Vibration-induced absorption (B band) of s^2-configuration ions in alkali halides. II. Magnetic circular dichroism

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(Received 25 February 1981)

The magnetic circular dichroism (MCD) spectra of the B band of In^+ centers ($KCl:In^+$), Sn^{2+} centers ($KI:Sn^{2+}$, $KBr:Sn^{2+}$, $KCl:Sn^{2+}$), and Tl^+ centers ($KCl:Tl^+$) have been measured at various temperatures and compared with a theoretical MCD line shape, which was calculated by Matsushima et al. A doublet-structured B band of the In^+ center, a triplet-structured B band of the Sn^{2+} center, and a nonstructured B band of the Tl^+ center have been interpreted consistently as being due to the Jahn-Teller effect for the P state. The coupling with the Jahn-Teller inactive A_{1g} vibrational mode is found to be as important as the coupling with the T_{2g} mode which gives rise to the splitting of the B band predominantly. It is found, from the MCD line shape of In^+, Sn^{2+}, and Pb^{2+} centers, that the B band of the In^+ and Sn^{2+} centers penetrates into the A-band region considerably.

I. INTRODUCTION

In alkali halides containing s^2-ground-configuration ions such as Tl^+, In^+, Ga^+, Sn^{2+}, and Ag^-, it is well known that the spin-orbit-allowed A and dipole-allowed C absorption bands have temperature-sensitive doublet and triplet structures by the dynamic Jahn-Teller effect, respectively. In our previous paper[1] (called paper I hereafter), by a systematic study of s^2-ion centers, we showed that the vibration-induced B band, too, has a temperature-sensitive fine structure in Sn^{2+} and In^+ centers. Unlike the cases of the A and C bands, however, the fine structure of the B band depends on the s^2 ion; a *triplet* structure is observed for the Sn^{2+} whereas a *doublet* structure is observed for the In^+ center. The three components of the B band in the Sn^{2+} centers were called B_1, B_2, and B_3 in order of increasing energy, whereas the two components in the In^+ centers were called B_2 and B_3. In paper I, we suggested that another component of the B band in In^+ centers, which corresponds to the B_1 band of Sn^{2+}, is located at the low-energy side of the B_2 band of In^+ and hidden under the high-energy tail of the A band. We had reason to believe that a similar fine structure should be observed for the B band of both Sn^{2+} and In^+ centers as was observed for the A and C bands, since Sn^{2+} and In^+ are isoelectronic ions. The present investigation was undertaken to examine whether the previous suggestion is correct. In this paper we show the magnetic circu-

lar dichroism (MCD) spectra of the B band in In^+ and Sn^{2+} centers. It is shown that the MCD data are useful not only to find the correspondence between the B-band components of In^+ and Sn^{2+} centers but also to find a part of the B band hidden under the A band.

Eight years have passed since paper I was published; in the meantime, a great theoretical effort has been made in trying to derive the absorption and MCD line shapes of the B bands.[2-7] Besides the theoretical study, MCD measurements have been done for the B band of s^2-ion centers. At present, there are many MCD data concerning the In^+ center.[7-13] Regarding Sn^{2+} centers, however, the experimental MCD datum published is limited. The only one is on $KI:Sn^{2+}$ but the measurement is confined to a single low temperature, 4.7 K.[7] We present the B-band MCD of various Sn^{2+}-doped alkali halides ($KCl:Sn^{2+}$, $KBr:Sn^{2+}$, and $KI:Sn^{2+}$) measured at various temperatures. As mentioned later, in the comparison between the experimentally observed MCD line shape and theoretically derived line shape, we can understand the *doublet* structure of the B band in the In^+ center and the *triplet* structure in the Sn^{2+} center consistently. In this paper are also shown the MCD spectra of Tl^+ and Pb^{2+} centers. We try to clarify the penetration of the B band into the A band by comparing with the MCD spectra of Tl^+ and Pb^{2+} centers where, unlike the cases of the In^+ and Sn^{2+} centers, the B band never overlaps with the A band.
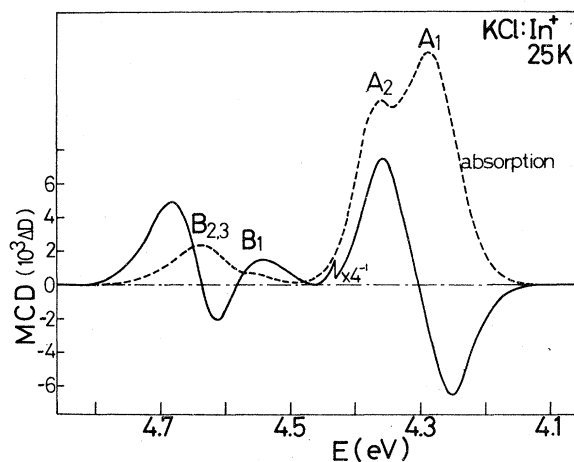
FIG. 1. Absorption and MCD spectra at KCl:In$^+$ (0.003 mole % in the melt) crystal at 25 K. The magnetic field applied is 13.8 kG.

II. EXPERIMENTAL PROCEDURE

Single crystals of KCl containing InCl, SnCl$_2$, or TlCl, KBr containing SnBr$_2$ or PbBr$_2$, and KI containing SnI$_2$ were grown by the Stockbarger or Kyropoulos method. The concentrations of s^2 ions were chosen to be less than 0.005 mole % in the melt so as to avoid the aggregation of s^2 ions. In order to produce the isolated s^2 ions uniformly, the crystals were warmed from room temperature and maintained at 450 °C for 15 min and rapidly quenched from room temperature, immediately before the absorption and MCD measurements. Care was taken to keep the crystals (especially KI and KBr) dry, since they are hygroscopic.

The MCD and absorption spectra were measured using a JASCO J-40A automatic recording spectropolarimeter. The absorption spectra were also measured using a Shimadzu MPS-50L spectrophotometer. Great care was taken to maintain the coincidence of wavelengths of the absorption and MCD spectra during the measurement at a specific temperature. Both absorption and MCD spectra were recorded on the same chart paper, so that the coincidence of wavelengths of the two spectra was well guaranteed. The slit width of the spectropolarimeter was set to be 0.15 nm. The MCD is given by the difference between the optical densities $\Delta D = D_+ - D_-$, for the right- and left-circularly polarized components of light. The polarized light travels through a crystal parallel to the magnetic field. A JASCO electromagnet was used to produce the magnetic field (up to 15 kG). The gap between the poles of the magnet was 15 mm. The field was applied along the [001] direction of the crystal.

III. EXPERIMENTAL RESULTS

Figure 1 shows the absorption and MCD spectra of a KCl:In$^+$ crystal at 25 K. The B band is observed to consist of a weak band peaking at 4.56 eV and a broad band peaking at 4.632 eV. The former is called B_1 band and the latter is called $B_{2,3}$ band hereafter.[14] The B_1 band shows a positive MCD with a peak at 4.542 eV, whereas the $B_{2,3}$ band shows a negative MCD with a peak at 4.612 eV and a positive MCD with a peak at 4.682 eV. When we look around the boundary between the A and B bands, we find that a valley is at 4.49 eV in the ab-
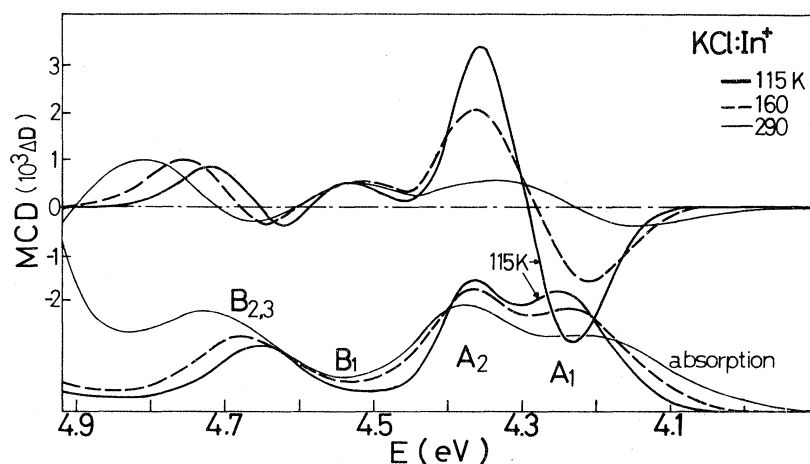

FIG. 2. Absorption and MCD spectra, measured at various temperatures, of the same KCl:In$^+$ crystal used in Fig. 1. The magnetic field applied is 14.3 kG.

sorption spectrum whereas such a valley is at 4.46 eV in the MCD spectrum. That is to say, the MCD of the high-energy side of the A band approaches zero earlier than the absorption itself. On the other hand, the MCD of the low-energy side of the A band approaches zero simultaneously with the absorption. This seems to be due to a penetration of the B band into the A_2 band. Such a penetration is more obvious at high temperatures as is seen in Fig. 2, the A_2 band being shifted toward the B band with increasing temperature.[15] In the absorption spectrum, as the temperature is raised, the weak B_1 band becomes unobservable and immersed under the tail of the $B_{2,3}$ or A_2 band. However, in the MCD spectrum, the positive MCD curve due to the B_1 band is clearly observed even at high temperatures. In this way, the B_1 band is confirmed to be present even at high temperatures.

A quite similar MCD spectrum (especially, the line shape and its temperature dependence) is observed in the B band of KI:Sn^{2+} as shown in Fig. 3 although, unlike the case of KCl:In$^+$, the B band of KI:Sn^{2+} consists of three bands B_1, B_2, and B_3. A low-energy-lying positive MCD is observed at the B_1 band region, a negative MCD at the B_2 band region. Therefore it is suggested that these three MCD parts arise from the B_1, B_2, and B_3 bands, respectively. Just as the case of KCl:In$^+$, the B_1 band is observed, from the MCD spectrum, to penetrate into the A_2 band. The similar MCD spectrum is also obtained in KBr:Sn^{2+} and KCl:Sn^{2+} (Figs. 4 and 5). When comparing the MCD spectra of KBr:Sn^{2+} and KCl:Sn^{2+} with that of KI:Sn^{2+}, we find the following difference: in the cases of KBr and KCl, a negative MCD curve is observed at

FIG. 4. Absorption and MCD spectra of a KBr:Sn^{2+} (0.005 mole % in the melt) crystal at 290 and 50 K. The magnetic field applied is 14.3 kG.

the low-energy side of the B_1 band.

As seen in Figs. 2 − 5, the MCD signal of the $B_{2,3}$ band of the In$^+$ center and of the B_2 and B_3 bands of the Sn^{2+} center grows with increasing temperature. Such a growth seems to be characteristic of vibration-induced absorption band, because the MCD signal of the *spin-orbit-allowed A* band decreases in intensity and becomes broad with increasing temperature, and the same phenomenon has been observed for the *dipole-allowed C* band.[8,9]

Figures 6 and 7 show the absorption and MCD spectra of KCl:Tl$^+$. In the Tl$^+$ center, the B band is located close to the C band and separated from the A band completely. The line shape is a non-structured single band[1,16] although the A and C bands have the doublet and triplet structures, respectively, just as the cases of the In$^+$ and Sn^{2+}
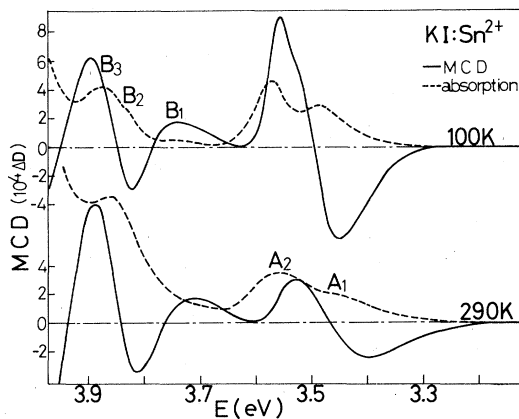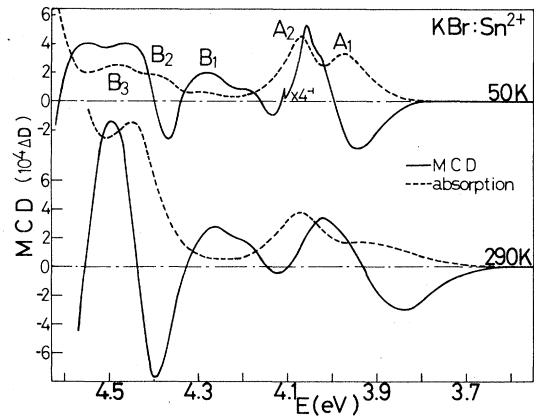

FIG. 3. Absorption and MCD spectra of a KI:Sn^{2+} (0.005 mole % in the melt) crystal at 290 and 100 K. The magnetic field applied is 14.3 kG.
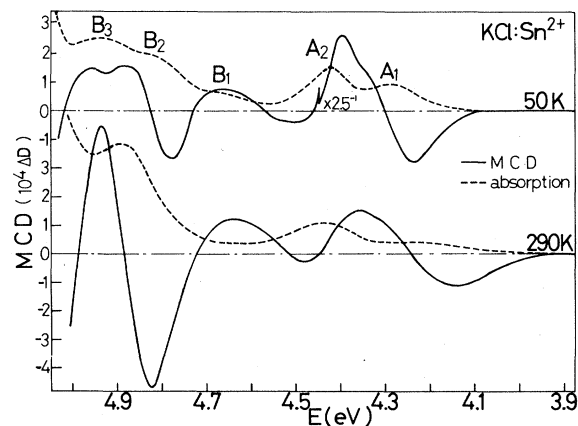

FIG. 5 Absorption and MCD spectra of a KCl:Sn^{2+} (0.005 mole % in the melt) crystal at 290 and 50 K. The magnetic field applied is 14.3 kG.
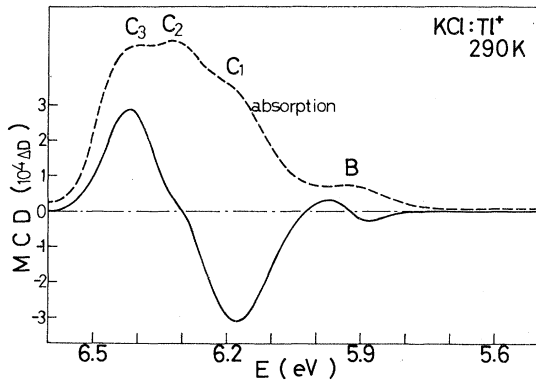
FIG 6. Absorption and MCD spectra of a KCl:Tl$^+$ (0.0003 mole% in the melt) crystal at 290 K in the B- and C-band region. The magnetic field applied is 14.3 kG.

centers. The B-band MCD of KCl:Tl$^+$ looks like the derivative of the absorption curve, with a negative MCD peak at the low-energy side and a positive peak at the high-energy side. In Fig. 7 one can see that, unlike the A band of the In$^+$ and Sn^{2+} centers, the MCD curve of the high-energy side approaches zero simultaneously with the absorption curve at both 50 and 290 K. The quite similar MCD curve is also observed in the A band of Pb^{2+} center as shown in Fig. 8.

IV. DISCUSSION

What are the MCD data obtained telling us? We can give three suggestions when we look closely at Figs. 1 — 8. Firstly, because of the similarity of the MCD spectra between the In$^+$ and Sn^{2+} centers, the B band of both centers would seem to arise
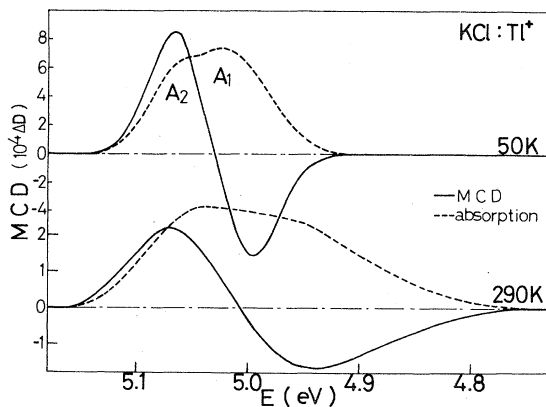

FIG. 7. Absorption and MCD spectra, in the A-band region, of the same KCl:Tl$^+$ crystal used in Fig. 6. The magnetic field applied is 14.3 kG.
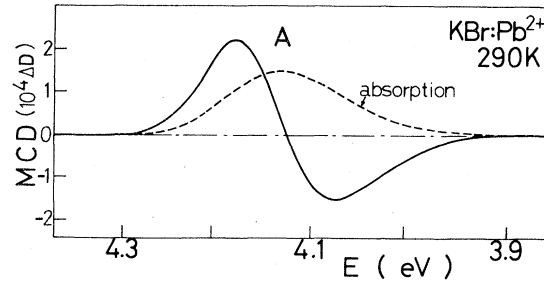

FIG. 8. Absorption and MCD spectra of the A band in a KBr:Pb^{2+} (0.002 mole% in the melt) crystal at 290 K. the magnetic field applied is 13.1 kG.

from the same origin although their absorption line shapes are different from each other. Secondly, the B band of the In$^+$ and Sn^{2+} centers penetrates into the A band. Thirdly, the influence of the B band on the A band depends on the distance of the two bands. In the following paragraphs, we consider how the MCD spectra observed among In$^+$, Sn^{2+}, and Tl$^+$ centers are explained consistently.

The A, B, and C bands have been attributed to the $(a_{1g})^2 \rightarrow (a_{1g})(t_{1u})$ transition in s^2-ion center.[17] Recently Matsushima et al.[5-7] derived the line shape of the A and B bands by solving a 12×12 matrix of linear electron-lattice interaction concerning the $a_{1g}t_{1u}$ state. According to their calculation, of the five adiabatic potential-energy surfaces $\epsilon^{(l)}$ ($l = 5$, 6, 7, 8, and 9) responsible for the B band, transitions from the ground state $^1A_{1g}$ to the three surfaces of $l = 6$, 7, and 8 contribute to the B-band line shape since the intensity of transitions to the $l = 5$ and 9 surfaces is considerably small. The $l = 6$ surface gives rise to a weak shoulder on the low-energy part of the B band whereas the $l = 7$ and 8 surfaces give rise to two big bands on the high-energy part,[7] giving an asymmetric triplet-structured B band as a whole. The theoretically derived line shape is in agreement with the B-band line shape of observed Sn^{2+} centers. Thus, it is concluded that the $l = 6$ surface is responsible for the B_1 band, whereas the $l = 7$ and $l = 8$ surfaces are predominantly responsible for the B_2 and B_3 bands, respectively.

In the case of the In$^+$ center, the $l = 7$ and 8 surfaces are close to each other and result in a formation of a single band.[7] Therefore the B band is expected to have a doublet structure, the $l = 6$ surface being separated from the $l = 7$ and 8 surfaces as the case of the Sn^{2+} center. The calculated line shape of the doublet structure is quite similar to the observed B band.[5] Therefore it is suggested that (1) the weak band located at the low-energy side of the

B band in KCl:In$^+$ (B_1 band of Fig. 1) corresponds to the B_1 band of the Sn^{2+} center, and (2) although each of the B_2 and B_3 bands has not been observed in the In$^+$ center by experiment distinctly, the $B_{2,3}$ band of the In$^+$ center is composed of two bands of low-energy-lying B_2 and high-energy-lying B_3 corresponding to the B_2 and B_3 bands of the Sn^{2+} center, respectively. In order to examine whether such a suggestion is correct or not, we investigate the MCD spectra of the In$^+$ and Sn^{2+} centers.

The transition from the ground state to the $l = 7$ surface (responsible for the B_2 band), gives rise to an MCD with a negative sign, whereas the transition to the $l = 8$ surface (responsible for the B_3 band) gives rise to an MCD with a positive sign.[7] As a result, if the above suggestion is correct, a quite similar S-shaped MCD curve (with a negative sign at the low-energy side) is theoretically expected both for the B_2-B_3 band region of the Sn^{2+} center and for the $B_{2,3}$ band region of the In$^+$ center. This is in quite satisfactory agreement with experiment (see Figs. 1—5). Therefore it is confirmed that the B_2 and B_3 bands associated with the $l = 7$ and 8 surfaces are also present in the In$^+$ center.

When an Sn^{2+}-doped crystal is cooled to a temperature less than 30 K, an additional fine structure appears on the B_2 and B_3 bands in the absorption spectrum as shown by arrows in Fig. 9. Previously, we[18] suggested that the fine structure is associated with the Jahn-Teller splitting of the fivefold-degenerate B state because just quintuplet structure was observed at that time. However, as seen in Fig. 9(b), there are at least six peaks (including the B_1 peak) in the B-band region as a whole, indicating the additional fine structure are not due to the Jahn-Teller splitting. It is believed that the fine structure is attributable to the phonon structure since, as is expected for the phonon structure, (1) the structure appears at very low temperatures clearly, (2) the structure consists of the successive components whose peak-to-peak interval is almost constant (~ 153 cm^{-1}), and (3) the components become sharp with decreasing temperature. A new MCD line shape, which is different from the line shape observed at high temperatures (Fig. 3), is obtained at such low temperatures as shown in Fig. 9(b). Since we are concerned about the fine structure due to the Jahn-Teller effect in the present paper, we do not refer to the MCD line shape deformed by the appearance of the phonon structure here.

Next, we examine whether the B_1 band of the In$^+$ center corresponds to the B_1 band of the Sn^{2+}
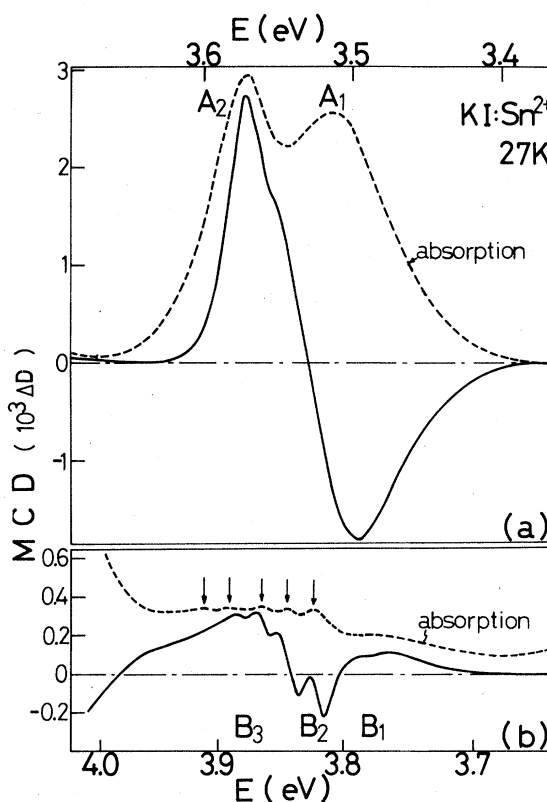

FIG. 9. Absorption and MCD spectra of the A band (a) and the B band (b), at 27 K, of the same KI:Sn^{2+} crystal used in Fig. 3. The magnetic field applied is 13.3 kG. Arrows indicate the peak positions of absorption bands resolved at low temperatures.

center truly, that is to say, whether the same $l = 6$ surface is responsible for the low-energy part of the B band in both the In$^+$ and Sn^{2+} centers. When the temperature is raised, neither the B_1 band of the In$^+$ center nor the B_1 band of the Sn^{2+} center show any remarkable increase in the MCD intensity nor any remarkable shift, unlike the cases of their B_2 and B_3 bands. This seems to indicate that both bands arise from the same origin. To make sure of it, let us examine the MCD line shape of the B_1 band.

The transition from the ground state to the $l = 6$ surface gives rise to a different MCD line shape between the In$^+$ and Sn^{2+} centers because of the difference of spin-orbit coupling strength.[7] The following MCD line shape has been theoretically derived for the B_1 band which is composed of the absorption band associated with the transition to the $l = 6$ surface and, partially, of the absorption band associated with the $l = 7$ surface; for the In$^+$ center, a sharp positive MCD and a sharp negative

peak are at the high-energy part of the B_1 band and a broad positive peak is at the low-energy part, whereas for the Sn^{2+} center, a sharp positive peak is at the high-energy part and a negative peak is at the low-energy part [see Figs. 5(d) and 6(d) of Ref. 7]. Such an MCD line shape was obtained from the Jahn-Teller coupling with T_{2g} lattice-vibrational mode. Taking into account the broadening of the so-called B_1, B_2, and B_3 bands by additional coupling with the Jahn-Teller inactive A_{1g} mode, the MCD curve is expected to be smoothed out as follows. In the case of the In^+ center, the sharp positive and negative peaks are extinguished since both peaks are located close to each other and, therefore, cancelled by each other. As a result, the B_1 band is expected to have an MCD line shape with only a positive sign. In the case of the Sn^{2+} center, both the positive and negative peaks are expected to be broadened considerably. Thus, a big difference between the In^+ and Sn^{2+} centers is that in the Sn^{2+} center, a negative MCD signal is expected at the low-energy part of the B_1 band. The experimental results of $KCl:In^+$, $KBr:Sn^{2+}$, and $KCl:Sn^{2+}$ are in satisfactory agreement with this expectation (see Figs. 1, 2, 4, and 5). This indicates that the B_1 band of the In^+ center certainly corresponds to the B_1 band of the Sn^{2+} center by reason that the $l = 6$ surface is predominantly responsible for both bands. At the same time, this indicates that any component of the B band is not present at the low-energy side beyond the observed B_1 band.

Contrary to the expectation, the B_1 band of $KI:Sn^{2+}$ observed does not have any negative MCD signal at the low-energy part. Let us consider the reason. The B-band MCD line shape of the s^2-ion center is sensitive to the position of the B band between the A and C bands.[6,7] A parameter x ($= E_{BA}/E_{CA}$, where E_{BA} means the separation between the A and C bands) is used to describe the relative position of the B band. Matsushima et al. derived the MCD line shapes at $x = 0.125, 0.25,$ $0.5,$ and 0.75 which were supposed to correspond to the cases of Ga^+, In^+, Sn^{2+}, and Tl^+ centers, respectively.[6,7] According to them (as mentioned above), the low-energy part of the B_1 band gives a positive MCD signal in the case of $x = 0.25$ (In^+), whereas it gives a negative MCD signal in the case of $x = 0.50$ (Sn^{2+}), suggesting that the MCD sign is changed as x is decreased from $x = 0.50$ to 0.25. We first supposed that the MCD line shape at $x = 0.50$ must be fitted for all of $KI:Sn^{2+}$, $KBr:Sn^{2+}$, and $KCl:Sn^{2+}$. However, it should be noted that the x value becomes small as the B band

approaches the A band. The separation of the A_2 and B_1 band peaks is $0.16, 0.23,$ and 0.25 eV for $KI:Sn^{2+}$, $KBr:Sn^{2+}$, and $KCl:Sn^{2+}$. Therefore we can understand the reason why the B_1-band MCD line shape of $KI:Sn^{2+}$ observed is not similar to those of $KBr:Sn^{2+}$ and $KCl:Sn^{2+}$ but rather similar to that of $KCl:In^+$.

Next let us examine the B band of the Tl^+ center. A theoretical B-band shape of the Tl^+ center, which was calculated in the case of $x = 0.75$ under a condition of the coupling with T_{2g} mode, has an asymmetrical triplet structure.[6] The corresponding MCD line shape is quite similar to that of the Sn^{2+} center, although the MCD curves due to the B_1, B_2, and B_3 bands are considerably closer to each other than in the case of the Sn^{2+} center (see Fig. 8 of Ref. 6). These disagree with the experimental result of each of absorption[1,15] and MCD (Fig. 6 of this paper). However, when the A_{1g} mode is additionally coupled to the B state, the triplet-structured absorption band is smoothed out and changed into a single nonstructured band, in agreement with the experiment. In this case, a weak MCD signal which appeared in the B_1-band region is supposed to be extinguished by cancellation between a negative MCD signal associated with the $l = 6$ surface and a positive MCD signal associated with the $l = 7$ surface. The remaining signal is a big, negative MCD signal associated with the $l = 7$ surface at the B_2-band region and a big, positive signal associated with the $l = 8$ surface at the B_3-band region, giving an S-shaped MCD line shape as a whole. This agrees with the experiment. Therefore it is suggested that the A_{1g} mode plays a very important role in determining not only the absorption line shape of the B band in Tl^+ center but also the MCD line shape.[19]

V. CONCLUSION

We have reached the following conclusions from the analysis of MCD line shape, which is consistent with the result of absorption line shape. (1) Transition from the $^1A_{1g}$ ground state to the three adiabatic potential-energy surfaces $\epsilon^{(l=6)}$, $\epsilon^{(l=7)}$, and $\epsilon^{(l=8)}$ are responsible for the B band observed in various s^2-ion centers. (2) The B_1 band of the Sn^{2+} center corresponds to the B_1 band of the In^+ center, and both are associated with the transition to the $\epsilon^{(l=6)}$ surface. (3) The B_2 and B_3 bands of the Sn^{2+} center are predominantly associated with the transi-

tions to the $\epsilon^{(l=7)}$ and $\epsilon^{(l=8)}$ surfaces, respectively. The same is also true for the In$^+$ center, but since the two bands are located close to each other, a single band called the $B_{2,3}$ band is observed at the high-energy side of the B_1 band. (4) In the case of

Tl$^+$ center, all three bands are located close to each other and, moreover, the coupling with the A_{1g} mode is as strong as the coupling with the T_{2g} mode which causes a triplet structure in the B band, resulting in a formation of a single B band.

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[14]This notation is different from that used in paper I.

[15]Theoretically, the A band of the s^2-ion center has an S-shaped MCD line shape which approaches zero simultaneously with the absorption itself at both the low- and high-energy sides [K. Cho, J. Phys. Soc. Jpn. 27, 646 (1969); Mastushima et al., Z. Phys. B 25, 211 (1976)]. Indeed, such an MCD line shape is observed for the doublet-structured A band of the Tl$^+$ center and for the nonstructured A band of the Pb^{2+} center (see

Figs. 7 and 8). In the cases of the In$^+$ and Sn^{2+} centers, however, the MCD of the high-energy side approaches zero earlier than the absorption (see Figs. 1 − 5). A big difference between the group of the Tl$^+$ and Pb^{2+} centers and the group of the In$^+$ and Sn^{2+} centers is that the B band, which is located at the high-energy side of the A band, is completely separated from the A band in the former group, whereas the B band is located close to the A band in the latter group. Therefore it is probable that the low-energy tail of the B band penetrates into the A band, although it cannot be observed in the absorption spectrum clearly. The penetration of the B band is clearly suggested from the fact that an MCD signal of the B_1 band follows the MCD signal of the A band at the high-energy region of the A band (see Sec. IV).

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lated the MCD line shape of the C band in several cases of $a:b:c = 0.2:1.0:1.0$, $0.4:1.0:1.0$, $0.6:1.0:1.0$, $0.8:1.0:1.0$, and $1.0:1.0:1.0$ (where a, b, and c are coupling strengths with the A_{1g}, E_g, and T_{2g} modes, respectively). The C-band MCD line shape observed in KCl:Tl$^+$ (Fig. 6) is quite similar to that calculated using a value of $a:b:c = 0.8:1.0:1.0$. On the other hand, the C-band MCD line shape observed in KCl:Sn^{2+} (T. Tsuboi, unpublished) is similar to the line shape calculated using a value of $a:b:c = 0.4:1.0:1.0$. This indicates that the coupling with the A_{1g} mode is certainly as strong as the coupling with the other modes in the Tl$^+$ center as was expected.