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Intensity parametrizations for electric-dipole transitions between Stark components in Er^{3+}:$Y_3Al_5O_{12}$

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ABSTRACT

Trivalent erbium Er^{3+} ($4f^{11}$), as a dopant in the laser host material $Y_3Al_5O_{12}$ (YAG), is a well-known and popular activator ion in a medium having optical, thermal, and mechanical properties suitable for numerous photonic applications. Despite its technological importance, a detailed intensity analysis of transitions between individual Stark components has not previously been attempted. This work presents an intensity analysis for Er:YAG, achieving good agreement between measured and calculated Stark-component transition intensities. Ambiguities in the parametrization due to different possible orientations of the quantization axes are addressed. Use of the "vector crystal field" parametrization resolves additional ambiguities that arise in the transition intensity parameters for low symmetry systems, and allows for a new definition for polarization-resolved Judd-Ofelt parameters, which can have wide-ranging applicability for polarized multiplet-to-multiplet intensity calculations.

1. Introduction

Recently there has been a renewed interest in the examination of crystal-field parametrizations for lanthanide systems having low symmetry. This arises from a need to characterize trivalent lanthanide ions in low symmetry hosts having technological importance [1–4]. One concern for the rationalization of low symmetry crystal-field parameters is that different orientations of the quantization axes may be used, yielding seemingly disparate parameter sets that give identical calculations of theoretical energy levels [5–7].

An issue that has received less attention is the ambiguity this produces in the determination of intensity parameters for calculating transitions between energy levels in these systems. So long as the standard isotropic Judd-Ofelt parametrization for multiplet-to-multiplet transitions is used, the intensity parameters will be independent of the selection choice made for crystal-field quantization direction [8]. However, for the characterization of transitions between Stark-component energy levels, an extended set of intensity parameters must be used that are sensitive to the orientation of the quantization axes.

The analysis of Stark-component transition intensities for low symmetry systems is far scarcer in the literature [9] than are the crystal-field energy level analyses, even though both must be understood to completely rationalize the observed data for technologically important materials. One prominent exception has been the analysis of intensities in D_2 symmetry for Nd:YAG [10]. However, that work neglected to consider parameter ambiguities that arise from multiple local minima [11] and from the multiplicity of identical solutions that arise due to having independent polarization directions [12].

In the present study, we analyze the crystal-field energy level and (Stark) transitions of Er^{3+} ($4f^{11}$) in YAG. Parameter ambiguities potentially arising from different sources are examined. First, we examine the multiple solutions due to alternate choices for the quantization axes. Then we examine the potential for multiple local minima and the multiplicity of identical solutions arising from the independent polarization directions. By redefining the standard Judd-Ofelt parameters as rotationally invariant intensity parameter strengths, we develop the idea of polarization-separated subsets of the Judd-Ofelt parameters.

2. Energy level analysis

The 125 experimental energy levels analyzed in this study span 29 $^{2S+1}L_J$ multiplet manifolds up to 44,000 cm^{-1}. Each energy level in the D_2 site symmetry of the Er^{3+} ion is a Kramer's doublet, the states of each doublet having irrep labels $\Gamma_{1/2}$ and $\Gamma_{3/2}$. The Er:YAG crystals used in the present study are the same samples that were used in the previously reported crystal-field splitting structure study of Gruber et al. [13] but their analysis did not include an intensity analysis of the individual transitions between Stark levels.

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Table 1
Crystal-field energy parameters (in cm^{-1}) for the six different D_2 symmetry orientations of Er:YAG.

Crystal-field parameter	Value Set 1	Value Set 2	Value Set 3
B_0^2	−443(14)	102(17)	341(15)
B_2^2	∓97(12)	±320(10)	∓223(11)
B_0^4	−1687(47)	678(58)	−173(51)
B_2^4	∓538(39)	∓958(31)	∓1496(28)
B_4^4	847(33)	−1132(33)	−420(41)
B_0^6	623(46)	∓628(52)	−1178(41)
B_2^6	±152(46)	∓422(40)	±323(32)
B_4^6	1010(24)	676(31)	529(28)
B_6^6	±187(32)	∓664(30)	±441(46)
D_0^4	15.4(1.7)	−6.2(0.7)	1.6 (0.2)
D_2^4	[±4.9]	[∓8.8]	[∓13.7]
D_4^4	[−7.8]	[10.4]	[3.9]
D_0^6	3.7(0.9)	−3.8(1.0)	−7.0(1.8)
D_2^6	[±0.9]	[∓2.5]	[±1.9]
D_4^6	[6.0]	[4.0]	[3.2]
D_6^6	[±1.1]	[∓4.0]	[±2.6]

The present analysis also has allowed identification of additional energy levels not reported in the previous study.

The electronic energy level structure of Er:YAG is analyzed by means of a model Hamiltonian defined to operate within the $4f^{11}$ electronic configuration of Er^{3+}. All parts of the Hamiltonian that depend upon 4f-electron radial coordinates or describe intermixing from excited configurations are represented as variable parameters [14,15]. Wavefunctions defined within the model Hamiltonian may then be used for evaluation of transition intensities between $4f^{11}$ states.

It is well understood that for lanthanide systems having low site symmetry, different possible orientations of the crystal-field quantization axes will result in different parameter sets that yield identical calculated energy levels. For D_2 symmetry, such as the Er:YAG system examined here, there are three different orientations of the quantization z-axis for which only the nine crystal-field parameters B_q^k listed in the first column of Table 1 are non-zero. For each of these three z-axis orientations, there exist two orientations of the x-axis, resulting in six alternative sets of crystal-field parameters.

Sets of crystal-field parameters corresponding to the six orientations of the quantization axes are presented in the three numerical columns of Table 1, identified as Sets #1 – 3 in Morrison and Leavitt [16] notation. Each column presents two possible sets of parameter values, indicated by the top and bottom symbols of the ±(∓) signs on the $q = 2$, 6 parameters. Set 1a (upper sign) parameters represent the "standardized" parameter set as defined by Rudowicz [17]. Set 3b (lower sign) parameters presented in Table 1 are similar to those previously reported by Gruber et al. [13]. Converted from unit-tensor to spherical-tensor (Wybourne) notation [8], Gruber's crystal-field parameters become: $B_0^2 = 326$, $B_2^2 = 227$, $B_0^4 = -199$, $B_2^4 = -1590$, $B_4^4 = -449$, $B_0^6 = -1164$, $B_0^6 = -283$, $B_0^6 = 496$, and $B_0^6 = -402\,cm^{-1}$.

Values of the correlation-crystal-field delta-function [18,19] parameters D_q^4 and D_q^6 are also presented in the three numerical columns of Table 1. The relationship between the orthogonal correlation-crystal-field parameters [20,21] and the delta-function-restricted parameters are given elsewhere [22]. Rather than directly fitting all nine correlation-crystal-field delta-function parameters, a simplifying assumption has been made that the q-dependence of the D_q^k scales with respect to the B_q^k. That is, the ratios D_q^k/D_0^k are fixed to the respective B_q^k/B_0^k ratios, reducing the number of freely fit correlation-crystal-field parameters to three: D_0^2, D_0^4, and D_0^6. The rank-two parameters D_q^2 do not have a statistically significant influence on the energy level fitting for Er^{3+}, and have therefore been omitted from the fitting presented in Table 1.

The standard deviation of the fitted energy levels with respect to experimentally determined values is $12.66\,cm^{-1}$ (rms error of $11.21\,cm^{-1}$), compared to the rms deviation of $13.20\,cm^{-1}$ reported by Gruber et al. [13]. This fitting improvement is predominately due to two factors: improved correlation-crystal-field parametrization from using the delta-function model rather than the previous arbitrary choice of a single parameter ($G_{10\,A}^4$), and corrected values for the atomic operators $\boldsymbol{m}_j$ and $\boldsymbol{p}_k$. [23] Details of the experimental and calculated energy levels will be presented elsewhere [24].

3. Intensity analysis

Following the notation of Reid and Richardson [25,26] transition linestrengths are calculated by evaluating:

$$S_{i\to f} = e^2 \left| \sum_{\lambda tp} A_{tp}^{\lambda} \sum_{\ell q} \langle \lambda \ell,\, 1(-q) | \mathrm{tp} \rangle (-1)^q \langle \psi_i | \boldsymbol{U}_{\ell}^{\lambda} | \psi_f \rangle \right|^2 + \left| \langle \psi_i | \boldsymbol{m} | \psi_f \rangle \right|^2 \tag{1}$$

where $q = 0, \pm 1$, $\ell = p + q$, and p is restricted by the D_2 site symmetry to even integers with $|p| \le t$. The first term, giving the electric-dipole contribution, is modeled by the A_{tp}^{λ} parameters and uses calculated $U_{\ell}^{(\lambda)}$ matrix elements. The second term gives the magnetic-dipole contribution and is calculated directly. For ground-state transitions of Er^{3+}, only the $^4I_{15/2} \to {}^4I_{13/2}$, $^2K_{15/2}$ and $^2K_{13/2}$ transitions have a magnetic dipole contribution greater than 2% of the observed intensity.

For D_2 site symmetry, there are 18 independent A_{tp}^{λ} parameters, nine which are allowed under the "superposition model" approximation: A_{32}^2, A_{32}^4, A_{52}^4, A_{54}^4, A_{52}^6, A_{54}^6, A_{72}^6, A_{74}^6, and A_{76}^6; and an additional nine parameters which arise through non-cylindrically symmetric ligand/ion interactions: [27] A_{20}^2, A_{22}^2, A_{40}^4, A_{42}^4, A_{44}^4, A_{60}^6, A_{62}^6, A_{64}^6, and A_{66}^6. Non-independent p = negative parameters are related to these by the relationship $(A_{tp}^{\lambda})^* = (-1)^{t+p+1} A_{t-p}^{\lambda}$. Previous work has shown that the additional "non-superposition model" parameters, which incorporate non-cylindrically symmetric interactions due to the "second-nearest" neighboring ligands, are necessary to adequately rationalize transition line strengths in the YAG system [10].

As was the case for the crystal-field and correlation-crystal-field energy level parameters, calculated intensity parameter values for transitions between Stark levels are dependent upon the orientation of the quantization axes. As well, it is now known that once the quantization axes are selected, there are additional ambiguities in the intensity parameters that yield multiple sets of parameter values which give identical calculated intensities.

In order to rationalize these multiple sets of parameters, we use an alternative, "vector crystal field" parametrization $B_{\ell q}^{\lambda}$ [12] which yields the following expansion for transition linestrengths:

$$S_{i\to f} = e^2 \left| \sum_{\lambda \ell q} B_{\ell q}^{\lambda} \langle \psi_i | U_{\ell}^{(\lambda)} | \psi_f \rangle \right|^2 + \langle \psi_i | m | \psi_f \rangle |^2 \tag{2}$$

Comparing Eq. (1) with Eq. (2), we see there is a direct linear transformation between the $B_{\ell q}^{\lambda}$ and A_{tp}^{λ} parametrizations, given by

$$B_{(p+q)q}^{\lambda} = \sum_t A_{tp}^{\lambda} (-1)^q \langle \lambda (p+q),\, 1-q | tp \rangle \tag{3}$$

Table 2
Transformations between the A^{λ}_{tp} and $B^{\lambda}_{\ell i}$ parameter sets in D_2 symmetry.

	A^2_{20}	A^2_{22}	A^2_{32}
B^2_{1x}	$-1/2$	$-1\sqrt{6}$	$1/\sqrt{3}$
B^2_{1y}	$1/2$	$-1\sqrt{6}$	$1/\sqrt{3}$
B^2_{2z}	0	$\sqrt{2/3}$	$1/\sqrt{3}$

	A^4_{32}	A^4_{40}	A^4_{42}	A^4_{44}	A^4_{52}	A^4_{54}
B^4_{1x}	$1/\sqrt{24}$	$-1/2$	$-3\sqrt{40}$	0	$\sqrt{7/30}$	0
B^4_{1y}	$1/\sqrt{24}$	$1/2$	$-3\sqrt{40}$	0	$\sqrt{7/30}$	0
B^4_{2z}	$-1\sqrt{3}$	0	$1/\sqrt{5}$	0	$\sqrt{7/15}$	0
B^4_{3x}	$-7\sqrt{24}$	0	$-\sqrt{7/40}$	$-1\sqrt{10}$	$-1\sqrt{30}$	$\sqrt{2/5}$
B^4_{3y}	$7/\sqrt{24}$	0	$\sqrt{7/40}$	$-1\sqrt{10}$	$1\sqrt{30}$	$\sqrt{2/5}$
B^4_{4z}	0	0	0	$2/\sqrt{5}$	0	$1/\sqrt{5}$

	A^6_{52}	A^6_{54}	A^6_{60}	A^6_{62}	A^6_{64}	A^6_{66}	A^6_{72}	A^6_{74}	A^6_{76}
B^6_{1x}	$\sqrt{5/78}$	0	$-1/2$	$-\sqrt{5/21}$	0	0	$\sqrt{18/91}$	0	0
B^6_{1y}	$\sqrt{5/78}$	0	$1/2$	$-\sqrt{5/21}$	0	0	$\sqrt{18/91}$	0	0
B^6_{2z}	$-4\sqrt{39}$	0	0	$\sqrt{2/21}$	0	0	$\sqrt{45/91}$	0	0
B^6_{3x}	$-3\sqrt{13}$	$1\sqrt{52}$	0	$-\sqrt{3/14}$	$-\sqrt{5/28}$	0	$-\sqrt{5/91}$	$\sqrt{55/182}$	0
B^6_{3y}	$3\sqrt{13}$	$1\sqrt{52}$	0	$\sqrt{3/14}$	$-\sqrt{5/28}$	0	$\sqrt{5/91}$	$\sqrt{55/182}$	0
B^6_{4z}	0	$-\sqrt{10/39}$	0	0	$\sqrt{8/21}$	0	0	$\sqrt{33/91}$	0
B^6_{5x}	0	$-\sqrt{55/156}$	0	0	$-\sqrt{11/84}$	$-1\sqrt{14}$	0	$-\sqrt{3/182}$	$\sqrt{3/7}$
B^6_{5y}	0	$\sqrt{55/156}$	0	0	$\sqrt{11/84}$	$-1\sqrt{14}$	0	$\sqrt{3/182}$	$\sqrt{3/7}$
B^6_{6z}	0	0	0	0	0	$\sqrt{6/7}$	0	0	$1\sqrt{7}$

where the $q = 0, \pm 1$ represent the spherical polarization bases. Converted to Cartesian bases via the following identification:

$$B^{\lambda}_{\ell x} = \frac{-B^{\lambda}_{\ell 1} + B^{\lambda}_{\ell -1}}{\sqrt{2}}, \quad B^{\lambda}_{\ell y} = \frac{i(B^{\lambda}_{\ell 1} + B^{\lambda}_{\ell -1})}{\sqrt{2}}, \quad B^{\lambda}_{\ell z} = B^{\lambda}_{\ell 0} \tag{4}$$

there are six x-polarization $B^{\lambda}_{\ell x}$ parameters, six y-polarization $B^{\lambda}_{\ell y}$ parameters, and six z-polarization $B^{\lambda}_{\ell z}$ parameters in D_2 symmetry. Table 2 presents the transformation matrices between the A^{λ}_{tp} and $B^{\lambda}_{\ell i}$ $(i = x, y, z)$ parametrizations.

In this vector-crystal-field parametrization, the multiplicity of parameter sets is resolved as independent overall signs on each separated-polarization subset of parameters. That is, for each independent polarization direction, the overall sign on the subset of intensity parameters for that polarization is indeterminate. Thus, for D_2 symmetry the three independent polarization directions result in $(2)^3 = 8$ parameter sets that yield identical line strengths.

Table 3 presents the $B^{\lambda}_{\ell i}$ $(i = x, y, z)$ parameters fitted to 88 ground-state transitions by minimizing the standard deviation:

$$\sigma = \sqrt{\sum_i \frac{\left[(E_i - C_i)/E_i\right]^2}{N - P}} \tag{5}$$

where E_i and C_i are the experimental and calculated values, respectively, $N = 88$ data points and $P = 18$ parameters. The fitting standard deviation is $\sigma = 0.280$ (rms error = 0.250), representing a 25% overall deviation between experimental and calculated values. Using a method of random starting parameter values [28] we found that this is an extremely robust solution, with the eightfold solution being the only minimum found from all reasonable ranges of starting parameters. This is in marked contrast to previous calculations of Nd:YAG [11] and oxydiacetate systems [29] where dozens of local minima were found. This provides us with some degree of confidence that the measured intensities are self-consistent and that the calculated parameter values are reliable.

The six numerical columns of Table 3 present parameter values for each of the six crystal-field quantization orientations given in Table 1. Sets "a" correspond to the top signs for the $p = 2, 6$ parameters given in Table 1, while sets "b" correspond to the bottom signs. Each set of parameter values presented in Table 3 represents an eightfold solution. The other seven solutions can be derived from the one presented in Table 3 by independently changing the sign on all $B^{\lambda}_{\ell x}$ parameters, changing the sign on all $B^{\lambda}_{\ell y}$ parameters, and/or changing the sign on all $B^{\lambda}_{\ell z}$ parameters.

The three panels of Table 4 present the same parameter solutions in standard A^{λ}_{tp} notation. The first column presents the solutions of Table 3, with the top sign on the $p = 2, 6$ parameters corresponding to sets "a" and the bottom sign corresponding to sets "b". The second, third, and fourth numerical columns of Table 4 present solutions from Table 3 with the sign on the $B^{\lambda}_{\ell z}$ parameters reversed ("−z" column), the sign on the $B^{\lambda}_{\ell x}$ ($B^{\lambda}_{\ell y}$ for set "b") parameters reversed ("−x/−y" column), and the sign on $B^{\lambda}_{\ell y}$ ($B^{\lambda}_{\ell x}$ for set "b") parameters reversed ("−y/−x" column). The other four solutions may be derived from these four solutions by reversing the sign on all parameters.

It is possible to relate the A^{λ}_{tp} intensity parameters to the Judd-Ofelt parameters for multiplet-to-multiplet transitions by the following expression:

$$\Omega_{\lambda} = \frac{1}{2\lambda + 1} \sum_{tp} |A^{\lambda}_{tp}|^2 = \frac{1}{2\lambda + 1} \sum_{t} \left[\left(A^{\lambda}_{t0}\right)^2 + 2 \sum_{p>0} |A^{\lambda}_{tp}|^2 \right] \tag{6}$$

Table 3
Intensity parameters in *vector crystal-field* notation corresponding to the six different D_2 symmetry orientations given in Table 1. Sets "a" ("b") correspond to the top (bottom) signs for the $p=2$, 6 parameters given in Table 1. The $B^{\lambda}_{\ell x}$ and $B^{\lambda}_{\ell z}$ parameters have units $i \times 10^{-12}$ cm; $B^{\lambda}_{\ell y}$ parameters have units 1×10^{-12} cm. The Ω_λ parameters have units 10^{-20} cm^2.

Parameter	Set 1a	Set 1b	Set 2a	Set 2b	Set 3a	Set 3b
B^2_{1x}	40(15)	−192(13)	192(13)	−88(15)	88(15)	−40(15)
B^2_{1y}	192(13)	−40(15)	88(15)	−192(13)	40(15)	−88(15)
B^2_{2z}	88(15)	−88(15)	40(15)	−40(15)	192(13)	−192(13)
B^4_{1x}	6(13)	−114(12)	−31(16)	−18(15)	−27(14)	−182(15)
B^4_{1y}	114(12)	−6(13)	18(15)	31(16)	182(15)	27(14)
B^4_{2z}	12(17)	−12(17)	−266(11)	266(11)	−117(15)	117(15)
B^4_{3x}	−282(13)	82(16)	−137(12)	−20(16)	3(17)	215(10)
B^4_{3y}	82(16)	−282(13)	−20(16)	−137(12)	215(10)	3(17)
B^4_{4z}	24(14)	24(14)	94(14)	94(14)	77(14)	77(14)
B^6_{1x}	−220(11)	−112(9)	35(8)	6(6)	−88(8)	66(12)
B^6_{1y}	112(9)	220(11)	−6(6)	−35(8)	−66(12)	88(8)
B^6_{2z}	60(7)	−60(7)	−56(9)	56(9)	−110(10)	110(10)
B^6_{3x}	84(11)	−52(8)	173(9)	130(8)	0(7)	160(10)
B^6_{3y}	−52(8)	84(11)	130(8)	173(9)	160(10)	0(7)
B^6_{4z}	−95(8)	−95(8)	−178(12)	−178(12)	−88(9)	−88(9)
B^6_{5x}	43(10)	129(10)	−25(10)	15(8)	97(6)	166(10)
B^6_{5y}	−129(10)	−43(10)	−15(8)	25(10)	−166(10)	−97(6)
B^6_{6z}	−67(7)	67(7)	−150(12)	150(12)	109(8)	−109(8)
Ω_2	1.85	1.85	1.85	1.85	1.85	1.85
Ω_4	2.22	2.22	2.22	2.22	2.22	2.22
Ω_6	1.63	1.63	1.63	1.63	1.63	1.63
Ω_{2x}	0.06	1.48	1.48	0.31	0.31	0.06
Ω_{2y}	1.48	0.06	0.31	1.48	0.06	0.31
Ω_{2z}	0.31	0.31	0.06	0.06	1.48	1.48
Ω_{4x}	1.76	0.44	0.44	0.02	0.02	1.76
Ω_{4y}	0.44	1.76	0.02	0.44	1.76	0.02
Ω_{4z}	0.02	0.02	1.76	1.76	0.44	0.44
Ω_{6x}	0.88	0.49	0.49	0.26	0.26	0.88
Ω_{6y}	0.49	0.88	0.26	0.49	0.88	0.26
Ω_{6z}	0.26	0.26	0.88	0.88	0.49	0.49

That is, under the specific conditions that individual Stark splittings within the initial and final multiplets can be neglected, each Stark level of the initial multiplet can be considered to be essentially equally populated, and the initial and final multiplet states have well-defined J character, summing the electric-dipole contribution of Eq. (1) over all Stark levels gives:

$$S^{\mathrm{ED}}_{\psi J\rightarrow\psi' J'} = e^2 \sum_{\lambda tp} \frac{1}{2\lambda+1} |A^{\lambda}_{tp}|^2 \langle \psi J || \boldsymbol{U}^{(\lambda)} || \psi' J' \rangle^2 = e^2 \sum_{\lambda} \Omega_\lambda \langle \psi J || \boldsymbol{U}^{(\lambda)} || \psi' J' \rangle^2 \quad (7)$$

where the second line reproduces the famous Judd-Ofelt equation. As the specific conditions requiring the absence of crystal-field mixings of different J-multiplets and the absence of crystal-field splittings within the multiplets are not well-met in real systems, one would not expect the Ω_λ parameters calculated from the A^{λ}_{tp} to equal published Ω_λ parameters from multiplet-to-multiplet fittings. However, treated as interaction-strength parameters, they provide rotationally invariant values that may be used for comparison purposes.

The bottom part of each section of Table 4 presents Ω_λ parameters calculated from Eq. (6). As can be seen from this table, the Ω_λ parameters are invariant with respect to both coordinate direction and multiple parameter solutions within a particular coordinate orientation. These values are $\Omega_2 = 1.85 \times 10^{-20}$ cm^2, $\Omega_4 = 2.22 \times 10^{-20}$ cm^2, and $\Omega_6 = 1.63 \times 10^{-20}$ cm^2.

For comparison, literature values of the Judd-Ofelt parameters are, $\Omega_2 = 0.740$, $\Omega_4 = 0.330$, and $\Omega_6 = 1.020 \times 10^{-20}$ cm^2, from Kaminskii [30] and $\Omega_2 = 0.724$, $\Omega_4 = 0.327$, and $\Omega_6 = 0.790 \times 10^{-20}$ cm^2, from Sardar et al. [31]. These values are the same order of magnitude, but somewhat smaller than our calculated values, a phenomenon that has been observed previously for Nd:YAG [10].

When the summation in Eq. (5) is restricted to t = odd terms, only the nine superposition-model allowed parameters are included in the calculated Ω_λ parameters. These values are presented as $\Omega_{\lambda\mathrm{super}}$ in Table 4, along with the percentage of the complete Ω_λ parameter represented. As can be seen from Table 4, these superposition-restricted Ω_λ parameters are not invariant with respect to the different identical parameter solutions. That is, different solutions intermix the t = even with t = odd parameter values differently. Thus, care should be taken with interpreting fitted A^{λ}_{tp} parameters to determine whether the superposition model might be valid in a particular system.

However, it should be noted that for each set of $\Omega_{\lambda\mathrm{super}}$ parameter values listed in Table 4, there is an identical set for each orientation of the crystal-field. This occurs because a rotation of the crystallographic parametrization axis will not intermix A^{λ}_{tp} parameters with different t (or λ), but will only intermix different values of p.

Alternatively, we can use the vector crystal-field $B^{\lambda}_{\ell q}$ parameters to define the Ω_λ parameters. Rewriting Eq. (6) in terms of the $B^{\lambda}_{\ell q}$ gives:

$$\Omega_\lambda = \frac{1}{2\lambda+1} \sum_{\ell q} |B^{\lambda}_{\ell q}|^2 = \frac{2}{2\lambda+1} \sum_{\ell>0} \left(|B^{\lambda}_{\ell x}|^2 + |B^{\lambda}_{\ell y}|^2 + |B^{\lambda}_{\ell z}|^2 \right) \quad (8)$$

where the 2 on the right hand side comes from the ℓ = negative contributions to the summation. As can be seen from the right hand side of Eq. (8), the contributions from each of the three polarizations are separable, allowing one to define polarization-dependent Judd-

Table 4
Intensity parameters in Reid-Richardson notation corresponding to the six different D_2 symmetry orientations given in Table 1. The ± (∓) signs correspond to the respective signs for $p = 2$, 6 parameters in Table 1. Parameters have units $i \times 10^{-12}$ cm. The Ω_λ parameters have units 10^{-20} cm^2.

Set 1 parameter	Value	$-z$ value	$-x/-y$ value	$-y/-x$ value
A^2_{20}	152(19)	152(19)	232(21)	232(21)
A^2_{22}	∓23(17)	∓167(11)	±10(15)	±134(14)
A^2_{32}	±185(11)	±83(17)	±139(13)	∓37(14)
A^4_{32}	±214(14)	±228(13)	∓93(18)	±79(13)
A^4_{40}	108(18)	108(18)	120(17)	−120(17)
A^4_{42}	±101(16)	±90(14)	∓129(17)	±140(18)
A^4_{44}	85(13)	41(16)	−93(14)	137(14)
A^4_{52}	±133(13)	±116(15)	±24(13)	∓7(16)
A^4_{54}	−115(17)	−137(14)	241(13)	−219(13)
A^6_{52}	∓131(10)	∓54(9)	±61(9)	∓138(8)
A^6_{54}	−49(9)	−145(8)	−22(10)	118(9)
A^6_{60}	332(14)	332(14)	108(15)	108(15)
A^6_{62}	±8(10)	∓29(9)	∓129(10)	±166(9)
A^6_{64}	−134(9)	−17(9)	−32(10)	−85(10)
A^6_{66}	∓39(7)	±85(8)	∓16(7)	∓108(7)
A^6_{72}	∓38(8)	∓122(10)	±198(8)	∓113(9)
A^6_{74}	−61(8)	53(9)	−143(9)	29(10)
A^6_{76}	∓81(10)	∓31(10)	∓138(9)	±87(9)
Ω_2	1.85	1.85	1.85	1.85
Ω_4	2.22	2.22	2.22	2.22
Ω_6	1.63	1.63	1.63	1.63
$\Omega_{2\ \mathrm{super}}$	1.37(74%)	0.28(15%)	0.77(42%)	0.05(3%)
$\Omega_{4\ \mathrm{super}}$	1.70(77%)	1.87(84%)	1.49(67%)	1.21(54%)
$\Omega_{6\ \mathrm{super}}$	0.48(30%)	0.66(40%)	1.27(78%)	0.83(51%)

Set 2 parameter	Value	$-y/-x$ value	$-z$ value	$-x/-y$ value
A^2_{20}	−104(23)	−280(15)	−104(23)	280(15)
A^2_{22}	∓82(15)	∓10(14)	∓147(13)	±75(17)
A^2_{32}	±185(11)	±83(17)	±139(13)	∓37(14)
A^4_{32}	±214(14)	±228(13)	∓93(18)	±79(13)
A^4_{40}	49(20)	13(24)	49(20)	−13(24)
A^4_{42}	∓64(14)	∓30(12)	±174(13)	∓207(11)
A^4_{44}	134(13)	121(13)	−34(14)	47(16)
A^4_{52}	∓166(17)	∓177(12)	±197(14)	∓186(14)
A^4_{54}	−57(13)	−32(17)	−141(12)	116(14)
A^6_{52}	±23(9)	∓99(8)	∓49(9)	±171(8)
A^6_{54}	138(10)	120(9)	−42(10)	61(9)
A^6_{60}	−41(9)	−30(11)	−41(9)	30(11)
A^6_{62}	∓52(9)	∓178(7)	∓17(8)	±143(7)
A^6_{64}	−234(9)	114(10)	−14(11)	−106(10)
A^6_{66}	∓128(11)	∓136(11)	±150(11)	∓142(12)
A^6_{72}	∓37(8)	∓92(8)	±43(9)	±13(8)
A^6_{74}	61(10)	−78(10)	275(8)	−136(10)
A^6_{76}	∓83(8)	∓64(10)	±31(9)	∓50(10)
Ω_2	1.85	1.85	1.85	1.85
Ω_4	2.22	2.22	2.22	2.22
Ω_6	1.63	1.63	1.63	1.63
$\Omega_{2\ \mathrm{super}}$	1.37(74%)	0.28(15%)	0.77(42%)	0.05(3%)
$\Omega_{4\ \mathrm{super}}$	1.70(77%)	1.87(84%)	1.49(67%)	1.21(54%)
$\Omega_{6\ \mathrm{super}}$	0.48(30%)	0.66(40%)	1.27(78%)	0.83(51%)

Set 3 parameter	Value	$-x/-y$ value	$-y/-x$ value	$-z$ value
A^2_{20}	−48(24)	128(18)	−128(18)	−48(24)
A^2_{22}	±105(14)	±177(12)	±137(16)	∓209(11)
A^2_{32}	±185(11)	±83(17)	±139(13)	∓37(14)
A^4_{32}	±214(14)	±228(13)	∓93(18)	±79(13)
A^4_{40}	209(20)	155(21)	−155(21)	209(20)
A^4_{42}	∓37(13)	∓60(14)	∓45(13)	±68(16)
A^4_{44}	0(15)	2(15)	136(14)	−138(15)
A^4_{52}	±34(16)	±61(17)	∓221(13)	±194(13)
A^4_{54}	173(15)	169(12)	−100(13)	103(15)
A^6_{52}	±108(9)	±153(9)	∓116(11)	∓33(9)
A^6_{54}	−89(9)	26(8)	64(8)	−178(8)
A^6_{60}	22(14)	−154(14)	154(7)	22(14)
A^6_{62}	±115(9)	±29(10)	∓97(9)	±183(8)
A^6_{64}	−217(8)	−147(8)	38(10)	−108(10)
A^6_{66}	±119(8)	±171(8)	±30(8)	∓82(8)
A^6_{72}	∓109(9)	∓30(10)	∓125(10)	±47(10)
A^6_{74}	1(9)	26(9)	−132(7)	107(9)
A^6_{76}	∓4(8)	∓131(9)	±212(8)	∓86(9)
Ω_2	1.85	1.85	1.85	1.85
Ω_4	2.22	2.22	2.22	2.22

Table 4 (*Continued*)

Set 1 parameter	Value	$-z$ value	$-x/-y$ value	$-y/-x$ value
Ω_6	1.63	1.63	1.63	1.63
$\Omega_{2\ \mathrm{super}}$	1.37(74%)	0.28(15%)	0.77(42%)	0.05(3%)
$\Omega_{4\ \mathrm{super}}$	1.70(77%)	1.87(84%)	1.49(67%)	1.21(54%)
$\Omega_{6\ \mathrm{super}}$	0.48(30%)	0.66(40%)	1.27(78%)	0.83(51%)

Ofelt parameters:

$$\Omega_{\lambda x} = \frac{2}{2\lambda+1}\sum_{\ell>0}|B^{\lambda}_{\ell x}|^2, \quad \Omega_{\lambda y} = \frac{2}{2\lambda+1}\sum_{\ell>0}|B^{\lambda}_{\ell y}|^2,$$

$$\Omega_{\lambda z} = \frac{2}{2\lambda+1}\sum_{\ell>0}|B^{\lambda}_{\ell z}|^2 \tag{9}$$

where

$$\Omega_{\lambda} = \Omega_{\lambda x} + \Omega_{\lambda y} + \Omega_{\lambda z} \tag{10}$$

The bottom part of Table 3 presents the complete Judd-Ofelt parameters Ω_λ along with the separated-polarization $\Omega_{\lambda x}$, $\Omega_{\lambda y}$, and $\Omega_{\lambda z}$ terms. As we have already noted, the Ω_λ parameters are invariant with respect to coordinate rotations. But more than this, each of the six sets of $B^{\lambda}_{\ell i}$ ($i = x, y, z$) parameters yields identical $\Omega_{\lambda i}$ parameter values, but with the x, y, and z subscripts permuted in all six possible ways. This means the $\Omega_{\lambda i}$ may be identified with the crystallographic a, b, and c axis directions, independent of the choice of quantization axes.

This provides justification for the idea of formally separating polarization-dependent parts of the Judd-Ofelt parameters. Presentation of separated polarization-dependent parts of the Judd-Ofelt parameters may be able to provide greater information than the current practice of presenting only one set of (isotropic) Judd-Ofelt parameters. See, for example, Ref. [32], where polarization-dependent measurements were taken, but only isotropic Judd-Ofelt parameters were reported.

4. Conclusion

In this study, we analyzed the energy (Stark) levels of Er^{3+} ($4f^{11}$) in YAG and the intensity of absorption transitions from the ground-state Stark level to individual excited Stark levels having an energy up to 50,000 cm^{-1}. Within this energy range, 88 experimental transition line strengths were analyzed in detail with a weighted $(E_i - C_i)/E_i$ standard deviation of 0.28 (rms deviation of 0.25), indicating a 25% overall deviation between calculated and experimental values.

We have presented the six sets of crystal-field parameters arising from the six alternative choices for parametrization axes in D_2 symmetry, and calculated intensity parameters based upon each of these parametrizations. The eightfold sets of intensity parameters arising from each parametrization have been resolved as three arbitrary sign choices for each polarization subset of vector crystal-field parameters. The vector crystal-field parametrization also leads to a new definition for polarization-resolved Judd-Ofelt parameters, which has the potential to have wide-ranging applicability for polarized Judd-Ofelt-type intensity calculations.

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