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Circular Dichroism of Chromium(III) Complexes. VII. Circular Dichroism in the Spin-forbidden Transitions of *cis*-$[Cr(N)_2(O)_4]$ Type Complexes with Ethylenediaminetetraacetate Analogues

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(Received January 5, 1981)

Potassium (2*S*,4*S*)-2,4-pentanediaminetetraacetatochromate(III) monohydrate was newly prepared and was found to be formed stereospecifically with a $\Lambda(\Delta\Delta\Lambda)$ absolute configuration as has been revealed for the corresponding Co(III) complex. The circular dichroism spectra in the spin-forbidden transitions of this new complex along with the known analogues, $(-)_{589}$-ethylenediamine-*N*,*N'*-diacetato-*N*,*N'*-dipropionatochromate(III) and $(+)_{589}$-(*S*,*S*)-2,2'-(ethylenediimino)disuccinatochromate(III) complexes, were measured and discussed in comparison with those in the first spin-allowed transitions on the basis of the theoretical relations between the rotational strengths for the spin-forbidden and the spin-allowed transitions in trigonal and tetragonal fields.

In our previous papers of this series,[1–3] circular dichroism(CD) spectra in the spin-forbidden d-d transitions of trigonal and tetragonal Cr(III) complexes have been elucidated in comparison with those in the first spin-allowed d-d transitions by using the theoretical relations between the rotational strengths for the spin-forbidden and the spin-allowed transitions. No CD datum in the spin-forbidden transitions and few CD data in the spin-allowed transitions of chiral Cr(III) complexes with sexidentate ethylenediaminetetraacetate(edta) type ligands have been reported, whereas the CD spectra of several Co(III) complexes with edta analogues have been studied and discussed in relation with their absolute configurations and theories of optical activity.[5–7] The CD spectra in the first band region of such complexes have been dealt with the assumption of C_2 symmetry descending from trigonal(D_3) or tetragonal(D_4) symmetry.[4,5a–c,7a,8] To clarify these CD behavior, it is significant to reveal whether the CD in the spin-forbidden transitions of Cr(III) complexes with edta analogues will be elucidated in terms of the theoretical considerations for the rotational strengths in the d-d transitions on either the trigonal or tetragonal symmetry assumption. In this instance, it is desirable to examine the CD spectra in the spin-forbidden and the spin-allowed transitions of chiral Cr(III) complexes containing edta analogues with four glycinate rings as well as those of the known Cr(III) complexes,[4] $(-)_{589}$-*trans*(O_5)†-ethylenediamine-*N*,*N'*-diacetato-*N*,*N'*-dipropionatochromate(III), $(-)_{589}$-$[Cr(eddda)]^-$, and $(+)_{589}$-*trans*(O_5)†-(*S*,*S*)-2,2'-(ethylenediimino)disuccinatochromate(III), $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$, both of which have two five-membered and two six-membered N–O chelate rings. Apart from some racemic complexes of the former type,[9] no chiral Cr(III) complexes have yet been known.

This paper reports the preparation and stereospecific formation of a new chiral Cr(III) complex containing (2*S*,4*S*)-2,4-pentanediaminetetraacetate(*S*,*S*-ptnta^{4-}) as one of the closely similar sexidentate ligands to the edta ligand. The CD data in the spin-forbidden transitions as well as in the spin-allowed transitions of this complex are examined together with those in the spin-forbidden transitions of chiral *trans*(O_5) isomers of $(-)_{589}$-$[Cr(eddda)]^-$ and $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$, of which the CD spectra in the spin-allowed transitions have been previously reported by Radanovic and Douglas.[4] Their CD spectra in the whole region of the d-d transitions are compared with one another and discussed on the basis of both trigonal and tetragonal symmetry assumptions.

Experimental

Preparation of Ligands and Complexes. *1) (2*S*,4*S*)-2,4-pentanediaminetetraacetic Acid (*S*,*S*-ptntaH$_4$·1.5H$_2$O):* This ligand was prepared from (2*S*,4*S*)-2,4-pentanediamine by the method for the racemic one.[6] Found: C, 43.27; H, 6.92; N, 7.63%. Calcd for *S*,*S*-ptntaH$_4$·1.5H$_2$O: C, 43.21; H, 6.97; N, 7.75%.

*2) $(+)_{589}$-K[Cr(*S*,*S*-ptnta)]·H$_2$O:* To a solution of 1.0 g of *S*,*S*-ptntaH$_4$·1.5H$_2$O in 15 cm^3 of water was added a solution containing 0.79 g of potassium hydroxide in 10 cm^3 of water. After dissolving 1.2 g of $Cr(NO_3)_3\cdot 9H_2O$ in this solution at 70 °C, the mixture was heated at 75—80 °C with stirring for about 16 h. The color of the solution changed gradually from violet to red violet. By adding ethanol to this chilled solution, red powder was obtained. Recrystallization was performed from warm water and ethanol. Found: C, 35.29; H, 4.61; N, 6.36%. Calcd for K[Cr(*S*,*S*-ptnta)]·H$_2$O: C, 35.53; H, 4.59; N, 6.38%.

*3) $(-)_{589}$-$[Cr(eddda)]^-$ and $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$:* The ligands, ethylenediamine-*N*,*N'*-diacetic-*N*,*N'*-dipropionic acid (edddaH$_4$)[5b] and (*S*,*S*)-2,2'-(ethylenediimino)disuccinic acid (*S*,*S*-eddsH$_4$),[7a] were prepared by the reported methods. $(-)_{589}$-$[Cr(eddda)]^-$ and $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$ were obtained by the method of Radanovic and Douglas.[4] The absorption and CD spectra along with the infrared spectra confirm that these complexes are *trans*(O_5) isomers which have been previously established.[4,10]

Measurements. Absorption spectra were recorded on a Shimadzu UV-200S spectrophotometer, and CD spectra on a JASCO MOE-1 spectropolarimeter. Infrared spectra were obtained by a JASCO DS-402S spectrophotometer.

Results and Discussion

*Stereospecificity of [Cr(*S*,*S*-ptnta)]$^-$.* The visible absorption spectrum of [Cr(*S*,*S*-ptnta)]$^-$ is quite similar in band position and intensities to that of the trimeth-

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† Oxygens of five-membered chelate rings occupy trans-axial coordination sites.

ylenediaminetetraacetatochromate(III) complex of the analogous type, [Cr(trdta)]$^-$, as shown in Fig. 1. This fact lends support to sexidentate coordination of an *S*,*S*-ptnta ligand as in the trdta complex. The CD intensities of the *S*,*S*-ptnta complex are found to be as large as those of $(-)_{589}$-[Cr(eddda)]$^-$ and $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$ as shown in Fig. 1 and Table 1. The CD pattern in the spin-allowed band region of the *S*,*S*-ptnta Cr(III) complex is much different from that of the corresponding Co(III) complex,6) whereas the CD spectra of $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$ and $(-)_{589}$-[Cr(eddda)]$^-$ behave similarly to those of the corresponding chiral *trans*(O_5) isomers of the *S*,*S*-edds and eddda Co(III) complexes, respectively.4,5b,7a) As has been revealed for [Co(*R*,*R*-ptnta)]$^-$,6) the stereospecific formation of [Cr(*S*,*S*-ptnta)]$^-$ is expected to take place with a $\Lambda(\Lambda\Delta\Lambda)$ absolute configuration owing to the preference of the equatorial orientation of two methyl groups to the chelate rings.6) It has been proposed for Co(III) complexes with edta type ligands5,7) that the CD signs of the lowest frequency component in the first band region are correlated to the absolute configurations; the complexes giving a positive CD component at the lowest frequency side of the first band take a $\Lambda(\Lambda\Delta\Lambda)$ configuration, and *vice versa*. For the *S*,*S*-ptnta Cr(III) complex, the absolute configuration on the basis of the CD criterion is consistent with that determined in terms of stereospecificity arising from preferential coordination of sexidentate chiral ligands to Cr(III). That is, as in Fig. 1, [Cr(*S*,*S*-ptnta)]$^-$ with a $\Lambda(\Lambda\Delta\Lambda)$ configuration gives a positive CD component at the lowest frequency side of the first band as has been confirmed for $\Lambda(\Lambda\Delta\Lambda)$-$(-)_{589}$-[Cr(eddda)]$^-$ and $\Lambda(\Lambda\Delta\Lambda)$-$(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$, of which the absolute configurations have been determined by the X-ray analysis10) and the stereospecific formation,4) respectively. Thus, the *S*,*S*-ptnta Cr(III) complex is formed stereospecifically with a $\Lambda(\Lambda\Delta\Lambda)$ configuration.

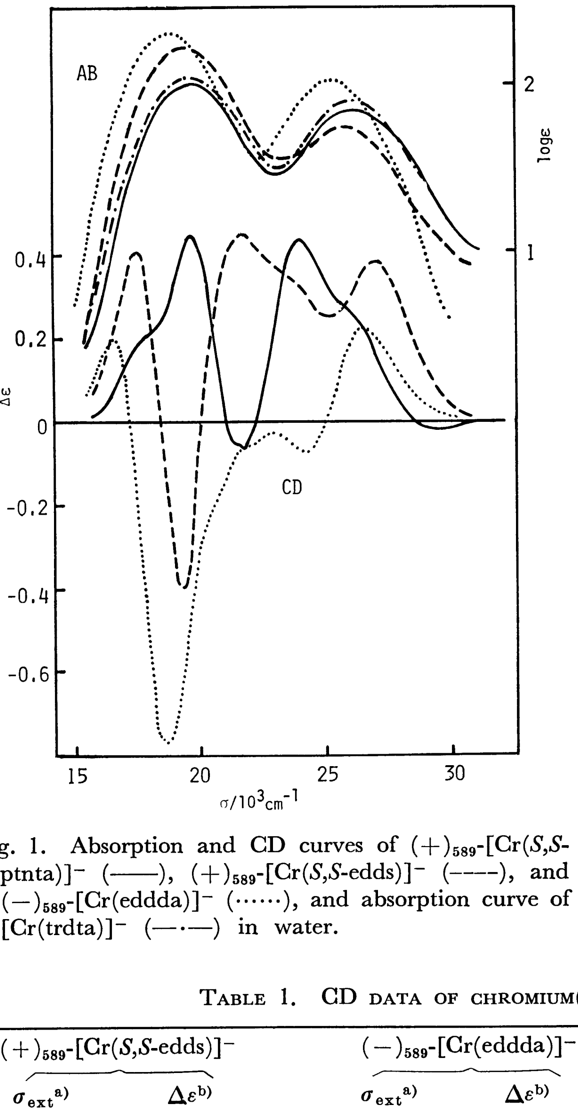

Fig. 1. Absorption and CD curves of $(+)_{589}$-[Cr(*S*,*S*-ptnta)]$^-$ (——), $(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$ (----), and $(-)_{589}$-[Cr(eddda)]$^-$ (······), and absorption curve of [Cr(trdta)]$^-$ (—·—) in water.

CD Spectra in the d-d Transitions. On the assumption of effective C_2 symmetry for the complexes of this type, the lowest-frequency CD component in the first band region is assigned to the $^4B(C_2)$ state, for which the CD signs are considered to be the same as those for the $^4B(C_2)$ state with $^4E(^4T_{2g})$ trigonal parentage or with $^4E(^4T_{2g})$ tetragonal parentage as in the case of the $^1T_{1g} \leftarrow {}^1A_{1g}$ transitions for Co(III) complexes.4,5b,c,11)

TABLE 1. CD DATA OF CHROMIUM(III) COMPLEXES WITH edta TYPE LIGANDS

$(+)_{589}$-[Cr(*S*,*S*-edds)]$^-$		$(-)_{589}$-[Cr(eddda)]$^-$		$(+)_{589}$-[Cr(*S*,*S*-ptnta)]$^-$		Assignment(D_4)
σ_{ext} a)	$\Delta\varepsilon$ b)	σ_{ext} a)	$\Delta\varepsilon$ b)	σ_{ext} a)	$\Delta\varepsilon$ b)	
				13.92	(−0.0038)	$^2E(^2T_{1g})$
14.21	(+0.0348)	14.25	(+0.0352)	14.35	(+0.0055)	$^2A_1$ or $^2A_1+{}^2B_1(^2E_g)$
14.38	(−0.0024)			14.46	(+0.0058)	
14.53	(+0.0036)			14.62	(−0.0017)	
14.87	(−0.0120)	14.87	(−0.0092)	14.85	(+0.0014)	
				15.00	(−0.0021)	
15.44	(+0.0690)	15.26	(+0.0780)	15.31	(+0.0260)	$^2A_2(^2T_{1g})$
17.54	(+0.409) c)	16.53	(+0.207) c)	18.00	(*ca.* +0.2) c)	$^4E(^4T_{2g})$
19.33	(−0.396)	18.62	(−0.765)	19.67	(+0.442)	
21.83	(+0.453)	20.33	(*ca.* −0.19)	21.67	(−0.064)	$^4B_2(^4T_{2g})$
23.33	(*ca.* +0.33)	22.17	(*ca.* −0.05)	24.00	(+0.435)	
27.03	(+0.386)	24.10	(−0.075)	25.25	(*ca.* +0.27)	
		26.53	(+0.23)	29.66	(−0.024)	

a) In units of 10^3 cm^{-1}. b) The $\Delta\varepsilon$ values are given in units of mol^{-1} dm^3 cm^{-1}. c) This band is assigned to the $^4B(C_2)$ state (see Text).

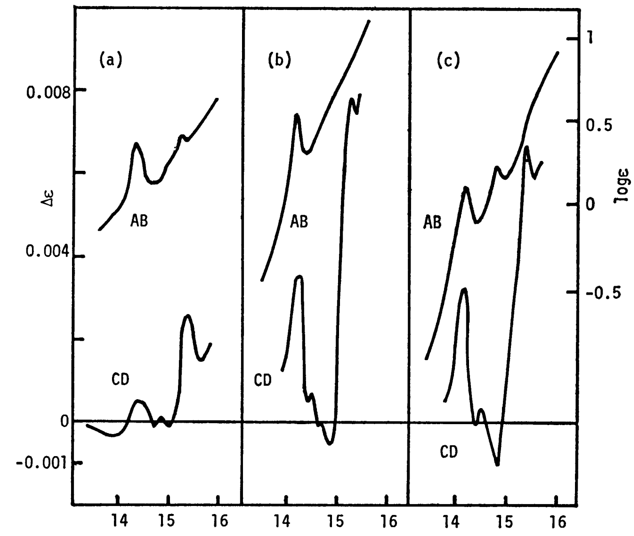

Fig. 2. Absorption (right side ordinate) and CD (left side ordinate) curves in the spin-forbidden transitions of (a) $(+)_{589}$-$[Cr(S,S\text{-ptnta})]^-$, (b) $(-)_{589}$-$[Cr(eddda)]^-$, and (c) $(+)_{589}$-$[Cr(S,S\text{-edds})]^-$ in water.

The CD spectra in the spin-forbidden transitions of the present complexes are shown together with the corresponding absorption spectra in Fig. 2. $(+)_{589}$-$[Cr(S,S\text{-edds})]^-$ and $(-)_{589}$-$[Cr(eddda)]^-$ give a positive CD component near 15300 cm^{-1} and a positive CD one at 14300 cm^{-1} where the lowest-frequency absorption peak is observed. The positions of these two CD peaks coincide with those of the low-temperature absorption bands observed for bis(iminodiacetato)chromate(III) complex.[12] In the region between two strong CD peaks with a positive sign, a weak negative CD component is observed at 14870 cm^{-1} for both the *S,S*-edds and $(-)_{589}$-eddda complexes as shown in Fig. 2 and Table 1. Assuming pseudo D_3 symmetry for the complexes, the three CD peaks, (+) near 14300 cm^{-1}, (−) at 14870 cm^{-1}, and (+) near 15300 cm^{-1}, may be assigned to the $^2E(^2E_g)$, $^2A_2(^2T_{1g})$, and $^2E(^2T_{1g})$ states from the lower frequency side as in the case of $(+)_{546}$-$[Cr(ox)_2(en)]^-$.[1] The similar CD behavior is observed in the spin-forbidden transitions of the *S,S*-ptnta Cr(III) complex, except for the lowest-frequency CD component with a negative sign as shown in Fig. 2. That is, the positive CD peak at 14350 cm^{-1} corresponding to the lowest-frequency absorption peak of the *S,S*-ptnta complex and the positive highest-frequency CD peak may be due to the $^2E(^2E_g)$ and $^2E(^2T_{1g})$ states, respectively, as for the *S,S*-edds and $(-)_{589}$-eddda complexes. In the region corresponding to the $^2A_2 \leftarrow {}^4A_2$ transition, three weak CD peaks with alternate signs are observed for the *S,S*-ptnta complex. In view of these CD behavior, it appears that the correlation between the CD in the spin-forbidden transitions and the absolute configurations for tris-chelate complexes[1] holds also for the present complexes; the complexes giving two $^2E(D_3)$ CD components with a positive sign take a $\Lambda(\Delta\Delta\Lambda)$ configuration. From a theoretical point of view,[1] the rotatianal strengths for the $^2E(^2E_g) \leftarrow {}^4A_2$ transition and those for the $^2E(^2T_{1g}) \leftarrow {}^4A_2$ transition in trigonal field are predicted to have the same signs as the net rotational strengths for the $^4T_{2g} \leftarrow {}^4A_{2g}$ transitions and the rotational strengths for the $^4E(^4T_{2g}) \leftarrow {}^4A_2$ transition, respectively. In the case of the *S,S*-edds and *S,S*-ptnta complexes, such a theoretical relation for the rotational strengths in trigonal field is applicable to CD in the region of the $^2E(^2E_g)$ and $^2E(^2T_{1g}) \leftarrow {}^4A_2$ transitions, because the net CD signs in the first band region and the signs of the $^4B(C_2)$ CD component with ^4E trigonal parentage are positive. The CD of $(-)_{589}$-$[Cr(eddda)]^-$, however, does not follow this relation, since this complex gives a major negative CD band in the first band region, which is presumed to result in the sign inversion of the lowest-frequency CD in the spin-forbidden transitions. Assuming tetragonal symmetry for the complexes, the theoretical relation predicts[2] that the positive highest-frequency CD peak near 15300 cm^{-1} is due to the $^2A_2(^2T_{1g})$ state and the lower frequency one near 14300 cm^{-1} to the $^2A_1(^2E_g)$ or $[^2A_1+{}^2B_1](^2E_g)$ state; the rotational strengths for the $^2A_2$ and $^2A_1 \leftarrow {}^4B_1$ transitions being borrowed from those for the $^4E(^4T_{2g}) \leftarrow {}^4B_1$ transition, and the sum of the rotational strengths for the $^2A_1$ and $^2B_1 \leftarrow {}^4B_1$ transitions from the net one for the $^4T_{2g} \leftarrow {}^4A_{2g}$ transition. The remaining CD peaks may correspond to the $^2E(^2T_{1g})$ state. For $(-)_{589}$-$[Cr(eddda)]^-$, the assignment of the lowest-frequency CD peak to the $[^2A_1+{}^2B_1](^2E_g)$ state is questionable as in the case of the trigonal assumption as mentioned before, because the CD sign of this component is opposite to that of the major component in the first band region. Such a failure of the theoretical relations for the CD in the d-d transitions may be explained by the following approach.

The CD pattern in the spin-forbidden transitions of the *S,S*-edds complex is similar as a whole to that of the $(-)_{589}$-eddda complex despite the different CD patterns at the higher frequency side of the first band for these complexes as in Figs. 1 and 2. This fact suggests that the rotational strengths for the spin-forbidden transitions are little affected by those for the higher-frequency components of the $^4T_{2g} \leftarrow {}^4A_{2g}$ transitions but are dominantly borrowed from those for the lowest-frequency $^4B(C_2)^4T_{2g}(O_h) \leftarrow {}^4B(C_2)^4A_{2g}(O_h)$ transition. This intensity borrowing mechanism may be ascribed mainly to the difference in energy intervals between the doublet states and the quartet excited states as has been considered for the elucidation of CD in the spin-forbidden transitions of *trans*-$[CrX_2(N)_4]$ type complexes.[2] This mechanism is supported by the fact that the CD intensities of the spin-forbidden bands for the present complexes increase in the order of increasing the lower-frequency shift of the lowest-frequency $^4B(C_2)$ CD component as shown in Figs. 1, 2, and Table 1. In such a situation, the contribution to the CD intensities of the spin-forbidden bands from the CD intensity of the negative major spin-allowed band of $(-)_{589}$-$[Cr(eddda)]^-$ is estimated to be only one-half that from the CD intensity of the positive lowest frequency band in the first band region. Therefore, it is probable that the relatively strong CD intensities of two positive peaks near 14300 and 15300 cm^{-1} are accounted for by the dominant contribution from the CD intensity of the positive lowest frequency $^4B(C_2)$ component. And the minor contribution from the CD intensity of the negative higher-frequency band in the first band region may be responsible for the weakness of the CD peaks near 15000 cm^{-1}. However, the following consideration for the $^2E(^2T_{1g})$ state suggests that the assignment of the negative CD peaks near 15000 cm^{-1} to the electronic transitions is doubtful.

At the lowest frequency side in the spin-forbidden absorption band region for the *S*,*S*-ptnta complex, a negative CD peak is observed as in Fig. 2. The assignment for this CD component is attempted as follows. On the one hand, this CD peak is assumed to correspond to one of the splitting components of the ^2E_g state which arise from the interplay of the spin-orbit coupling and trigonal field,[13] as in the case of $(+)_{546}$-[Cr(acac)$_2$(en)]$^+$ and $(-)_{589}$-[Cr(biguanide)$_3$]$^{3+}$.[1] Then, provided that two positive lower-frequency CD components and a negative higher-frequency one are assigned to the $^4E(D_3)$ and $^4A_2(D_3)$ states, respectively, for the present complex, it may be predicted by the use of Eq. 6 and Table 2 of Ref. 1 that the CD components of the $^2E(^2E_g)$ splitting states, $\bar{E}$ and $2\bar{A}$, have positive and negative sings, respectively, and locate in this order from the lower frequency side. For the *S*,*S*-ptnta complex, however, this predicted CD pattern is reverse to the observed one. Thus, the lowest-frequency CD peak of this complex may not be assigned to one of the splitting components of the ^2E_g state.

On the other hand, the assumption of the holohedrized tetragonal symmetry for this complex makes alternative assignments possible. In this case, the positive lower-frequency and the negative higher-frequency CD components in the first band region are due to the ^4E and $^4B_2$ states, respectively. From the detailed analyses of the low temperature spin-forbidden absorption and luminescence spectra of Na[Cr(ida)$_2$]·1.5H$_2$O (ida^{2-}=iminodiacetate),[12] it has been revealed that the $^2E(^2T_{1g})$ state is largely shifted to the lower frequency than the $^2A_1$ and $^2B_1(^2E_g)$ states. And this shift is considered to occur owing to the large configurational interaction between the $^2E(^2T_{1g})$ and $^2E(^2T_{2g})$ states as in the case of *trans*-[CrF$_2$(en)$_2$]ClO$_4$.[14] The present *S*,*S*-ptnta complex has the identical chromophore to the bis(iminodiacetato)chromate(III) complex except that two ida^{2-} ligands are joined by a 1,3-dimethyltrimethylene linkage. The doublet states for the *S*,*S*-ptnta complex are expected to behave similarly to the bis-ida complex as has been revealed from the magnetic circular dichroism measurements of several Cr(III) complexes with edta analogues.[15] Accordingly, the lowest-frequency CD component with a negative sign may be assigned to the $^2E(^2T_{1g})$ state in tetragonal field, though the position of this CD peak is shifted to the higher frequency than that of the absorption and phosphorescence bands.[12] The negative sign of this CD peak is elucidated by taking into account both the configurational interaction between the upper $^2E(^2T_{2g})$ and the lower $^2E(^2T_{1g})$ states and the strong spin-orbit coupling between the $^2E(^2T_{2g})$ state and the nearby $^4B_2(^4T_{2g})$ state, which may give rise to the splitting of the negative CD peaks near 22000 cm^{-1} as shown in Fig. 1. Then, the $^2E(^2T_{1g})\leftarrow{}^4B_1(^4A_{2g})$ transition attains the negative rotational strength by acquiring a piece of the negative rotational strength for the $^4B_2(^4T_{2g})\leftarrow{}^4B_1(^4A_{2g})$ transition to a larger extent than a piece of the positive rotational strength for the $^4E(^4T_{2g})\leftarrow{}^4B_1(^4A_{2g})$ transitions as has been found for the CD of *trans*-[CrX$_2$(N)$_4$] type complexes with chiral diamines.[2] Thus, the assignments of these CD components to the electronic transitions suggest that the remaining weak CD peaks are due to the vibronic origin rather than the electronic one.

In conclusion, the CD behavior in the whole region of the spin-forbidden transitions for $(+)_{589}$-[Cr(*S*,*S*-ptnta)]$^-$ and related complexes is elucidated on the tetragonal symmetry assumption more consistently than on the trigonal one as summarized in Table 1.

The authors wish to express their thanks to Prof. Yoichi Shimura, Osaka University, for his kind guidance and continuous encouragement throughout this work and also for making the circular dichroism and infrared spectra available for the present work.

References

1) S. Kaizaki, J. Hidaka, and Y. Shimura, *Inorg. Chem.*, **12**, 142 (1973).

2) S. Kaizaki and Y. Shimura, *Bull. Chem. Soc. Jpn.*, **48**, 3611 (1975).

3) S. Kaizaki and M. Ito, *Bull. Chem. Soc. Jpn.*, **54**, 2499 (1981).

4) D. J. Radanovic and B. E. Douglas, *J. Coord. Chem.*, **4**, 191 (1975).

5) a) C. W. Van Saun and B. E. Douglas, *Inorg. Chem.*, **8**, 1145 (1969); b) W. Byers and B. E. Douglas, *ibid.*, **11**, 1470 (1972); c) D. J. Radanovic and B. E. Douglas, *ibid.*, **14**, 6 (1975); d) G. G. Hawn, C. A. Chang, and B. E. Douglas, *ibid.*, **18**, 1266 (1979).

6) F. Mizukami, H. Ito, J. Fujita, and K. Saito, *Bull. Chem. Soc. Jpn.*, **43**, 3633 (1970).

7) a) J. A. Neal and N. J. Rose, *Inorg. Chem.*, **7**, 2405 (1968); b) J. I. Legg and J. A. Neal, *ibid.*, **12**, 1805 (1973); c) W. T. Jordan and J. I. Legg, *ibid.*, **13**, 2271 (1974).

8) C. J. Hawkins and E. Larsen, *Acta Chem. Scand.*, **19**, 1969 (1965).

9) J. A. Weyh and R. E. Hamm, *Inorg. Chem.*, **7**, 2431 (1968); H. Ogino, J. J. Chung, and N. Tanaka, *Inorg. Nucl. Chem. Lett.*, **7**, 125 (1971).

10) F. T. Helm, W. H. Watson, D. J. Radanovic, and B. E. Douglas, *Inorg. Chem.*, **16**, 2351 (1977).

11) A. J. McCaffery, S. F. Mason, and B. J. Norman, *J. Chem. Soc.*, **1965**, 5094.

12) C. D. Flint and A. P. Matthews, *J. Chem. Soc., Faraday Trans. 2*, **71**, 379 (1975).

13) S. Sugano and Y. Tanabe, *J. Phys. Soc. Jpn.*, **13**, 880 (1958).

14) C. D. Flint and A. P. Matthews, *J. Chem. Soc., Faraday Trans. 2*, **70**, 1307 (1974).

15) S. Kaizaki and H. Mori, *Chem. Lett.*, **1981**, 567.