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The effect of pressure on the position and fluorescence lifetime for the $^5D_0 \rightarrow {}^7F_2$ transition in $Y_{1.9}Eu_{0.1}O_3$

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Abstract. The aim of this study was to assess the effect of pressure on the position and fluorescence lifetime for the $^5D_0 \rightarrow {}^7F_2$ transition in $Y_{1.9}Eu_{0.1}O_3$ within the 0–104 kbar range. The relationship between the pressure and the line position is given by linear regression: λ (nm) $= 611.29 + 0.011P$ (kbar). The change of the fluorescence lifetime for the $^5D_0 \rightarrow {}^7F_2$ transition with pressure can be expressed by an exponential decay: τ (ms) $= 1.175 + 0.818[1 + 0.006(P - 16.796)^2]^{-1}$.

1. Introduction

Eu^{3+}-doped Y_2O_3 is a well known red phosphor material. The luminescence properties of Eu^{3+} in Y_2O_3 were previously reported by several authors [1, 2, 3]. In Y_2O_3:Eu^{3+} two possible symmetry sites for Eu^{3+} ions are present, namely S_6 and C_2 [1]. In Y_2O_3 host material, Y^{3+} is substituted for with Eu^{3+} and surrounded by six oxygen atoms located at the corners of a cube. The emission spectrum is primarily due to C_2 Eu^{3+} ions [1]. The $^5D_0 \rightarrow {}^7F_2$ transition is the dominant emission (at 611.29 nm) [2], related to one line connected to the $^5D_0 \rightarrow {}^7F_0$ transition and four lines connected to $^5D_0 \rightarrow {}^7F_1$ [1]. The intensity of the $^5D_0 \rightarrow {}^7F_2$ line is three times stronger than any other mentioned [1]. Also, $^5D_0 \rightarrow {}^7F_2$ is far from the nearest line; therefore it is well separated from them. Due to these characteristics, the effect of high pressure on the spectral position can be easily determined. Good separation and high intensity make measuring the fluorescence lifetime for the $^5D_0 \rightarrow {}^7F_2$ line very easy. As Y_2O_3:Eu^{3+} is commonly used as the red phosphor in lamp tubes and lasers, it is of theoretical and technological interest to investigate the effect of high pressure on it. The effect of high pressure and temperature on the crystallographic properties of Y_2O_3:Eu^{3+} is well known [4]. Although the effects of the temperature and the concentration of Eu^{3+} ions, as regards the shifting and lifetime for the $^5D_0 \rightarrow {}^7F_2$ transition in Y_2O_3:Eu^{3+}, have been well examined, the effect of high pressure has not [1, 2]. Therefore the aim of this paper was to investigate the effect of high pressure on the positions and fluorescence lifetime for the $^5D_0 \rightarrow {}^7F_2$ transition in Y_2O_3:Eu^{3+}.

2. Experimental techniques

For pressure generation, a diamond-anvil cell of the NBS type [5] with 1/3 carat stones has been used. Small (≈30 mm) chips of ruby (≈5000 ppm Cr^{3+}) and chips of $Y_{1.9}Eu_{0.1}O_3$

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(5 mol% Eu^{3+}) (≈50 μm) were put in the 300 μm hole of a preindented stainless-steel gasket, together with a methanol–ethanol (4:1) mixture which served as a pressure medium. The starting components for crystal preparation (99.99% pure Y_2O_3 and 99.99% pure Eu_2O_3) were thoroughly mixed in appropriate molar ratios and heated at 1323 K for about 24 h. The presintered material was crushed, reground and again sintered under the same conditions. A spectrofluorometer connected with a multiscaler card in a personal computer was used as the measuring apparatus. For time-resolving measurements, a mechanical chopper was used. The positions of the line R1 and the line connected with the $^5D_0 \rightarrow {}^7F_2$ transition were obtained by using a double optical monochromator with 0.05 nm spectral resolution and a photon counter with multiscaler. The pressure was determined from the redshift of the ruby R1 line [6]. The samples were excited with a green He/Ne laser at 541 nm with an IF filter (541 nm ± 2 nm) in front of the laser. The decay curves were measured at the maximum (for the given pressure) of the line connected with the $^5D_0 \rightarrow {}^7F_2$ transition using a well known method for lifetime measurement [7]. Illumination lasted about 1 ms, and the total measuring time (for one scan) was 10 ms. The data collected by the multiscaler after 10 000 excitation pulses were transferred to a PC to obtain the decay curve, lifetime and standard deviation. The standard deviation for the lifetime measurement was less than 2%.

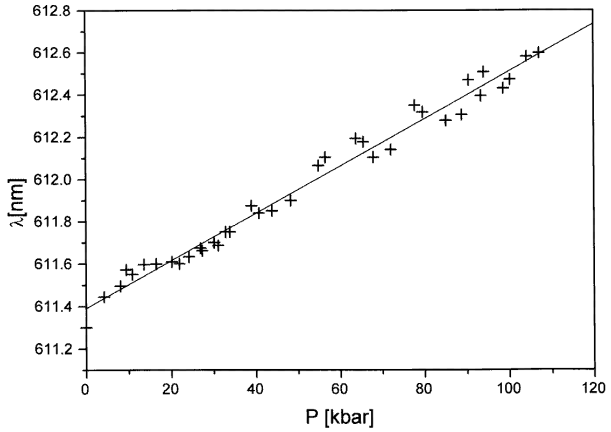

Figure 1. The line shift of the $^5D_0 \rightarrow {}^7F_2$ fluorescence line up to 104 kbar at room temperature. +: experimental data; ——: the curve fitted according to equation (1).

3. Results and discussion

The effect of changing the position of the line connected with the $^5D_0 \rightarrow {}^7F_2$ transition is shown in figure 1. The increasing of the pressure causes a shifting of the line to longer wavelengths. The dependence of the position of the line can be expressed by linear regression:

$$\lambda \text{ (nm)} = 611.29 + 0.011P \text{ (kbar)}. \tag{1}$$

The goodness of fit is 0.9822. In the pressure range examined, the linear dependence of the position of the line connected with $^5D_0 \rightarrow {}^7F_2$ on the pressure is different from those observed for several other crystals. For $Y_2O_2S{:}Eu^{3+}$ [8] the rate is 0.0168 nm $kbar^{-1}$, while it is 0.0162 nm $kbar^{-1}$ for $LaOCl{:}Eu^3$ [9] and 0.0075 nm $kbar^{-1}$ for $P_5O_{14}{:}Eu^{3+}$

[10]. Eu^{3+}:Y_2O_3 shows a smaller pressure shift for the $^5D_0 \rightarrow {}^7F_2$ transition than does ruby for the $^2E \rightarrow {}^4A_2$ transitions (0.0365 nm kbar^{-1}) [6]. Also, all of the luminescence lines derived from $^5D_0 \rightarrow {}^7F_0$ and $^5D_0 \rightarrow {}^7F_1$ transitions of Eu^{3+} ions in Y_2O_3 tend to redshift with increasing pressure but with different rates (-0.0052 nm kbar^{-1} and -0.0046 nm kbar^{-1}, respectively) [11]—less than that for the $^5D_0 \rightarrow {}^7F_2$ transition (0.011 nm kbar^{-1}).

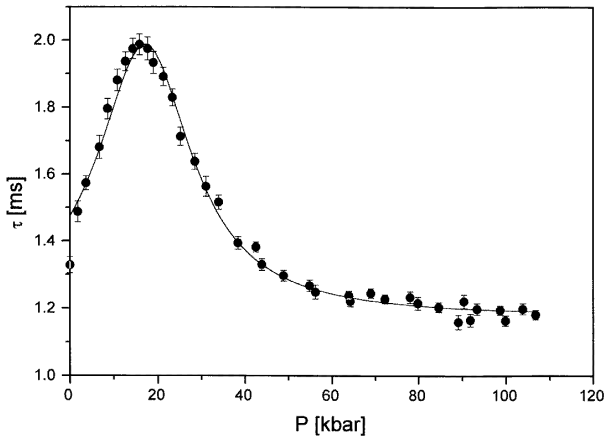

Figure 2. The pressure dependence of the fluorescence lifetime τ for the $^5D_0 \rightarrow {}^7F_2$ transition at room temperature. ●: experimental data; ——: the curve fitted according to equation (2).

Like in ruby, decay curves for the $^5D_0 \rightarrow {}^7F_2$ transition for any pressure are exponential. On increasing the pressure, starting from 1 kbar, the lifetime τ for the $^5D_0 \rightarrow {}^7F_2$ transition suddenly rises, reaching its maximum at about 17 kbar. On further increasing the pressure, the lifetime τ, after reaching its maximum, suddenly decreases, and at about 40 kbar reaches a value which stays unchanged until the maximum pressure is reached. The dependence of the lifetime τ for the $^5D_0 \rightarrow {}^7F_2$ transition upon pressure is neither linear, as in ruby [5], nor exponential, as in alexandrite [12] and YAG:Cr^{3+} [13]. For the experimentally achieved values of τ, at different pressures, this can be written as an exponential decay:

$$\tau \text{ (ms)} = 1.175 + 0.818[1 + 0.006(P - 16.796)^2]^{-1}. \tag{2}$$

The pressure P in equation (2) is in kbar. A graphical representation of the experimental points and fitting curve is given in figure 2. Full squares show experimental data with standard deviations. The parameter for the goodness of fit is $r^2 = 0.9877$.

4. Conclusions

According to our experiment the following conclusions can be stated.

(1) In the 0–104 kbar pressure range a linear increase of the position for the $^5D_0 \rightarrow {}^7F_2$ line with increasing pressure has been found.

(2) The relationship between the pressure and line position is given by linear regression: λ (nm) $= 611.29 + 0.011P$ (kbar).

(3) In the same pressure range the change of the fluorescence lifetime τ with pressure can be expressed as an exponential decay: τ (ms) $= 1.175 + 0.818[1 + 0.006(P - 16.796)^2]^{-1}$.

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