

Superposition Model Analysis of Cr^{3+} ions Doped CSA at Orthorhombic Symmetry Site

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Abstract— Superposition model (SPM) is employed to determine zero field splitting (ZFS) parameters (ZFSPs) and crystal field parameters (CFPs) of Cr^{3+} doped $\text{Cs}_2\text{NaAl}_3\text{F}_{12}$ (CSA). Substitutional and interstitial sites for Cr^{3+} ion in CSA crystal and distortion are taken into consideration for computation. The ZFSPs thus obtained are in good agreement with the experimental values when distortion is incorporated. The optical energy band positions for Cr^{3+} in CAS are calculated using CFPs determined from SPM and CFA package. The results show that Cr^{3+} ions substitute CAS lattice at Al^{3+} sites.

Keywords— Superposition model; Crystal field: zero-field splitting; Optical spectroscopy; Cr^{3+} ions in CSA.

I. INTRODUCTION

The present investigation gives a contribution to a general work about the correlations between spin Hamiltonian parameters of 3d ions in an octahedral crystal field and the host crystal structure. The superposition model (SPM) suggests that the spin Hamiltonian parameters result from individual contributions of nearest neighbours of the paramagnetic ion. It has been frequently employed to obtain spin Hamiltonian parameters of various metal ions doped in crystals [1-6]

The EPR study of Cr^{3+} ion doped $\text{Cs}_2\text{NaAl}_3\text{F}_{12}$ (CSA) crystals [7] at room temperature has been performed [8] and the parameters g, D, and E are obtained. The present paper deals with the superposition model (SPM) analysis of the zero field splitting parameters (ZFSPs) and the crystal field parameters (CFPs) for Cr^{3+} ions in CAS. The aim is to find the ZFSPs for Cr^{3+} ions at different sites, to obtain the CFPs for Cr^{3+} ions and to know the existence of structural distortion about the Cr^{3+} ions at various sites. The optical energy band positions for Cr^{3+} in CSA are computed using CFPs and CFA package. The predicted ZFSPs and CFPs may be useful in future investigations for technological applications of various crystals.

This section was dedicated to the introduction of the subject and aim of the study carried out. The rest of the paper is organized as given below. Section II presents the related work associated with the SPM/ZFS and SPM/CF analysis. Section III discusses the methods used for computation of ZFSPs and CFPs. Section IV gives the results and its discussion. In section V, conclusion of the study and future scope for improvement are presented.

II. RELATED WORK

The SPM/ZFS and SPM/CF analysis have been done for various ion-host systems [9-19]. The crystal structure of $\text{Cs}_2\text{NaAl}_3\text{F}_{12}$ (CSA) crystal is trigonal (rhombohedral) having unit cell parameters $a = 0.7310$ nm, $\alpha = 57.450^\circ$, space group $R\bar{3}m$ and $Z = 1$ [7]. The structure is built with $(\text{AlF}_4)_n$ layers perpendicular to the trigonal axis. The orientations of the (x, y, z) axes (SAAS-symmetry adopted axis system) relative to the (a, b, c) crystallographic axes are $(x \rightarrow a, y \rightarrow b, z \rightarrow c)$ as shown in Fig. 1.

The structural data in spherical polar coordinates for the Cr^{3+} ions in CSA using positions x, y, z of ligands [7] are shown in Table 1. These data are used for SPM/ZFS and SPM/CF calculations given here for CAS: Cr^{3+} .

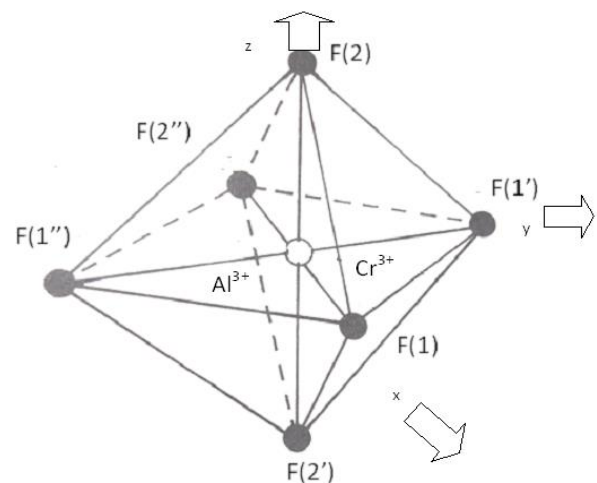


Fig.1. Crystal structure of CSA.

Table 1. Fractional coordinates of Cr³⁺ ion and spherical co-ordinates (R, θ, φ) of ligands in CSA crystal.

Position of Cr ³⁺	Ligands	Spherical co-ordinates of ligands		
		R ^A	θ ⁰	φ ⁰
ND: Substitutional Al (0.5, 0, 0)	F(1)	1.8298	107.8	-55.6
	F(1')	1.8298	0	20.7
	F(1'')	3.5458	52.2	-27.2
	F(2)	3.8207	42.9	8.2
	F(2')	3.8207	99.3	-33.2
	F(2'')	1.7478	110.7	35.8
WD: substitutional Al(0.5, 0, 0)	F(1)	7.6292	64.8	-76.5
	F(1')	7.5861	38.0	-67.0
	F(1'')	8.7768	47.1	-55.9
	F(2)	7.2119	23.7	-38.8
	F(2')	7.2685	64.0	-58.0
	F(2'')	5.3341	53.3	-76.0

ND = No distortion, WD = With distortion.

III. MATERIALS AND METHODS

The ground state energy levels of transition ions in crystals may be found by the spin Hamiltonian with electronic Zeeman (Ze) and ZFS terms [9, 10, 11]:

$$\mathcal{H} = \mathcal{H}_{Ze} + \mathcal{H}_{ZFS} = \mu_B \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S} + \sum B_k^q O_k^q = \mu_B \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S} + \sum f_k b_k^q O_k^q \quad (1)$$

where \mathbf{g} is the spectroscopic splitting factor, μ_B is the Bohr magneton, \mathbf{B} is the applied magnetic field, \mathbf{S} is the effective spin operator and $O_k^q(S_x, S_y, S_z)$ are the extended Stevens operators (ESO) [12, 13]. B_k^q and b_k^q are the related ZFSPs, $f_k = 1/3$ and $1/60$ are the scaling factors for $k = 2$ and 4 , respectively. The ZFS terms in Eq. (1) for Cr³⁺ ion with $S = 3/2$ at orthorhombic symmetry sites are given as [14, 15, 16]:

$$\mathcal{H}_{ZFS} = B_2^0 O_2^0 + B_2^2 O_2^2 = \frac{1}{3} b_2^0 O_2^0 + \frac{1}{3} b_2^2 O_2^2 = D(S_z^2 - \frac{1}{3} S(S+1)) + E(S_x^2 - S_y^2) \quad (2)$$

The conventional orthorhombic ZFSPs (D , E) and (B_k^q , b_k^q) are related as:

$$b_2^0 = D = 3 B_2^0, \quad b_2^2 = 3E = 3 B_2^2 \quad (3)$$

Using SPM [14-16], the ZFSPs (in ESO notation) for a ML_n complex are obtained as:

$$b_k^q = \sum_i \bar{b}_k(R_0) \left(\frac{R_0}{R_i} \right)^{t_k} K_k^q(\theta_i, \phi_i) \quad (4)$$

where (R_i , θ_i , ϕ_i) are the spherical polar coordinates of the i -th ligand. The intrinsic parameters \bar{b}_k give the strength of

the k -th rank ZFS contribution from a ligand located at the distance R_i , and the coordination factors K_k^q provide the geometrical information. K_k^q with $k = 1$ to 6 in the ESO notation are given in [17, 18]. The distance dependence of the intrinsic parameters for a ML_n complex is provided in (5) [14-16, 18], where t_k are the power-law exponents and R_0 is the reference distance [18, 19-22].

From Eq. (4) conventional ZFSPs, D and E are given as [18]:

$$b_2^0 = D = \frac{\bar{b}_2(R_0)}{2} \left[\left(\frac{R_0}{R_i} \right)^{t_2} \sum_i (3 \cos^2 \theta_i - 1) \right] \quad (5)$$

$$b_2^2 = 3E = \frac{b_2^2}{3} = \frac{\bar{b}_2(R_0)}{2} \left[\left(\frac{R_0}{R_i} \right)^{t_2} \sum_i \sin^2 \theta_i \cos 2\phi_i \right]$$

IV. RESULTS AND DISCUSSION

Cr³⁺ ion in CSA may be considered to substitute at the Al³⁺ ion site, and the interstitial site having similar ligand arrangement. The local symmetry at Cr³⁺ ion site is taken to be approximately orthorhombic. In octahedral coordination of Cr³⁺ ion for LiNbO₃ having Cr³⁺-O²⁻ bond, $\bar{b}_2(R_0) = 2.34 \text{ cm}^{-1}$ and $t_2 = -0.12$ [23] have been used to find b_2^0 and b_2^2 . As Cr³⁺ ion in CSA has distorted octahedral coordination (Fig.1) with fluorine as ligands, the b_k^q in the present analysis are determined using the parameters $\bar{b}_2(R_0) = 2.34 \text{ cm}^{-1}$ and $t_2 = -1.035$.

The position of metal ion and spherical coordinates of ligands given in Table 1 are used for calculation. The conventional ZFSPs, D and E of Cr³⁺ ion in CSA crystal are obtained using Eq. (5). The reference distance of 0.120 nm was taken for the determination of ZFSPs [24], and the obtained values of conventional ZFSPs are $|D| = 0.0268 \text{ cm}^{-1}$ and $|E| = 0.0077 \text{ cm}^{-1}$. The ratio b_2^2 / b_2^0 should be within the range (0, 1) for orthorhombic symmetry [25]. In the present study, the ratio $|b_2^2| / |b_2^0| = 0.865$ and $|E| / |D| = 0.288$, in agreement with above for b_2^2 / b_2^0 . However, the values of $|D|$ and $|E|$ do not agree with the experimental values. Hence, with above values of t_2 and reference distance, the ZFSPs $|D|$ and $|E|$ are evaluated for Cr³⁺ at the Al³⁺ site with distortion having position Al³⁺ (0.5, -0.556, -0.5205). The conventional ZFSPs obtained now are $|D| = 0.1527 \text{ cm}^{-1}$, $|E| = 0.0305 \text{ cm}^{-1}$, which are in good agreement with the experimental ones. The ratio $|b_2^2| / |b_2^0| = 0.599$ and $|E| / |D| = 0.199$ which is consistent with [25]. Further, with above values of t_2 and reference distance, the conventional ZFSPs $|D|$ and $|E|$ are determined for Cr³⁺ at the interstitial site but the values

found are quite different from the experimental values and hence these data are not being given here.

The calculated and experimental ZFSPs for Cr³⁺ ion doped CSA are given in Table 2 from which it is found that the ZFSPs |D| and |E| are in good agreement with the experimental ones [8] when the distortion is taken into account.

The CF energy levels of transition metal ions in crystals [26-29], using \mathcal{H}_{CF} [30, 31] and Wybourne operators [9, 30], are obtained as:

$$\mathcal{H}_{CF} = \sum_{kq} B_{kq} C_q^{(k)} \quad (6)$$

Using SPM [14-16], the CFPs in Eq. (6) for a ML_n complex are found as:

$$B_{kq} = \sum_i \bar{A}_k \left(\frac{R_0}{R_i} \right)^{t_k} K_{kq}(\theta_i, \varphi_i). \quad (7)$$

Table 2. Calculated and experimental ZFSPs of Cr³⁺ doped CSA single crystal together with reference distance.

R_0^{A}	Calculated ZFS parameters (cm ⁻¹)				Conventional ZFS parameters (cm ⁻¹)		
	$ b_2^0 $	$ b_2^2 $	$b_2^2/ b_2^0 $	$ b_2^0 $	D	E	E / D
ND	1.2	0.0268	0.0232	0.865	0.0268	0.0077	0.288
WD	1.2	0.1527	0.0915	0.599	0.1527 ^e	0.0305 ^e	0.199
					0.1527 ^e	0.0305 ^e	0.199

ND = No distortion, WD = With distortion, Al³⁺ (0.5, -0.5560, -0.5205)
^e = experimental.

where R₀ represents the reference distance for the site; R_i, θ_i, φ_i are the polar coordinates of the ith ligand and K_{kq} are the coordination factors [26]. To find B_{kq} (k = 2, 4; q = 0, 2, 4); $\bar{A}_2 = 40, 400 \text{ cm}^{-1}$, $t_2 = 1.3$, $\bar{A}_4 = 11, 700 \text{ cm}^{-1}$ and $t_4 = 3.4$ are taken [26]. The calculated B_{kq} parameters are given in Table 3. The ratio B₂₂/B₂₀ = -0.714, which indicates that B_{kq} parameters are standardized [25]. Using B_{kq} parameters in Table 3 and CFA program [27, 28], the CF energy levels of Cr³⁺ ion in CSA crystals are computed. The energy levels of Cr³⁺ ion are calculated by diagonalizing the complete Hamiltonian containing the Coulomb interaction (in terms of the Racah parameters B and C), Trees correction, the spin-orbit interaction, the crystal field Hamiltonian, the spin-spin interaction and the spin-other orbit interaction.

Table 3. B_{kq} parameters of Cr³⁺ doped CSA single crystal.

R_0^{A}	Calculated B _{kq} (cm ⁻¹) Parameters used for CFA package						
	B ₂₀	B ₂₂	B ₄₀	B ₄₂	B ₄₄	B ₂₂ /B ₂₀	
WD	1.2	3695.1	-2638.97	-41.5487	-227.4	-747.50	-0.714

WD = With distortion.

The calculated energy values are shown in Table 4. Since optical absorption data on Cr³⁺: CSA could not be found in literature, the experimental energy values for similar ligand

system Cr³⁺: MgF₂ [32] are given here for comparison. From Table 4, it is seen that there is reasonable agreement between theoretical and experimental band positions. Thus the theoretical study of Cr³⁺ ions entering CSA lattice at Al³⁺ sites supports the experimental finding [32].

Table 4. Experimental and calculated (CFA package) energy band positions of Cr³⁺ doped CSA single crystal.

Transition from $^4A_{2g}(F)$	Experimentally observed band (cm ⁻¹) [32]	Calculated energy band from CFA (cm ⁻¹)
$^4T_{2g}(F)$	14948	14312, 14506, 14599, 14758, 14908, 14961
$^2E_g(G)$	15267	15162, 15492
$^4T_{1g}(F)$	20284	19281, 19392, 19482, 19515, 20655, 20918

(Racah parameters in A, B and C, spin-orbit coupling constant and Trees correction are 0, 516, 3773, 276 and 70 cm⁻¹, respectively)

V. CONCLUSION AND FUTURE SCOPE

The zero-field splitting (ZFS) parameters (ZFSPs) and the crystal field (CF) parameters (CFPs) are evaluated using superposition model (SPM) for Cr³⁺ ions in CSA single crystals. Cr³⁺ ions entering the CSA lattice at Al³⁺ sites, interstitial site and distortion models are considered for calculation. The computed conventional ZFSPs for Cr³⁺ ion at Al³⁺ sites in CSA crystal are in good agreement with the experimental values when distortion is taken into account. It is concluded that the Cr³⁺ ions substitute at Al³⁺ sites in the lattice of CSA. The CF energy values for Cr³⁺ ions at Al³⁺ sites obtained using CFA package and CFPs are in reasonable agreement with the experimental ones. Thus the theoretical results support the experimental conclusion. Modeling procedures considered here may be applicable in future to correlate EPR and optical absorption data for different ion-host systems.

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