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Abstract. On the basis of both the crystal-field theory and the Judd–Ofelt method, the relative intensities and polarization characteristics of the individual transitions between the manifolds $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{9/2}$ for Nd^{3+} ions in an $NdAl_3(BO_3)_4$ crystal were studied by experimental fluorescence spectroscopy and branching ratios. The theoretical results are in agreement with those of laser and fluorescence experiments. In this study, the crystal-field wavefunctions were obtained by fitting the Stark energy sublevels with a constraint condition imposed on the electrostatic ratios of the crystal-field parameter belonging to the same order of k, in a similar manner to that proposed by the present authors previously and referred to as the quasi-three-parameter method. It was shown that the minimum obtained in this way was the only solution corresponding to physical reality.

1. Introduction

Since Judd and Ofelt (JO) [1, 2] presented the general theoretical framework for calculating the intensities of electric dipole transitions within the 4f shell of rare-earth (RE) ions in solutions and solids, it has been used by many workers. It is used to calculate the transition intensities between any two J manifolds $|J\rangle$ and $|J'\rangle$ within the ground $4f^n$ configuration of a trivalent RE in terms of three phenomenological intensity parameters denoted as Ω_2, Ω_4 and Ω_6 [3, 4]. These parameters are determined experimentally by fitting the theoretical line strength to the corresponding line strength measured in absorption spectra. These parameters are then used to calculate the transition intensities between any pair of J manifolds, including fluorescence or excited-state absorption transitions which are inaccessible by direct measurement of absorption spectra. The JO parameters Ω_2, Ω_4 and Ω_6 have been measured for a large number of RE laser materials [3–7].

However, the JO three-parameter method cannot be used to study the relative transition intensities between Stark energy sublevels of different J manifolds. For a laser crystal doped with RE ions, the relative intensities of individual crystal-field (CF) transitions are important in predicting the possibility of laser emission and in describing the characteristics of laser operation. Although the parametrization scheme introduced by Axe [8] can be used to obtain the information mentioned, it is often difficult to calculate all the required parameters because the absorption intensity data resolved at the level of individual crystal-field transitions cannot be obtained under the usual experimental conditions.

In this paper, on the basis of the CF theory and the JO three-parameter method, we try to introduce a procedure for the calculation of the relative transition intensities between Stark sublevels in the same pair of J manifolds together with polarization information about these transitions. The experimental data used are the Stark splitting obtained from fluorescence spectra, and the fluorescence branching ratios of the transitions between the J manifolds considered. As an example, we applied this procedure to a self-activated laser crystal $NdAl_3(BO_3)_4$ (NAB) [9, 10], which had many desirable features, such as a low laser threshold, a high gain, a linearly polarized output, a small beam divergence, and excellent physical and chemical properties. The relative transition intensities between different Stark sublevels belonging to $^4F_{3/2} \rightarrow \, ^4I_{11/2}$ and $^4F_{3/2} \rightarrow \, ^4I_{9/2}$, corresponding to 1.06 μm and possibly 0.9 μm laser emission [9–11], respectively, will be calculated and compared with fluorescence and laser experiments.

2. Crystal field and wavefunction

In the NAB crystal the Nd^{3+} ions occupy D_3 point symmetry sites [12]. The detailed CF Hamiltonian can be expressed as

$$H_{cf} = B_{20}C_{20} + B_{40}C_{40} + B_{43}C_{43} + B_{60}C_{60} + B_{63}C_{63} + B_{66}C_{66} + B_{33}C_{33} + B_{53}C_{53}$$

$$(1)$$

where C_{kq} is the tensor operator and B_{kq} is the crystal field parameter (CFP). B_{43} and B_{63} have been made real by a rotation about the Z axis. The k-even crystal-field components result in the experimentally observed Stark splitting in RE spectra while the k-odd CF components, via configuration admixing, bring about the electric dipole transitions within the $4f^n$ configuration according to the JO theory.

For the NAB crystal, the Stark energy sublevels of $^4F_{3/2}$, $^4I_{11/2}$ and $^4I_{9/2}$ were assigned by the fluorescence spectra of $^4F_{3/2} \rightarrow \, ^4I_{11/2}$ and $^4F_{3/2} \rightarrow \, ^4I_{9/2}$ at 77 K [9]. According to the quasi-three-parameter method proposed [13], the k-even CFPs B_{kq} can be obtained [13] and the eigenfunctions of the Stark sublevels of $^4F_{3/2}$, $^4I_{11/2}$ and $^4I_{9/2}$ manifolds can be calculated simultaneously. The eigenfunctions of these Stark sublevels are expressed as linear combinations of normalized wavefunctions $|Jz\rangle$ simply as

$^4F_{3/2}(2)$ $\quad 0.7071\,i|\tfrac{3}{2}\rangle - 0.7071|-\tfrac{3}{2}\rangle$

$\quad\quad\quad\quad\quad 0.7071|\tfrac{3}{2}\rangle - 0.7071\,i|-\tfrac{3}{2}\rangle$

$^4F_{3/2}(1)$ $\quad |\tfrac{1}{2}\rangle$

$\quad\quad\quad\quad\quad -|-\tfrac{1}{2}\rangle$

$^4I_{11/2}(6)$ $\quad -0.0726|\tfrac{7}{2}\rangle + 0.9950|\tfrac{1}{2}\rangle + 0.0400|-\tfrac{5}{2}\rangle + 0.0560|-\tfrac{11}{2}\rangle$

$\quad\quad\quad\quad\quad -0.0726|-\tfrac{7}{2}\rangle - 0.9950|-\tfrac{1}{2}\rangle + 0.0400|\tfrac{5}{2}\rangle - 0.0560|\tfrac{11}{2}\rangle$

$^4I_{11/2}(5)$ $\quad (0.2296 + 0.0879\,i)|\tfrac{9}{2}\rangle + (0.0879 - 0.2296\,i)|-\tfrac{9}{2}\rangle$

$\quad\quad\quad\quad\quad + (-0.2621 - 0.6090\,i)|\tfrac{3}{2}\rangle + (0.6090 - 0.2621\,i)|-\tfrac{3}{2}\rangle$

$\quad\quad\quad\quad\quad - (-0.2296 + 0.0879\,i)|-\tfrac{9}{2}\rangle + (0.0879 + 0.2296\,i)|\tfrac{9}{2}\rangle$

$\quad\quad\quad\quad\quad + (-0.2621 + 0.6090\,i)|-\tfrac{3}{2}\rangle + (-0.6090 - 0.2621\,i)|\tfrac{3}{2}\rangle$

$^4I_{11/2}(4)$ $\quad 0.4946|\tfrac{7}{2}\rangle - 0.0116|\tfrac{1}{2}\rangle + 0.8446|-\tfrac{5}{2}\rangle + 0.2045|-\tfrac{11}{2}\rangle$

$\qquad\qquad 0.4946|-\tfrac{7}{2}\rangle + 0.0116|-\tfrac{1}{2}\rangle + 0.8446|\tfrac{5}{2}\rangle - 0.2045|\tfrac{11}{2}\rangle$

$^4I_{11/2}(3)$ $\quad (-0.0413 - 0.6617\,i)|\tfrac{9}{2}\rangle + (-0.6617 + 0.0413\,i)|-\tfrac{9}{2}\rangle$

$\qquad\qquad + (0.1651 - 0.1822\,i)|\tfrac{3}{2}\rangle + (0.1822 + 0.1651\,i|-\tfrac{3}{2}\rangle$

$\qquad\qquad (0.0413 - 0.6617\,i)|-\tfrac{9}{2}\rangle + (-0.6617 - 0.0413\,i)|\tfrac{9}{2}\rangle$

$\qquad\qquad + (0.1651 + 0.1822\,i)|-\tfrac{3}{2}\rangle + (-0.1822 + 0.1651\,i)|\tfrac{3}{2}\rangle$

$^4I_{11/2}(2)$ $\quad 0.8434|\tfrac{7}{2}\rangle + 0.0965|\tfrac{1}{2}\rangle - 0.4182|-\tfrac{5}{2}\rangle - 0.3234|-\tfrac{11}{2}\rangle$

$\qquad\qquad 0.8434|-\tfrac{7}{2}\rangle - 0.0965|-\tfrac{1}{2}\rangle - 0.4182|\tfrac{5}{2}\rangle + 0.3234|\tfrac{11}{2}\rangle$

$^4I_{11/2}(1)$ $\quad 0.3560|\tfrac{7}{2}\rangle - 0.3250|\tfrac{1}{2}\rangle + 0.7643|-\tfrac{5}{2}\rangle + 0.4282|-\tfrac{11}{2}\rangle$

$\qquad\qquad 0.3560|-\tfrac{7}{2}\rangle + 0.3250|-\tfrac{1}{2}\rangle + 0.7643|\tfrac{5}{2}\rangle - 0.4282|\tfrac{11}{2}\rangle$

$^4I_{9/2}(5)$ $\quad 0.1746|\tfrac{7}{2}\rangle - 0.9842|\tfrac{1}{2}\rangle + 0.0295|-\tfrac{5}{2}\rangle$

$\qquad\qquad - 0.1746|-\tfrac{7}{2}\rangle - 0.9842|-\tfrac{1}{2}\rangle - 0.0295|\tfrac{5}{2}\rangle$

$^4I_{9/2}(4)$ $\quad - 0.9390|\tfrac{7}{2}\rangle - 0.1755|\tfrac{1}{2}\rangle - 0.2957|-\tfrac{5}{2}\rangle$

$\qquad\qquad 0.9390|-\tfrac{7}{2}\rangle - 0.1755|-\tfrac{1}{2}\rangle + 0.2957|\tfrac{5}{2}\rangle$

$^4I_{9/2}(3)$ $\quad (0.3694 - 0.0536\,i)|\tfrac{9}{2}\rangle + (0.0536 + 0.3694\,i)|-\tfrac{9}{2}\rangle$

$\qquad\qquad + (-0.2404 + 0.5504\,i)|\tfrac{3}{2}\rangle + (0.5504 + 0.2404\,i)|-\tfrac{3}{2}\rangle$

$\qquad\qquad (0.3694 + 0.0536\,i)|-\tfrac{9}{2}\rangle + (-0.0536 + 0.3694\,i)|\tfrac{9}{2}\rangle$

$\qquad\qquad + (0.2404 + 0.5504\,i)|-\tfrac{3}{2}\rangle + (0.5504 - 0.2404\,i)|\tfrac{3}{2}\rangle$

$^4I_{9/2}(2)$ $\quad (-0.1575 - 0.5796\,i)|\tfrac{9}{2}\rangle + (0.5796 - 0.1575\,i)|-\tfrac{9}{2}\rangle$

$\qquad\qquad + (-0.3576 - 0.1070\,i)|\tfrac{3}{2}\rangle + (-0.1070 + 0.3576\,i)|-\tfrac{3}{2}\rangle$

$\qquad\qquad (-0.1575 + 0.5796\,i)|-\tfrac{9}{2}\rangle + (-0.5796 - 0.1575\,i)|\tfrac{9}{2}\rangle$

$\qquad\qquad + (0.3576 - 0.1070\,i)|-\tfrac{3}{2}\rangle + (-0.1070 - 0.3576\,i)|\tfrac{3}{2}\rangle$

$^4I_{9/2}(1)$ $\quad - 0.6578|\tfrac{7}{2}\rangle - 0.5706|\tfrac{1}{2}\rangle - 0.4916|-\tfrac{5}{2}\rangle$

$\qquad\qquad 0.6578|-\tfrac{7}{2}\rangle - 0.5706|-\tfrac{1}{2}\rangle + 0.4916|\tfrac{5}{2}\rangle.$

The wavefunctions are listed in order of decreasing energy. All the Stark sublevels are doubly degenerate and their wavefunctions have the following relationship [14]:

$$\psi = \sum_{J,J_z} a_{J,J_z}|J, J_z\rangle$$

$$\bar{\psi} = \sum_{J,J_z} a_{J,J_z}^*(-1)^{J-J_z}|J, -J_z\rangle.$$

These wavefunctions are normalized and will be used to describe the states of Stark sublevels in the following electric dipole transition calculations.

3. Calculation of relative transition intensities

The JO theory has demonstrated that the k-odd CFPs, via configuration admixing, allow electric dipole transitions within the $4f^n$ configurations to occur. For Nd^{3+} ions in the NAB crystal, as shown in equation (1), the k-odd CF components $B_{33}C_{33}$ and $B_{53}C_{53}$ make the electric dipole transitions possible. The electric dipole moment operators are given by

$$P_\rho^{(1)} = -e \sum_i r_i (C_\rho^{(1)})_i, \qquad C_0^{(1)} = z/r \qquad C_{\pm 1}^{(1)} = (x \pm iy)/r \quad (2)$$

where r_i is the radial coordinate of the ith electron; $\rho = 0$ and ± 1 correspond to π and σ polarization absorption or emission respectively.

Assuming that the $nl^{N-1}l'$ configuration admixes to the nl^N configuration, the electric dipole transition is allowed. For non-allowed mixing of the J states $\langle A|$ and $|B\rangle$, the transition matrix element can be written as

$$\langle A|P^{(1)}|B\rangle = -e \sum_{J_z, J_{z'}} a_{J_z} a_{J_{z'}} \sum_{\lambda, k, q} (-1)^{q+\rho} (2\lambda+1) \begin{pmatrix} 1 & \lambda & k \\ \rho & -(\rho+q) & q \end{pmatrix}$$

$$\times B_{\lambda, k, q} (-1)^{J-J_z} \begin{pmatrix} J & \lambda & J' \\ -J_z & \rho+q & J_z' \end{pmatrix} \langle l^n \alpha SLJ \| U^{(\lambda)} \| l^n \alpha' SL'J' \rangle$$

$$(3)$$

$$B_{\lambda, k, q} = 2B_{k,q} \begin{Bmatrix} l & l & \lambda \\ 1 & k & l' \end{Bmatrix} \langle l \| C^{(1)} \| l' \rangle \langle l' \| C^{(k)} \| l \rangle \langle nl|r|n'l' \rangle / E_{\alpha\nu} \qquad (4)$$

where B_{kq} are the k-odd CFPs and $E_{\alpha\nu}$ is the energy separation between the initial (or final) states and the intermediate states belonging to a configuration with different parity.

In the case discussed, the energy separations between the initial and final states in the $4f^3$ configuration are much smaller than that between the configurations of $4f^3$ and $4f^2 5d$. Therefore, in the calculation, $E_{\alpha\nu}$ is referred to as the average energy separation of these two configurations.

As the first step, the parameters $B_{\lambda, k, q}$ must be evaluated. The non-zero of the $6-j$ symbols in which λ is a component require that $\lambda \geqslant |J - J'|$. For the configuration f^n, $\lambda = 2, 4, 6$, and only three terms B_{433}, B_{453} and B_{653} in equation (3) should be calculated. The admixing of $4f^3$ and $4f^2 5d$ configurations requires that $l = 3$ and $l' = 2$ in equation (4). Using the relationship

$$\langle l \| C^{(k)} \| l' \rangle = (-1)^l [(2l+1)(2l'+1)]^{1/2} \begin{pmatrix} l & k & l' \\ 0 & 0 & 0 \end{pmatrix} \qquad (5)$$

the following equations result:

$$B_{433} = 0.4467 (B_{33}/E_{\alpha\nu}) \langle 4f|r|5d \rangle = 0.4467 D_{33}$$
$$B_{453} = -0.0724 (B_{53}/E_{\alpha\nu}) \langle 4f|r|5d \rangle = -0.0724 D_{53} \qquad (6)$$
$$B_{653} = -0.4859 (B_{53}/E_{\alpha\nu}) \langle 4f|r|5d \rangle = -0.4859 D_{53}$$

where

$$D_{kq} = (B_{kq}/E_{\alpha\nu}) \langle 4f|r|5d \rangle.$$

Considering the intermediate-coupling approximation, the irreducible matrix elements appearing in equation (3) can be written as [15]

$$\langle f^3 \, {}^4I_{9/2} \| U^{(4)} \| f^3 \, {}^4F_{3/2} \rangle = -0.4779$$

$$\langle f^3 \, {}^4I_{9/2} \| U^{(6)} \| f^3 \, {}^4F_{3/2} \rangle = -0.2316$$

$$\langle f^3 \, {}^4I_{11/2} \| U^{(4)} \| f^3 \, {}^4F_{3/2} \rangle = 0.3769 \tag{7}$$

$$\langle f^3 \, {}^4I_{11/2} \| U^{(6)} \| f^3 \, {}^4F_{3/2} \rangle = 0.6416.$$

To calculate the intensities for the transition from the sublevels of the ${}^4F_{3/2}$ manifold to that of the ${}^4I_{11/2}$ and ${}^4I_{9/2}$ manifolds, the ratio of the particle distribution in the upper Stark level to that in the lower Stark sublevels of ${}^4F_{3/2}$ should be considered. If ΔE is the energy separation of two ${}^4F_{3/2}$ sublevels, then this factor is, by the Boltzman distribution, $\exp(\Delta E/kT)$. Hence, the transition intensities between the sublevels of ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$, all of them containing the parameters D_{kq}, are evaluated. To do this the following equation for the transition intensities of spontaneous radiation was used:

$$A(\alpha J, \alpha' J') = [64\pi^4 \sigma^3 e^2 / 3h(2J' + 1)][n(n^2 + 2)^2 / 9] S_{JJ'}$$

$$S_{JJ'} = \frac{1}{e^2} \sum_{\psi_J, \psi_{J'}} \langle \psi_J | P^{(1)} | \psi_{J'} \rangle^2 \tag{8}$$

where σ is the wavenumber corresponding to the transition and n is the refractive index at this wavelength. If the fluorescence ratio β is expressed as

$$\beta(\alpha J, \alpha' J') = A(\alpha J, \alpha' J') \Big/ \sum_{\alpha', J'} A(\alpha J, \alpha' J') \tag{9}$$

then using the experimental data listed in table 1, the following relationships can be established:

$$D_{33} = 1.1848 A^{1/2} \qquad D_{53} = 1.8963 A^{1/2} \tag{10}$$

where A is a factor which is proportional to the total transition intensities $\sum_{\alpha', J'} A(\alpha J, \alpha' J')$. From equation (9), the relative intensities and polarizations of radiative transitions between sublevels in ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ and ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ can be estimated and these are listed in tables 2–5. In these tables, the emission light polarized parallel and perpendicular to the C axis corresponds to π and σ transitions, respectively.

4. Discussion

The calculated relative values of the transition intensity demonstrate that the transitions between the Stark sublevels of ${}^4F_{3/2}(1) \rightarrow {}^4I_{11/2}(2)$ and ${}^4F_{3/2}(1) \rightarrow {}^4I_{11/2}(3)$ are the most intense transitions. On the other hand, it is shown that the transition of ${}^4F_{3/2}(1) \rightarrow {}^4I_{11/2}(2)$ emits light polarized parallel and perpendicular to the C axis, while that of the ${}^4F_{3/2}(1) \rightarrow {}^4I_{11/2}(3)$ is polarized perpendicular to the C axis only. The results of the laser experiment at 1.06 μm [9] agree well with the above prediction in both the laser emission channel and the polarization characteristics.

Table 1. Spectroscopic experimental results for the NAB crystal.

	n	σ (cm^{-1})	β (%)
${}^4F_{3/2} \rightarrow {}^4I_{11/2}$	1.70977	9.35×10^3	53.5
${}^4F_{3/2} \rightarrow {}^4I_{9/2}$	1.71201	11.4×10^3	34.7

Table 2. Calculated relative values of the transition intensity for $^4F_{3/2}(1) \rightarrow {}^4I_{11/2}$ sublevels in the NAB crystal.

$^4I_{11/2}$	Wavelength (μm)	Transition intensity		
		$\perp C$	$\parallel C$	Total
(1)	1.0622	0.0021	0.0219	0.0240
(2)	1.0638	0.0537	0.1475	0.2012
(3)	1.0660	0.2661	0	0.2661
(4)	1.0670	0.0010	0.0436	0.0446
(5)	1.0794	0.0419	0	0.0419
(6)	1.0821	0.0004	0.0011	0.0015

Table 3. Calculated relative values of the transition intensity for $^4F_{3/2}(2) \rightarrow {}^4I_{11/2}$ sublevels in the NAB crystal.

$^4I_{11/2}$	Wavelength (μm)	Transition intensity		
		$\perp C$	$\parallel C$	Total
(1)	1.0547	0.0404	0	0.0404
(2)	1.0563	0.0031	0	0.0031
(3)	1.0584	0	0.0026	0.0026
(4)	1.0594	0.0485	0	0.0485
(5)	1.0717	0	0.0293	0.0293
(6)	1.0743	0.0072	0	0.0072

Table 4. Calculated relative values of the transition intensity for $^4F_{3/2}(1) \rightarrow {}^4I_{9/2}$ sublevels in the NAB crystal.

$^4I_{9/2}$	Wavelength (μm)	Transition intensity		
		$\perp C$	$\parallel C$	Total
(1)	0.8836	0.0615	0.0119	0.0734
(2)	0.8851	0.0494	0	0.0494
(3)	0.8921	0.0060	0	0.0060
(4)	0.9024	0.1136	0.0018	0.1154
(5)	0.9065	0.0038	$\simeq 0.0$	0.0038

Table 5. Calculated relative values of the transition intensity for $^4F_{3/2}(2) \rightarrow {}^4I_{9/2}$ sublevels in the NAB crystal.

$^4I_{9/2}$	Wavelength (μm)	Transition intensity		
		$\perp C$	$\parallel C$	Total
(1)	0.8784	0.0011	0	0.0011
(2)	0.8799	0	0.0027	0.0027
(3)	0.8868	0	0.0003	0.0003
(4)	0.8970	0.0005	0	0.0005
(5)	0.9010	0.0005	0	0.0005

In order to compare the calculated relative intensities of the transition between Stark sublevels with the experimental fluorescence intensities quantitatively, the factor $f_{calc} = A_{ij}/A$ for the calculated relative intensities and the factor $f_{exp} = T_{ij}/T$ for the corresponding experimental fluorescence intensities are introduced. A_{ij} are the calculated values of transition intensities listed in tables 2–5, $A = \sum_{i,j} A_{ij}$ is the sum of the transitions in a pair of manifolds such as $^4F_{3/2} \rightarrow {}^4I_{11/2}$, T_{ij} are the line strengths of fluorescence between related sublevels and $T = \sum_{i,j} T_{ij}$ is the sum

Table 6. Calculated and experimental line strengths of the fluorescence transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$ for Nd^{3+} in the NAB crystal.

$^4F_{3/2} \rightarrow {}^4I_{11/2}$		f_{exp}	f_{calc}	f_{calc}[a]
(1)	(1)	0.2101	0.0338	0.1640
(1)	(2)	0.3754	0.2832	0.1365
(1)	(3)	0.0543	0.3745	0.1181
(1)	(4)	0.0503	0.0628	0.0417
(1)	(5)	0.0583	0.0589	0.0262
(1)	(6)	0.0327	0.0021	0.1896
(2)	(1)	0.0903	0.0568	0.0313
(2)	(2)	0.0719	0.0044	0.0357
(2)	(3)	0.0256	0.0037	0.0167
(2)	(4)	0.0120	0.0683	0.0816
(2)	(5)	0.0088	0.0412	0.1559
(2)	(6)	0.0104	0.0102	0.0026
RMS			2.4774	5.3276

[a] Values calculated by the conventional CPF method.

of all fluorescence intensities between the manifolds. The experimental fluorescence data of the transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$ published in [9] are used and the results are listed in table 6. To compare the values of f_{calc} and f_{exp}, the RMS deviation of the experimental and calculated line strengths is defined as

$$\text{RMS} = \left(\sum_{i,j} \frac{[(f_{exp} - f_{calc})/f_{exp}]^2}{N} \right)^{1/2} \tag{11}$$

where N is the number of transitions. The calculated RMS values are listed in table 6.

The recent generation of 0.9 μm radiation is very attractive because of the possibility of creating a solid state coherent blue light by frequency doubling of the fundamental 0.9 μm laser. However, there are difficulties arising from the smaller probability of the related transition and the high thermal population of the lower laser level. The prerequisite of $^4F_{3/2} \rightarrow {}^4I_{9/2}$ laser emission (about 0.9 μm) is that the Stark splitting of $^4I_{9/2}$ should be larger, for example, than that in Nd^{3+}: YAG [16], in which the most suitable upper and lower laser levels are $^4F_{3/2}(1)$ and $^4I_{9/2}(5)$. However, from the calculated relative intensities for $^4F_{3/2} \rightarrow {}^4I_{9/2}$ sublevels listed in tables 4 and 5, one can find that the most intense transitions are those of $^4F_{3/2}(1) \rightarrow {}^4I_{9/2}(4)$ and $^4F_{3/2}(1) \rightarrow {}^4I_{9/2}(1)$ but not of $^4F_{3/2}(1) \rightarrow {}^4I_{9/2}(5)$. Therefore, the emission of the laser at 0.9 μm in NAB would be much more difficult than that in Nd: YAG [17, 18].

All the above calculations are based successfully on the quasi-three-parameter method. Utilizing the results from the conventional CPF method [19], which is used to obtain the wavefunctions of the Stark sublevels of $^4F_{3/2}$, $^4I_{11/2}$ and $^4I_{9/2}$ terms as well as the CFPs concerned, the relative transition intensities between sublevels of $^4F_{3/2} \rightarrow {}^4I_{11/2}$ can also be calculated. However, these results are not consistent with those of the laser experiments in both emission channels and polarization characteristics. For example, the most intense transition calculated by the method in [19] is neither of the two transitions shown by both experiments and this work to exhibit laser emission, and the value of the RMS deviation for f_{calc} is 5.33 (table 6), which is much larger than that calculated by the quasi-three-parameter method. Therefore, it is safe to say that, in the crystal-field energy fitting, the quasi-three-parameter method can provide a solution

corresponding to the observed physical situation.

In this way, the crystal-field theory can be used not only in the Stark energy sublevel fitting and CFP evaluation but also in the calculations on the radiative transition between these sublevels. In contrast, the conventional CFP calculation without the constraint condition mentioned above may provide a mathematical minimum which has no real physical meaning [20, 21]. This is one of the reasons why the conventional crystal-field calculation method cannot be further used to analyse radiative emission and absorption problems quantitatively.

5. Conclusion

By means of the quasi-three-parameter method proposed previously, the crystal-field energy levels can be fitted, and the wavefunctions of the Stark energy sublevels, which are the eigenfunctions in this calculation, can be expressed as a linear combination of $|Jz\rangle$. By using the JO theory, the relative transition intensities between Stark energy sublevels in $^4F_{3/2} \rightarrow {}^4I_{11/2}$ and $^4F_{3/2} \rightarrow {}^4I_{9/2}$ were calculated from the experimental data of fluorescence spectra. The calculated results agree well with those of the laser and fluorescence experiments, and the reason for the difficulty in obtaining a laser emission at 0.9 μm in the NAB crystal is discussed. It is demonstrated that the results obtained from conventional CPF calculation cannot be used to explain the observed spectral experimental results other than a fitting of the Stark energy sublevels. This is another way to show that the quasi-three-parameter method in crystal-field theory does correspond to a solution of physical reality.

A method for calculating relative transition intensities between Stark energy sublevels was proposed, by which the laser emission and its polarization and wavelength can be predicted. This feature is useful in the investigations of laser material, and in particular of new laser crystals.

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