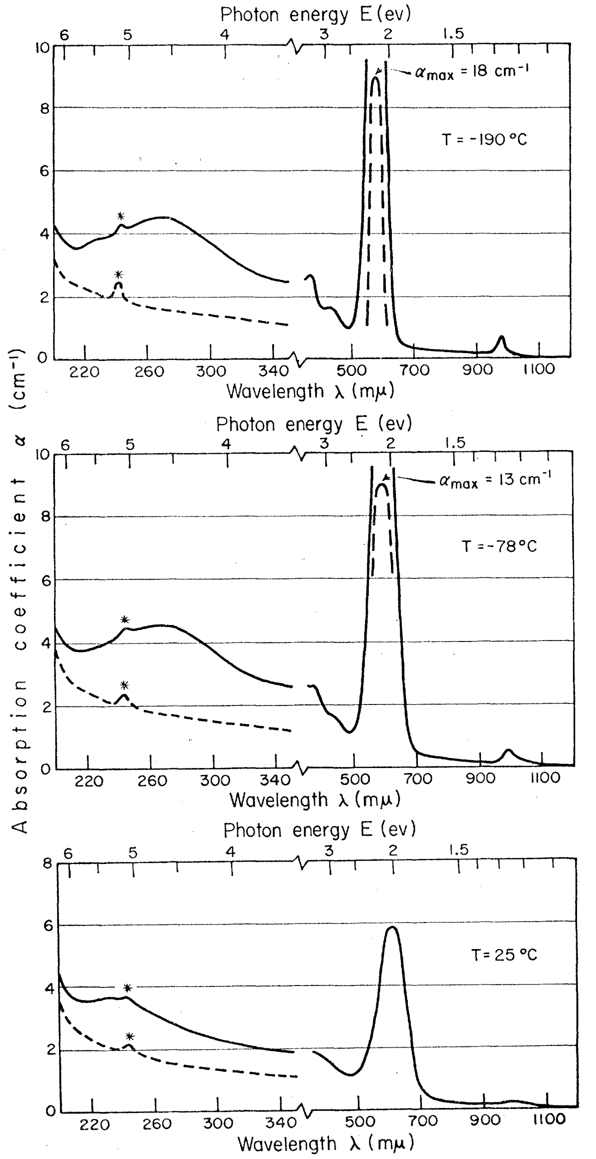

FIG. 2. Absorption spectrum of a melt-grown CsCl crystal (4.8 mm thick) after irradiation with 3.0-Mev electrons (dose: 8×10^4 rad) at −78°C. The dashed curves give the ultraviolet absorption of the uncolored crystal. (*) Impurity band.

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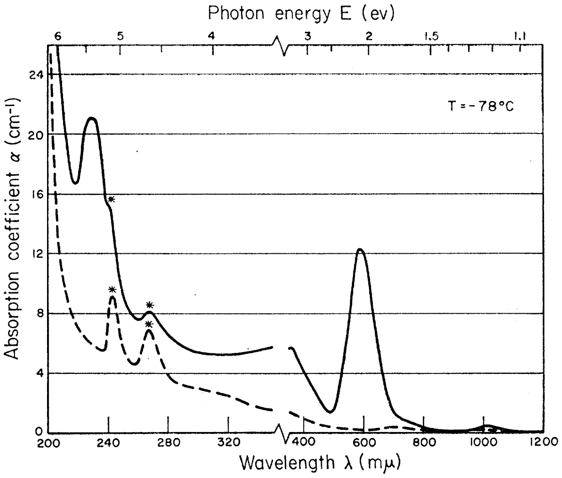

FIG. 3. Absorption spectrum at −78°C of a CsCl crystal (1.67 mm thick) grown from a water solution with 10% urea additive after irradiation with x rays (130 kv, 9 ma) for 4½ hours at −78°C. The dashed curve represents the absorption prior to irradiation. (*) Impurity bands.

tungsten (or hydrogen) source (with a slit opening of about 1 mm).

Cesium iodide could be colored by electrolysis only. Up to 2500 volts were applied to the crystal (at ~480°C) and a half-megohm current-limiting resistor. A platinum electrode, a set of point electrodes, or an edge electrode was used as the cathode. The crystal was held against a carbon anode with a platinum-rhodium tension clip which also applied the voltage to the cathode. The crystals were quenched in a stream of dry nitrogen.

RESULTS

Cesium Chloride

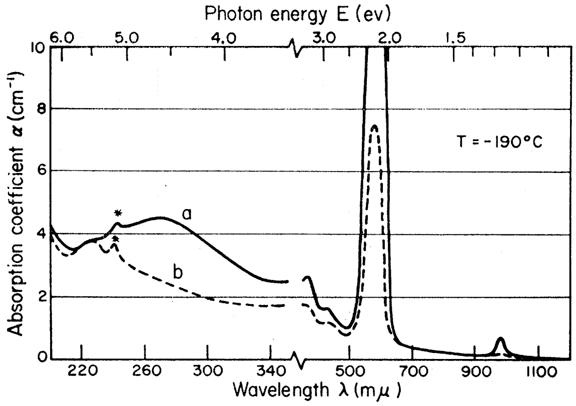

FIG. 4. Absorption spectrum of a melt-grown CsCl crystal (4.8 mm thick) after irradiation with 3.0-Mev electrons (dose: 8×10^4 rad) recorded at −190°C. (a) directly after the irradiation, (b) after warming to 25°C and recooling to −190°C. (*) Impurity band.

The absorption spectra of a CsCl crystal irradiated at −78°C and recorded at −78°, −190°, and 25°C are displayed in Fig. 2. The spectrum recorded at −190°C shows five new bands at 980, 579, 430, 370, and 270 mμ, and a sixth is indicated at 227 mμ. This last band is quite intense for crystals grown from solution (Fig. 3). The 270-mμ band is much broader than the others. Its half-width of 1.0 ev upon irradiation at −78°C, compared to 0.65 ev for irradiation at −190°C, indicates the presence of more than one band in this region. When the colored crystal is warmed to room temperature, all bands decrease except that at 227 mμ, and those at 270 and 980 mμ practically disappear (Fig. 4). Very strong electron irradiation (10^6 rad) produces additional bands (Fig. 5). Table I lists the spectral position of color-centered band maxima and half-

TABLE I. Spectral position of color-center absorption band-maxima and their half-widths for CsCl.

	Position of the maxima[a] (mμ)				Half-widths of the bands[b] (ev)		
	Temperature, °C				Temperature, °C		
	−190°	−78°	+25°		−190°	−78°	+25°
λ_1	227	229	231	H_1	0.3	0.4	0.45
λ_2				H_2			
λ_3	270	270		H_3	0.65	0.7	
λ_4	370	~370	(370)[c]	H_4	(0.3)[c]		
λ_5	430	~430	(430)[c]	H_5			
λ_6	579	590	603	H_6	0.23	0.30	0.36
λ_7	715	735		H_7	0.08	0.08	
λ_8	780	780		H_8	0.04	(0.07)[c]	
λ_9	855	860		H_9	0.06	0.07	
λ_{10}	980	990	1000	H_{10}	0.04	0.06	0.07
λ_{11}	1145	1170		H_{11}	0.06	0.08	

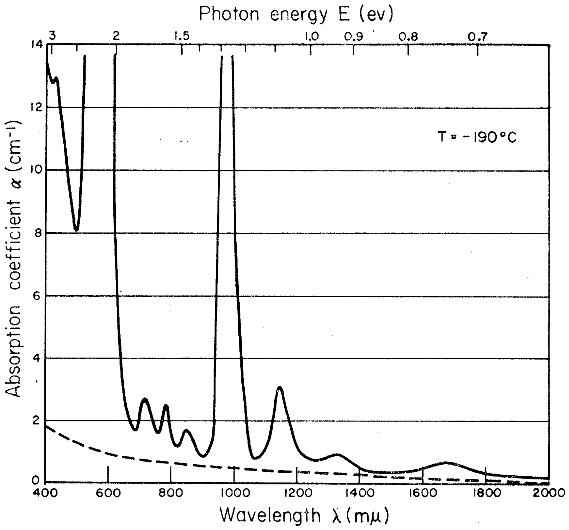

FIG. 5. Absorption spectrum at −190°C of a CsCl crystal (3.15 mm thick) grown from the melt after irradiation with 3.0-Mev electrons (dose: 10^6 rad) at −78°C. The dashed curve gives the absorption prior to irradiation.

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[a] Average uncertainty is between 1 and 5 mμ depending on the intensity, position, and half-width of the band. There are also bands at 1325 and 1675 mμ (at −190°C).
[b] Average uncertainty is 10 to 15%.
[c] Parentheses signify indication only.

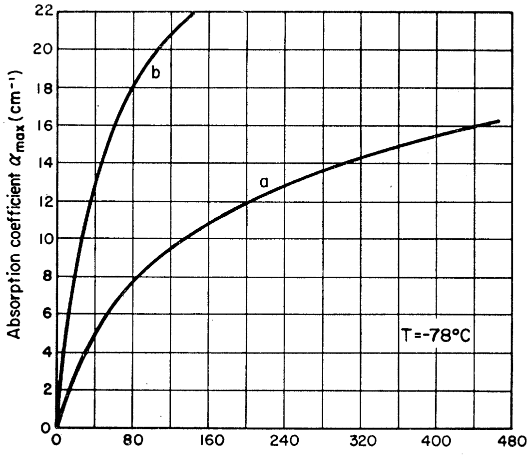

FIG. 6. Intensity of the most prominent band (at 590 mμ) as a function of time upon irradiation with x rays at −78°C of a CsCl crystal: (a) grown from a water solution with urea additive (1.67 mm thick), and (b) grown from the melt (1.76 mm thick).

widths for CsCl. Crystals grown from the melt colored more deeply than those grown from solution (Fig. 6).

The most prominent band, at 603 mμ, can be bleached by exposing the crystal to light in its spectral region. The band at 370 mμ is also affected but not that at 231 mμ. At −78°C the 990-mμ band increases during the initial bleaching of the 590-mμ band (at 603 mμ at room temperature); it then decreases while other long-wavelength bands increase slowly but continuously.

Cesium Bromide

As in CsCl, CsBr crystals were colored with x rays and electrons, mostly at −78°C, and some by electrolysis.

The absorption spectrum of colored CsBr (Fig. 7) is similar to that for CsCl (Fig. 2). Again the broad ultraviolet band (at 315 mμ) and the infrared band

TABLE II. The spectral position of color-center absorption-band maxima and their half-widths for CsBr.

	Position of the maxima[a] (mμ)				Half-widths of the bands[b] (ev)		
	Temperature, °C				Temperature, °C		
	−190°	−78°	+25°		−190°	−78°	+25°
λ_1	241	244	245	H_1	0.25	0.3	0.35
λ_2	~270[c]	~280[c]		H_2	0.3		
λ_3	~315[c]	~315[c]		H_3	0.5	0.6	
λ_4	390	390	(400)[d]	H_4	0.35		
λ_5	480	~490		H_5			
λ_6	646	662	675	H_6	0.20	0.28	0.32
λ_7	(780)			H_7			
λ_8	840	(850)[d]		H_8			
λ_9	(~930)[d]	(~930)[d]		H_9	(0.1)[d]		
λ_{10}	1055	1065	~1070	H_{10}	0.04	0.06	

[a] Average uncertainty is between 1 and 5 mμ depending on the intensity position, and half-width of the band.

[b] Average uncertainty is 10 to 15%.

[c] Exact position seems to depend on factors such as temperature of irradiation.

[d] Parentheses signify indication only.

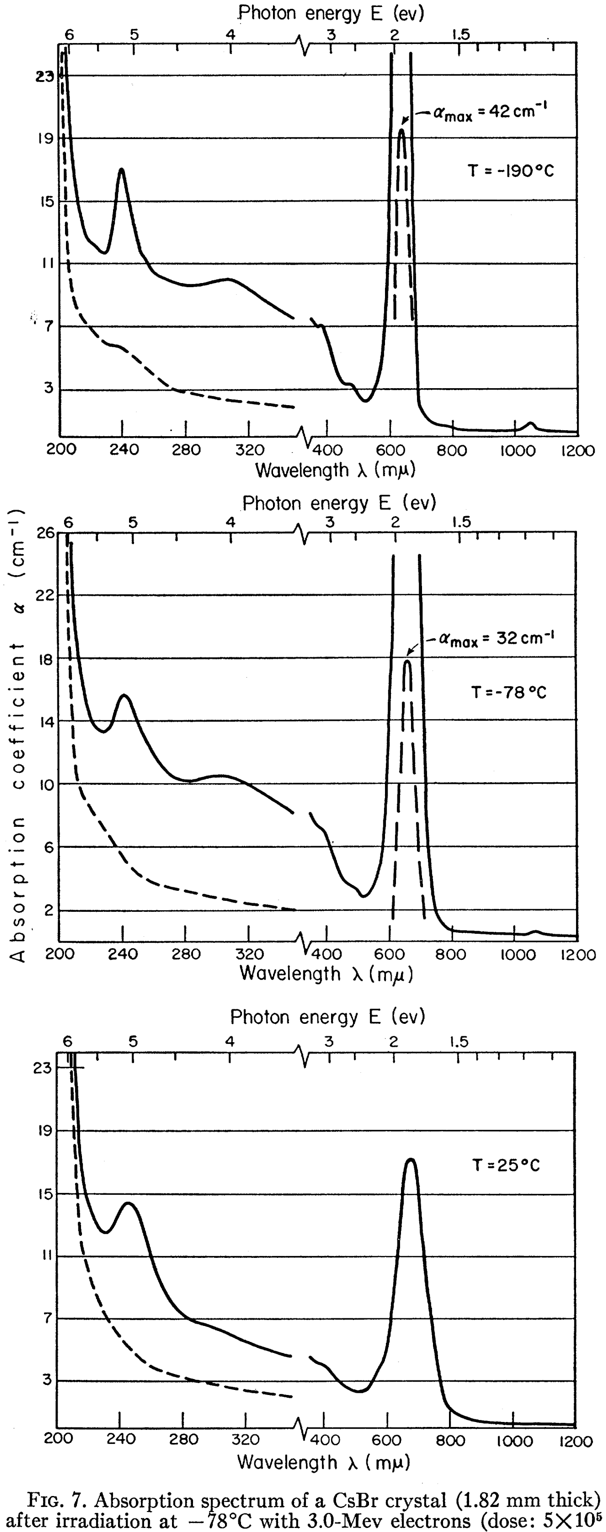

FIG. 7. Absorption spectrum of a CsBr crystal (1.82 mm thick) after irradiation at −78°C with 3.0-Mev electrons (dose: 5×10⁵ rad). The dashed curves give the ultraviolet absorption of the uncolored crystal.

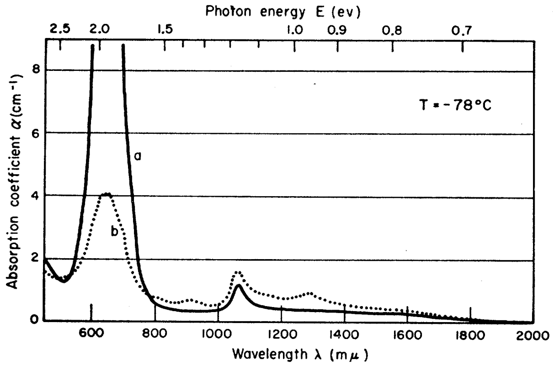

FIG. 8. Absorption spectrum of a CsBr crystal (5.07 mm thick) at −78°C: (a) immediately after irradiation with 3.0-Mev electrons (dose: 5×10^6 rad) at −78°C, and (b) after exposure to light in the 662-mμ band for $\frac{3}{4}$ hour.

(1065 mμ at −78°C) are unstable at room temperature. Coloration at −190°C results in a new ultraviolet band near 270 mμ and in some changes in the infrared region. Crystals colored by electrolysis showed a spectrum similar to that of irradiated ones.

Table II lists the spectral positions and half-widths of color-center absorption bands in CsBr. Several bands not shown in Fig. 7 but indicated for some other samples are also included.

The growth rate of the most prominent band (662 mμ at −78°C) resembles that in solution-grown CsCl (Fig. 6, Curve *a*) but is somewhat slower. Upon photochemical bleaching of the 646-mμ band at −190°C, a broad band developed toward longer wavelengths, with the superposition of a narrower band at 840 mμ. Bleaching at −78°C yielded more bands (Fig. 8). As a result of bleaching, the 662-mμ band tends to broaden. As in CsCl the band shows some asymmetry even before bleaching.

The band near the absorption edge (245 mμ at 25°C), similarly to the analogous band in CsCl, did not bleach upon illumination with the spectrophotometer hydrogen lamp.

TABLE III. The spectral position of color-center absorption-band maxima and their half-width for CsI.

	Position of the maxima[a] (mμ)				Half-width of the bands[b] (ev)		
	Temperature, °C				Temperature, °C		
	−190°	−78°	+25°		−190°	−78°	+25°
λ_1	270	273	275	H_1	0.25	0.35	0.4
λ_2	330[c]	333[c]	335[c]	H_2	0.15	0.20	0.25
λ_3				H_3			
λ_4	425	430	430	H_4	0.4	0.5	0.6
λ_5	535	540	∼540	H_5	0.2	0.25	∼0.25
λ_6	750	768	785	H_6	0.23	0.31	0.36
λ_7				H_7			
λ_8				H_8			
λ_9	1050	1065	1080	H_9			
λ_{10}	1185	1200	1220	H_{10}	0.05	0.07	0.1

[a] Average uncertainty is between 1 and 5 mμ depending on the intensity, position, and half-width of the band. There is also an indication for bands at 1010 and 1480 mμ (at −190°C).
[b] Average uncertainty is 10 to 15%.
[c] Assignment of λ_2 to this band is uncertain.

Cesium Iodide

Although color centers were produced in CsCl and CsBr by irradiation with x rays within a few minutes, such irradiation did not produce measurable coloration in CsI, even after several hours, nor did coloration result from electron doses of 10^6 rad and more. Cesium iodide crystals could be colored by electrolysis only.

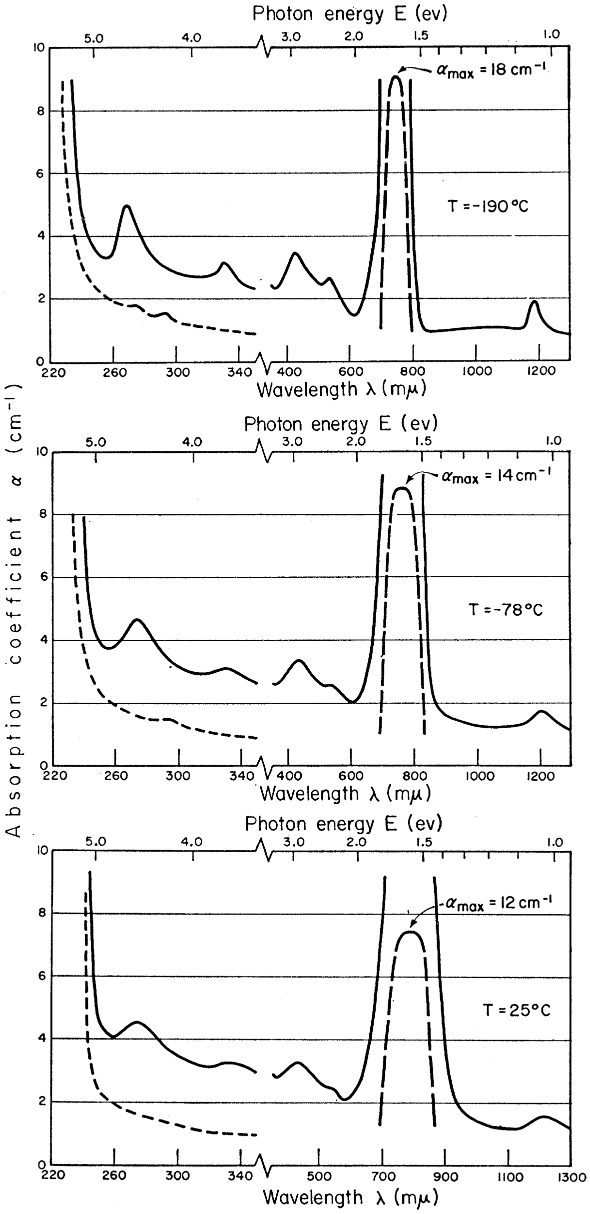

FIG. 9. The absorption spectra at −190°, −78°, and 25°C of a CsI crystal (4.65 mm thick) colored by electrolysis in N_2 atmosphere at 460°C. The dashed curves represent the ultraviolet absorption of the uncolored crystal.

The absorption spectra of a colored CsI crystal (Fig. 9) are similar to those of CsCl and CsBr. Table III shows the positions and half-widths of color-center

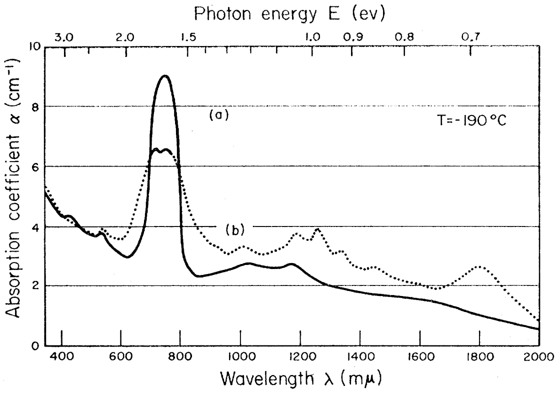

FIG. 10. Absorption spectrum at $-190°C$ of a CsI crystal (2.3 mm thick): (a) after coloration by electrolysis, (b) after illumination for 38 minutes with light in the 770-mμ band at $-78°C$.

bands in CsI. It lists some bands not shown in the sample of Fig. 9. In particular, the 1080-mμ band was much broader than others in its vicinity.

Illuminating the crystal with light in the spectral region of the strongest band ($\sim$770 mμ) caused a decrease in its intensity and an increase of absorption at longer wavelengths. At room temperature the increased absorption is mostly in the 1220-mμ band, while at lower temperatures a variety of bands can be produced.

The results of bleaching at $-78°C$ are illustrated in Fig. 10. After the first four minutes of illumination, the intensity of the 768-mμ band diminished by about 20%. Additional illumination for 34 minutes decreased the band intensity by only 18% more, while the absorption at longer wavelengths increased substantially. In addition to enhancement of the 1010-mμ and 1185-mμ bands, the spectrum recorded at $-190°C$ shows new bands at 1255, 1335, 1450, and 1800 mμ; while the band at 535mμ remained essentially unaffected, that at 425 mμ almost disappeared. As a result of the bleaching (for 38 minutes), the half-width of the 768-mμ band at $-78°C$ increased from 0.31 to 0.37 ev and at $-190°C$ from 0.23 to 0.36 ev. The shape of the top of the band (at $-190°C$) furthermore indicates a slight splitting in the band. The crystal was restored to its unbleached state by storage at room temperature in darkness for half a day.

DISCUSSION

It is quite certain that the coloration in cesium halides is due to electrons and holes trapped in lattice vacancies that are partly present in uncolored crystals and partly created by irradiation. The appearance of several absorption bands suggests the existence of a variety of traps. The absorption spectra of the three crystals show certain similarities in position, shape, and intensity of bands (see Figs. 2, 7, and 9). The strongest band in all three crystals appears in or near the visible spectral range. Its half-width is 0.32 to 0.37 ev at room temperature, and 0.20 to 0.23 ev at $-190°C$. In NaCl-type crystals the analogous band, the *F* band, has a half-width of 0.31 to 0.47 ev at room temperature, and 0.22 to 0.34 ev at $-190°C$.[20] As temperature decreases to $-190°C$, the maximum shifts by about 0.08 ev toward higher energies (or shorter wavelengths), compared to about 0.06 ev in other alkali halides.[20] Another similarity appears upon bleaching with light which is absorbed in the band. During bleaching, the band decreases while another one between 0.95 and 1.25 μ increases.

The growth of the band intensity in CsCl (Fig. 6) and CsBr as function of time during irradiation with x rays is similar to that of the *F* band in the face-centered cubic alkali halides. The concentrations of color centers for this band, computed from Smakula's equation,[21] is of the same order as those achieved in other alkali halides.

The half-width and its change with temperature, the shifting of the band maximum with temperature, and its partial conversion into a new band upon bleaching support the assignment of this band in cesium halides (in or near the visible spectral range) as the *F* band (i.e., absorption due to an electron trapped at a negative ion vacancy).

The spectral position of the *F* band in NaCl-type alkali halides is related to the interionic distance by the Mollwo relation ($\nu_{\max}d^2=$ constant).[22] The exponent, according to Ivey,[23] is 1.84 rather than 2.0. From the slope of the logarithmic plot of the *F*-band maximum $\lambda_{\max}$ versus the lattice constant a in cesium halides (Fig. 11) one obtains a value 2.5 for the exponent.

According to Levy,[24] the ratio of the *F*-band excitation energy to $3/8(e^2/k_0R)$ is close to unity for the face-centered cubic alkali halides. In the above expression, e is the electronic charge, k_0 the high-frequency dielectric constant, and R the cation radius. The expression gives the excitation energy for a $1s$-$2p$ transition of an electron associated with a hydrogen-type impurity imbedded in a dielectric medium, when R is allowed to stand for the Bohr radius. The ratio ranges from 1.0 to 1.24 for the NaCl-type alkali halides. Levy reports 1.64 for CsCl. The present data yield ratios of 1.67, 1.57, and 1.48 for CsCl, CsBr, and CsI, respectively. Therefore, Levy's ratio is about 1.1 for the face-centered and 1.6 for the simple cubic alkali halides.

The partial conversion of the *F* band into a new band by bleaching (with light in the *F* band) suggests that the new band between 0.95 and 1.25 μ corresponds to the *M* band (i.e., absorption due to an electron trapped at a negative ion vacancy adjacent to a cation-anion vacancy pair[4]). The assignment of the *M* band is also

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supported by the fact that this band is already present in moderately colored CsCl and CsBr after irradiation with x rays or electrons, even prior to exposure to "*F* light." The logarithmic plot of the spectral position of the *M*-band maximum with respect to the lattice constant approximates a straight line. The average exponent for the Mollwo-Ivey relation for −190°, −78°, and 25°C is 1.8. According to Ivey,[23] the exponent for the *M* band in face-centered cubic alkali halides is 1.56.

In analogy with face-centered cubic alkali halides, the bands between the *F* and *M* bands in cesium halides might be called *R* bands. According to Seitz,[4] R_1 and R_2 centers are, respectively, one and two electrons at an anion-vacancy pair. Upon exposing the colored crystal at −78°C to light absorbed in the *F* band, the *M* band at first increases most rapidly. As bleaching continues, however, it starts to decrease in intensity while the growth of bands between the *F* and *M* bands and toward longer wavelengths from the *M* band continues, as in NaCl-type crystals. The band beyond the *M* band might be correlated to the *N* band.[25]

A correlation of absorption bands at shorter wavelengths from the *F* band is more difficult. The broad bands observed in CsCl around 270 mμ and in CsBr near 315 mμ might possibly be due to *V* centers (i.e., one or more trapped holes[4]). They appear only upon irradiation of the crystals at low temperatures (−78°, and −190°C), disappear at room temperature, and are about twice as broad as the *F* band. Some electrolytically colored samples, however, also show bands in this region.

At present one can say very little about the remaining absorption bands. Several bands in the three salts can be correlated approximately by a Mollwo-Ivey type empirical relation. As one moves from the *F* band toward shorter wavelengths, the first two bands (in the region of 430 and 370 mμ in CsCl, 485 and 390 mμ in CsBr, and 540 and 430 mμ in CsI) conform to an average exponent of 2.0 and 1.4, respectively. The narrow intense band near the ultraviolet absorption edge (around 229 mμ in CsCl, 243 mμ in CsBr, and 273 mμ in CsI) has an average exponent of 1.7. The appearance of these bands in both irradiated and additively colored samples implies that they are caused by trapped electrons. (With the introduction of excess metal into the crystal, only excess electrons are introduced.[3])

The broadening of the *F* band during bleaching with light in the *F* band and the indication for the production of a band just to the long- and short-wavelength sides of the *F* band may correspond to the *A* and *B* bands reported by Petroff.[26]

A slight asymmetry appears to be present in the *F* band. Duerig and Markham's[27] measurements suggest the same on other alkali halides. At −190°C, especially for highly bleached samples, there is already some indication for the "*F*-band structure" reported by Rabin *et al.*[28,29] at liquid-helium temperature.

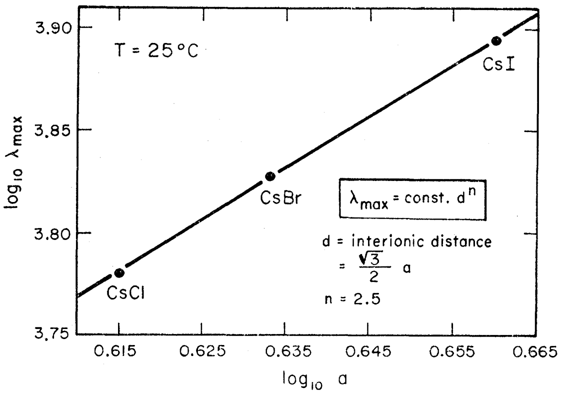

FIG. 11. Relation between the *F*-band maximum and interionic distance (Mollwo relation). Both λ_{max} and a are in angstroms.

Since during the early stages of *F*-center formation electrons are trapped mainly in existing vacancies,[30,31] the lower coloration in a solution-grown, as compared to a melt-grown, CsCl crystal (Fig. 6) indicates that the former had fewer vacancies. Exposure of melt-grown crystals to high temperatures because of their method of growth would tend to form vacancies that could be frozen into the crystal upon cooling. The transformation in the solid state of the melt-grown CsCl crystal from the face-centered cubic to the simple cubic modification probably also introduces vacancies into the crystal because of the distortions accompanying the transformation.

Why CsI can be colored only by electrolysis is not understood at present. Since CsCl and CsBr both color deeply upon irradiation with x rays or electrons, the same would be expected for CsI. One can assume that the electrons and holes produced during irradiation of CsI recombine easily before they can be trapped at vacancies in the lattice, or that an insufficient number of vacancies are present and new ones cannot be formed readily.

Rabin and Klick[32] support the view that at liquid-helium temperature the formation of *F* centers occurs as a result of the creation of halogen-ion vacancies by ejection of halogen ions from their normal lattice positions and their capture at interstitial sites as halogen atoms. These investigators have shown that the total x-ray energy required to form an *F* center at liquid-helium temperature in the NaCl-type alkali

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halides increases strongly upon decrease of the ratio S/D (where S is the space between adjacent halogen ions in a [110] direction of the normal lattice and D the diameter of the halogen atom). In going from KBr to NaBr, S/D diminishes by a factor of approximately two, while the energy needed to form an F center becomes 600 times greater. In the cesium halides, as one goes from CsCl to CsI, the corresponding S/D ratio decreases by a factor of 2.5. Consequently, one might expect a much lower colorability for CsI than for CsCl, at least at low temperatures.

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Microwave Modulation of Light in Paramagnetic Crystals*

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The considerations of Dehmelt and several other workers about the modulation of light by radio-frequency signals in atomic vapors are extended to paramagnetic solids. It is shown that these materials, driven near a microwave resonance at low temperatures may be used both to create and to detect modulation of light at microwave frequencies. Experimental design criteria are discussed at the hand of two numerical examples, modulated circular dichroism in ruby and modulated Faraday rotation in a broad class of ionic rare earth compounds. Some possible applications of microwave modulated light are reviewed.

I. INTRODUCTION

THE modulation of light has a long history. As early as 1881 Righi[1] obtained fluctuations in an interference pattern by a periodic mechanical rotation of one in a pair of crossed Nicols. Rupp[2] produced unresolved side bands on light by operating a Kerr cell near 10^8 cps and detected their passage through a vapor cell absorbing only the central component. More recently Dehmelt[3] proposed that a precessing magnetization of atoms in alkali vapors should give a modulation of the absorption coefficient for circular polarized light propagating in the plane of the precessing component. Such a time dependent magnetization may be created by polarization of the alkali atoms (e.g., by optical pumping) and driving the resonance between two Zeeman levels with a radio-frequency magnetic field.

The successful outcome of such an experiment at 550 kc/sec on Na vapor has been reported by Bell and Bloom.[4] Bloom has announced a similar experiment on a higher frequency transition in K vapor. Series[5] has reported related experiments on modulation in Hg vapor where the precessing magnetization is induced in the excited state.

It is well known that large precessing components of magnetization can be produced by exciting the spin resonance in paramagnetic crystals at low temperatures. It is the purpose of this paper to investigate what happens to a circularly polarized light beam which passes through a crystal in which a precessing magnetization is established by an exciting microwave field.

Several optical experiments and their interplay with the spin resonances in ruby have recently been reported.[6–9] The analogy with corresponding experiments in alkali vapors has been pointed out. In paramagnetic crystals the polarization is established by thermal relaxation processes and need not be established by optical pumping. The characteristics of an experiment in ruby analogous to the Bell and Bloom experiment in Na vapor will be analyzed in Secs. II and III.

Kastler[10] has pointed out that the Faraday rotation in paramagnetic crystals could be decreased by saturation of the microwave resonance(s) in the ground-state multiplet. Opechowski[11] has given a detailed theory of the effect, which can be described simply by saying that microwave saturation changes the effective magnetic temperature. Daniels and Wesemeyer[12] have demonstrated the disappearance of optical Faraday rotation in

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