SPECTRAL PROPERTIES AND QUANTUM EFFICIENCY OF OPTICAL EMISSION IN PR^{3+}-DOPED $CaAl_4O_7$ AND $SrAl_4O_7$ CRYSTALS

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We have studied the spectral properties of Pr^{3+}-doped $CaAl_4O_7$ and $SrAl_4O_7$ crystals at 77 and 290 K. Emissions from the 4f5d band are detected using two-photon stepwise excitation. We demonstrated a new method to conveniently measure the quantum efficiency of deep UV emission using the two-step excitation and the difference of the integrated spectral intensities. The quantum efficiencies of the emission from the lowest 4f5d state in the two systems have been estimated and are less than 10% at room temperature. The effect of excited state absorption of the lowest 4f5d on the measurement of the quantum efficiency is analyzed. The actual efficiency may be higher when the effect is taken into account, especially under the condition of strong excitation.

Assuming the number of electrons excited from the $^3P_0$ to 4f5d band and relaxed to the lowest state of the band is Δn_0 and the QE of the state is η_1, the number of photons emitted from the state equals to $\eta_1\Delta n_0$. If the QE of $^3P_0$ state is η_0 and branch ratios of 4f5d-$^3H_{4,5,6}$, $^3F_{2,3,4}$ 4f5d-^3P_J/$^1I_6$, and $^3P_0$-$^3F_2$ transitions are β_1, β_1', and β_0, respectively, the emission intensity of 4f5d-$^3H_{4,5,6}$, $^3F_{2,3,4}$, I_1, and the decrease of $^3P_0$-$^3F_2$ emission due to the UV pump, ΔI_0, can be expressed as follows,

$$I_1 = \beta_1\eta_1\Delta n_0, \quad (1)$$

$$\Delta I_0 = \beta_0\eta_0\Delta n_0(1-\beta_1'\eta_1) \quad (2)$$

The transitions from the lowest 4f5d state to ^3P_J or $^1I_6$ are forbidden, nearly all the electrons from 4f5d state relax to $^3H_{4,5,6}$ and $^3F_{2,3,4}$ states, i.e., $\beta_1' \approx 0$ and $\beta_1 \approx 1$. Taking the ratio of the two equations, we have,

$$R = \frac{I_1}{\Delta I_0} = \frac{\eta_1}{\beta_0\eta_0}, \quad (3)$$

or we find the QE of the band,

$$\eta_1 = R\beta_0\eta_0. \quad (4)$$

All the information on the right of Eq. (4) can be obtained from the spectral measurements. Modifications are needed when considering ESA effect of the lowest 4f5d state on the QE measurements.

Fig. 1 shows the excitation spectra of SAO:Pr when the $^3P_0$-$^3F_2$ emission at 647 nm is monitored at 290 K (solid line) and 77 K(dashed line), respectively, and the intensity corrected emission spectra under 463 nm ($^3H_4$-$^3P_1$) excitation also at 290 K (solid line) and 77 K(dashed line), respectively. Fig. 2 represents the Pr:SAO emission spectra (intensity corrected) used for the QE calculation at 290 K. The QE measurement will be intensity dependent. More general, let $y = \gamma_1/(F\sigma_1+\gamma_1)$ and $x = (F\sigma_1+\gamma_1)/(F\sigma_0+\gamma_0)$, $\eta_1/\eta_1(y = 1)$ against x has been plotted in Fig. 3 for different y values, where F is photon flux density, σ_0 and $\sigma_1$absorption cross-sections of $^3P_0$ and the lowest 4f5d state, respectively, and γ_0 and $\gamma_1$decay rates of $^3P_0$ and the lowest 4f5d, respectively.

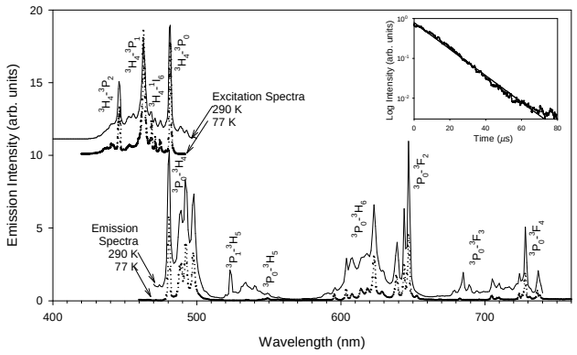

Fig. 1. The excitation spectra of SAO:Pr when the $^3P_0$-$^3F_2$ emission at 647 nm is monitored at 290 K (solid line) and 77 K(dotted line), respectively, and the intensity corrected emission spectra under 463 nm ($^3H_4$-$^3P_1$) excitation also at 290 K (solid line) and 77 K(dotted line), respectively. The detailed peak identifications are also shown in the figure. $^3P_1$-$^3H_5$ emission appearing only at 290 K is due to the thermally populated electrons in $^3P_1$ state from $^3P_0$. The energy difference of the two levels is about 800 cm^{-1}, as shown in the excitation spectra. The inset in the figure depicts the relaxation of $^3P_0$ state at 290 K, yielding a lifetime of 15 μs.

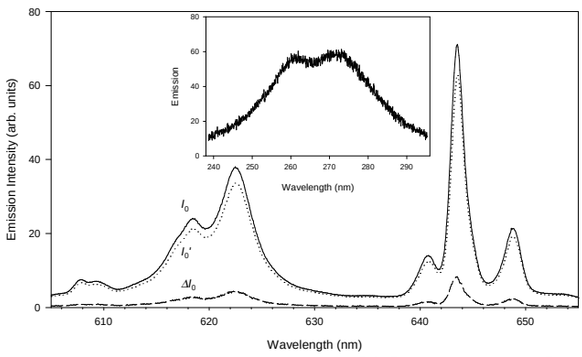

Fig. 2. The emission spectra of Pr:SAO used for the QE calculation at 290 K. I_0 is the spectrum of the $^3P_0$-$^3F_2$ transition under 463 nm excitation, I_0' the spectrum of the $^3P_0$-$^3F_2$ transition under both 463 nm and 355 nm excitations, and ΔI_0 the difference of I_0 and I_0'. Inset: up-converted UV emission (intensity corrected) from the lowest 4f5d state under excitations of 463 nm and 355 nm.

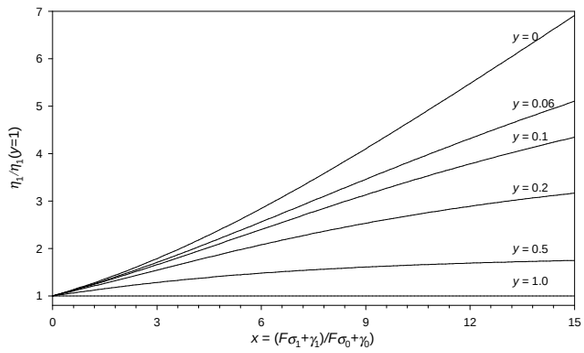

Fig. 3. Ratio $\eta_1/\eta_1(y = 1)$ against x for different y values, where $x = (F\sigma_1+\gamma_1)/(F\sigma_0+\gamma_0)$ and $y = \gamma_1/(F\sigma_1+\gamma_1)$.