

Supporting Information for

**Judd-Ofelt analysis of high erbium content yttrium-aluminum and
yttrium-scandium-aluminum garnet ceramics**

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Judd-Ofelt theory

The Judd-Ofelt theory[1,2] is the most suitable one to characterize the intensities of electric-dipole $4f$ transitions in lanthanide ions. This theory is correct both for well-crystalline and disordered media such as single crystals and glasses.

According to the Judd-Ofelt theory, the value of the oscillator strength for the $J \rightarrow J'$ transition is determined by the expression:

$$f_{JJ'} = \left(\frac{8\pi^2 mc}{3hn^2 \bar{\lambda}(2J+1)} \right) [\chi^{ed} s_{JJ'}^{ed} + \chi^{md} s_{JJ'}^{md}] \quad (1)$$

where m is the mass of the electron, c is speed of light in vacuum, h is Planck's coefficient, $\bar{\lambda}$ is a mean wavelength of inter-multiplet transition $J \rightarrow J'$, n is a wavelength-dependent refractive index which is determined from Sellmeier's dispersion equation,[3,4] $\chi^{ed} = n(n^2 + 2)^2/9$, $\chi^{md} = n^3$, $s_{JJ'}^{ed}$ and $s_{JJ'}^{md}$ are the line strengths of the electric-dipole and magnetic-dipole transitions, respectively.

The line strength for the electric dipole transition is:

$$s_{JJ'}^{ed} = \sum_{t=2,4,6} \Omega_t |\langle 4f^N J \| U^t \| 4f^N J' \rangle|^2 \quad (2)$$

where $|\langle 4f^N J \| U^t \| 4f^N J' \rangle|^2$ ($t = 2, 4, 6$) are the doubly reduced matrix elements of the unit tensor operators $U(t)$ of rank t ($t = 2, 4, 6$) which depend only on angular momentum while are independent of the host, [5,6] Ω_t are JO parameters.

Line strength for magnetic dipole transitions $s_{JJ'}^{md}$ is calculated by the formula:

$$s_{JJ'}^{md} = \left(\frac{h}{4\pi mc} \right)^2 \Omega_t |\langle 4f^N J \| L + 2S \| 4f^N J' \rangle| \quad (3)$$

where $L+2S$ is magneto-dipole transition operator $J \rightarrow J'$.

However, the values of the oscillator strengths $f_{JJ'}$ of intermultiplet transitions $J \rightarrow J'$ are determined experimentally by the formula:

$$f_{JJ'} = \frac{mc^2}{4\pi N \bar{\lambda}^2} \int \alpha(\lambda) d\lambda \quad (4)$$

where $\int \alpha(\lambda) d\lambda = \Gamma$ is an integral absorption coefficient for transition between the ground and excited states of rare earth ions, N is an activator ion concentration.

The line strengths for the corresponding transitions of rare earth ions were determined experimentally from the values of the integral absorption coefficients by the formula:

$$s_{JJ'} = \frac{3hc(2J+1)}{8\pi^3 ke^2 N \bar{\lambda}} \frac{9n}{(n^2+2)^2} \int \alpha(\lambda) d\lambda \quad (5)$$

where e is the charge of the electron, k is Coulomb's constant. The factor $[9n/(n^2+2)^2]$ represents the local field correction for electric dipole transition for the effective field for the ion in the dielectric host medium of isotropic refractive index n .

The system of linear equations with three variables Ω_2 , Ω_4 , Ω_6 is solved by the Gauss–Jordan method. [7]

The root mean square (*RMS*) of the line strength of the electric-dipole transition (S_{calc}) is deviation between the experiment and calculated line strengths and determined by (6).

$$RMS \Delta s = \sqrt{\sum_{i=1}^M (s_{\text{exp}_i} - s_{\text{calc}_i})^2 / (M-3)} \quad (6)$$

where M is the number of experimental bands using in the above calculation.

The relative error is defined as:

$$RMS \text{ error} = \frac{RMS \Delta s}{RMS s} \times 100\% \quad (7)$$

where

$$RMS s = \sqrt{\sum_{i=1}^M s_{\text{exp}_i}^2 / M} \quad (8)$$

The deviation between the experiment and calculated absorption oscillator strengths, f_{calc} , and relative error are determined similarly.

The radiative transition probabilities $A_{\text{rad}}(J \rightarrow J')$ is calculated for the transitions by using the following expression:

$$A_{\text{rad}}(J \rightarrow J') = \frac{64\pi^4 e^2}{3h(2J+1)\bar{\lambda}^3} \left(\left[\frac{n(n^2+2)^2}{9} \right] s_{ed} + n^3 s_{md} \right) \quad (9)$$

The radiative lifetime τ_{rad} for an excited states (J) is calculated by:

$$\tau_{\text{rad}} = \frac{1}{\sum A_{\text{rad}}(J \rightarrow J')} \quad (10)$$

The emission cross section has been calculated by means of the Fuchtbauer–Ladenburg formula:

$$\sigma(J, J'; \lambda) = \frac{\lambda^2}{8\pi c n^2} \frac{\beta(J, J')}{\tau_j} g(\tilde{\nu}) \quad (11)$$

where λ is the wavelength at the peak emission, $\beta(J, J')$ is the fluorescence branching ratio for the transition from the upper manifold J to the lower manifold J' , τ_j is the radiative lifetime of the excited manifold $^4I_{13/2}$, and $g(\tilde{\nu})$ is the line-shape function. The line-shape function is obtained from the fluorescence spectrum using the following expression:

$$g(\tilde{\nu}) = \frac{I(\tilde{\nu})}{\int I(\tilde{\nu}) d\tilde{\nu}} \quad (12)$$

The intrinsic quantum yield (η) is evaluated from the ratio of the fluorescence (τ_{lum}^{exp}) to radiative decay time (τ_{rad}^{calc}):

$$\eta = \frac{\tau_{lum}^{exp}}{\tau_{rad}^{calc}} \cdot 100 \% \quad (13)$$

The non-radiative multiphonon decay rates may be given by expression (14):

$$W_{NR} = 1/\tau_{lum}^{exp} - A_{rad}(J \rightarrow J') \quad (14)$$

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