Exploration of the electric and magnetic dipole transition mechanism for Nd^{3+} doped Yttrium aluminum borate

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Introduction

In recent years, the borates have attracted considerable attention owing to their excellent optical and chemical behavior in laser and lighting technologies. Among various borates, the most widely used borate for host lattice is probably the yttrium aluminum borate (YAB). When YAB is doped with the rare earth ion called neodymium (Nd^{3+}), it becomes a promising system for generating near-infrared lasers[1] and for self-frequency-doubling lasers because of its high nonlinear optical coefficient and huge stimulated emission cross section.[2]

The crystal structure of $YAl_3(BO_3)_4$ is trigonal with the space group *R*32. Yttrium cations occupy sites in trigonal prisms while aluminum and boron atoms are located in sites with octahedrally coordinated oxygen, respectively. It has been confirmed by previous study that the rare-earth ions will substitute the position of Y^{3+} in the crystal lattice.[3]

Figure 1 gives a visual micro-structure of Nd^{3+} doped YAB.

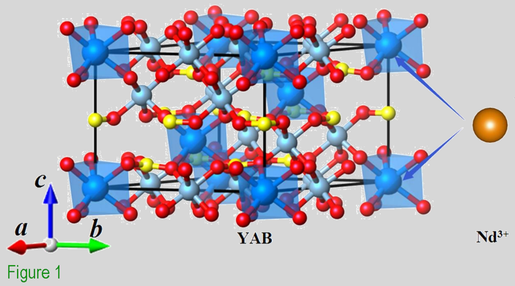

Figure 1

Research Methodology

The atomic energy levels of Nd^{3+} ions in YAB are calculated using our developed crystal-field (CF) theory method.

The parametric Hamiltonian for a $4f^3$ configuration ion can be expressed as:[4-5]

$$H_f = E_{AVE} + \sum_{k=2,4,6} F^k f_k + \zeta_{4f} \times \sum_i \vec{l}_i \cdot \vec{s}_i + \alpha L(L+1) + \beta G(G_2) + \gamma G(R_7) + \sum_{k=2,k=5}^{8} t_k T^k + \sum_{j=0,2,4} M^j m_j + \sum_{k=2,4,6} P^k p_k \quad (1)$$

The CF interaction operator H_{CF} will be introduced when the RE^{3+} ion is doped into the crystal. For the Wybourne normalization, the crystal field Hamiltonian for the D_3 symmetry can be written as:[6]

$$H_{CF} = B_2^0 C_2^0 + B_4^0 C_4^0 + B_6^0 C_6^0 + B_4^3 (C_4^{-3} - C_4^3) + B_6^3 (C_6^{-3} - C_6^3) + B_6^6 (C_6^{-6} + C_6^6). \quad (2)$$

where C_q^k are the normalized spherical-tensor operators. The B_q^k, also known as the crystal field parameters (CFPs), are closely related to the crystal environment and can be determined by the least-squares fit to the observed energy levels or estimated by the superposition model through the use of some crystal field invariants.[7]

Results and discussion

To fully explore the likely transition mechanisms for this system, we carry out a systematic calculations of the transition intensities and radiative features within the $4f^3$ configuration of Nd^{3+} by using a complete free-ion Hamiltonian for the f-shell in conjunction with the crystal-field theory and the Judd-Ofelt theory.

We first focused on the atomic energy calculation of Nd^{3+} ions by using the fitted values of free ion and crystal-field parameters reported by Cascales *et. al*.[8] Noticeably, our result of the energy levels showed reasonable agreements with the previous studies, thus providing significant support for the reliability of our calculation method. The Stark levels as well as the corresponding atomic energy levels are illustrated quantitatively in Figure 2.

Then, the electric dipole (ED) induced transitions for excited states of Nd^{3+} ion in YAB are calculated. It can be clearly seen from Table 1 that the $^4F_{5/2} \rightarrow {}^4I_{9/2}$ and $^2H_{(2)9/2} \rightarrow {}^4I_{9/2}$ transitions are good candidates for laser action at the near-infrared region.

In addition, the magnetic dipole (MD) transitions are also shown with detailed values, providing a large number of strong absorption lines and spontaneous emissions for experimental use.

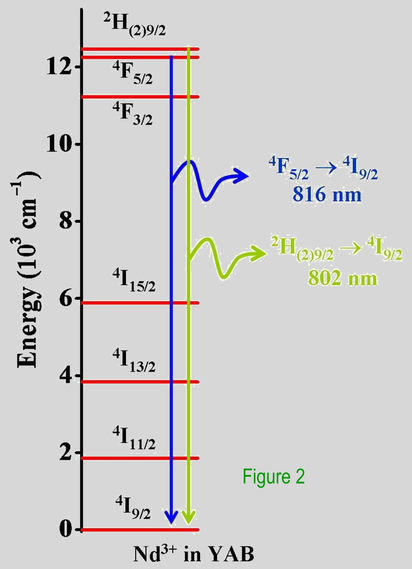

Figure 2

Table 1. Calculated wavelengths (λ), ED (A_{ED}) and MD (A_{MD}) radiative decay rates, branching ratios (β) and radiative lifetimes (τ) for spontaneous emission transitions between J-multiplets in Nd:YAB.

Transition		λ (nm)	A_{ED} (s-1)		A_{MD} (s-1)	β		τ (μs)		
$^4I_{11/2}$	$^4I_{9/2}$	5391	7.8		1.4	1		106835		
$^4I_{13/2}$	$^4I_{9/2}$	2611	21.1		0	0.66		31256		
	$^4I_{11/2}$	5063	9.0		2.0	0.34				
$^4I_{15/2}$	$^4I_{9/2}$	1700	6.3		0	0.17		26837		
	$^4I_{11/2}$	2482	19.6		0	0.53				
	$^4I_{13/2}$	4870	10.0		1.5	0.30				
$^4F_{3/2}$	$^4I_{9/2}$	891	1664.4	1637[1]	0	0.49	0.49[1]	298	302[1]	53[1]
	$^4I_{11/2}$	1067	1441.3	1430[1]	0	0.43	0.43[1]			
	$^4I_{13/2}$	1352	235.1	235[1]	0	0.07	0.07[1]			
	$^4I_{15/2}$	1872	11.5	12[1]	0	0.003	0.005[1]			
$^4F_{5/2}$	**$^4I_{9/2}$**	**816**	**2627.1**		**0**	**0.63**		**241**		
	$^4I_{11/2}$	961	642.5		0	0.16				
	$^4I_{13/2}$	1187	763.0		0	0.18				
	$^4I_{15/2}$	1570	109.1		0	0.03				
$^2H_{(2)9/2}$	**$^4I_{9/2}$**	**802**	**296.7**		**6.1**	**0.50**		**1653**		
	$^4I_{11/2}$	942	40.1		3.7	0.07				
	$^4I_{13/2}$	1158	122.9		0	0.20				
	$^4I_{15/2}$	1519	135.2		0	0.23				
$^4F_{7/2}$	$^4I_{9/2}$	756	1573.8		0	0.39		246		
	$^4I_{11/2}$	879	1405.5		0	0.34				
	$^4I_{13/2}$	1064	608.6		0	0.15				
	$^4I_{15/2}$	1361	475.8		0	0.12				

Conclusion

In summary, we performed a systematic exploration of the electric and magnetic dipole transitions between the excited states of Nd^{3+} doped YAB crystal. Based on the fitted values of free ion and crystal-field parameters reported by previous study, the wavelengths, radiative decay rates, branching ratios and radiative lifetimes for spontaneous emission transitions between J-multiplets in Nd:YAB are reasonably predicted and compared with some finding available in the literature. More importantly, our theoretical results indicate that the $^4F_{5/2} \rightarrow {}^4I_{9/2}$ and $^2H_{(2)9/2} \rightarrow {}^4I_{9/2}$ transitions are good candidates for laser action at the near-infrared region.

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