VUV studies of charge trapping in BaF_2:Ce

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Although the hipped interest in undoped BaF_2, the fastest known inorganic scintillator material disappeared soon after the SSC project was discarded, the RE (rare earth)-doped material still receives a fair amount of attention. In particular BaF_2:Ce, a reasonably fast and rugged scintillator material, offers a decent stopping power, favorable refraction index and availability of large size crystals [1]. With the sole exception of less dense and much slower CaF_2:Eu, it is also the most efficient of all the RE-doped alkaline earth fluorides [2]. It comes therefore as no surprise that both undoped and RE-doped BaF_2 have been a focus of a major effort at HASYLAB [3]. In this note we report more details of the current studies on BaF_2:Ce which was initiated at SUPERLUMI station in 1998 [4]. A full account of this work will soon be published in a regular journal [5].

The VUV excitation spectra, luminescence spectra and time profiles of Ce and STE (self-trapped exciton) emissions have been presented earlier [4]. Of special interest is the peculiar disparity between time profiles obtained at RT (~300 K) for 60 and 75 nm excitations, shown in Fig. 1. Although under both excitations the emission spectra consist exclusively of the Ce^{3+} *d-f* bands only the 60 nm profile shows a ~30 ns Ce^{3+} radiative lifetime decay. The 75 nm profile displays a longer rise and decay times, as well as significantly higher background indicative of larger contribution of even slower components.

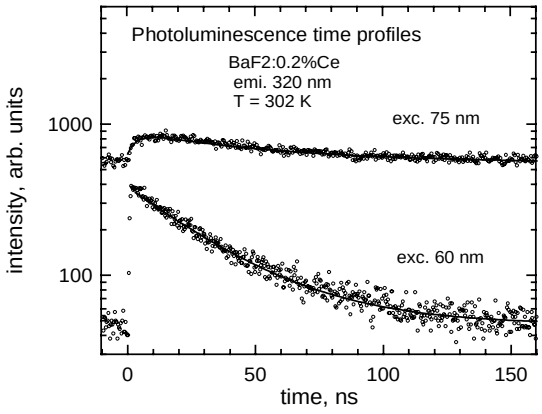


Figure 1: Emission time profiles of BaF_2:Ce under VUV excitations by synchrotron pulses at room temperature. Experimental points are shown by small empty circles. Solid lines depict one- (31.8 ns) and three-exponential (3.9, 33.1 and 64.2 ns) fits with an additional fitting constant to correct for background and longer decay time components.

The explanation, illustrated in Fig. 2, was offered by Wojtowicz *et al.* [4,5]. They note that the photon energy corresponding to the 60 nm wavelength is above, while that for the 75 nm is below the threshold for the Ba^{2+}5p core excitation. Consequently the 60 nm excitation creates a core hole and a cold electron in the conduction band that are not likely to separate. On the contrary, the 75 nm excitation produces a valence band hole and a hot conduction band electron that have an opportunity to separate and become trapped introducing slower, trap mediated components to radiative recombination of electrons and holes.

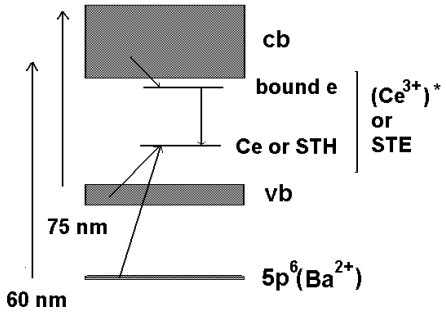


Figure 2: BaF_2:Ce energy diagram. The 60 nm excitation creates a $5p^5(Ba^{3+})$ core hole that self-trappes (creating self-trapped hole, STH) or is captured by Ce^{3+}. The slow conduction band electron is then immediately trapped at the site producing STE or $(Ce^{3+})^*$. On the contrary the 75 nm excitation creates a valence band hole and a hot electron that have a chance to separate.

To study the influence of charge trapping on radiative recombination of charge carriers in BaF_2 we have measured time profiles under 75 nm synchrotron excitation for various temperatures, shown in Fig. 3. In Fig. 4 we present decay and rise times (filled circles and filled triangles) obtained from fits to those profiles as a function of inverse temperature, 1/T. For comparison we have also included decay times (empty diamonds) of profiles excited at 300 nm (into the lowest energy absorption band of the Ce^{3+} ion).

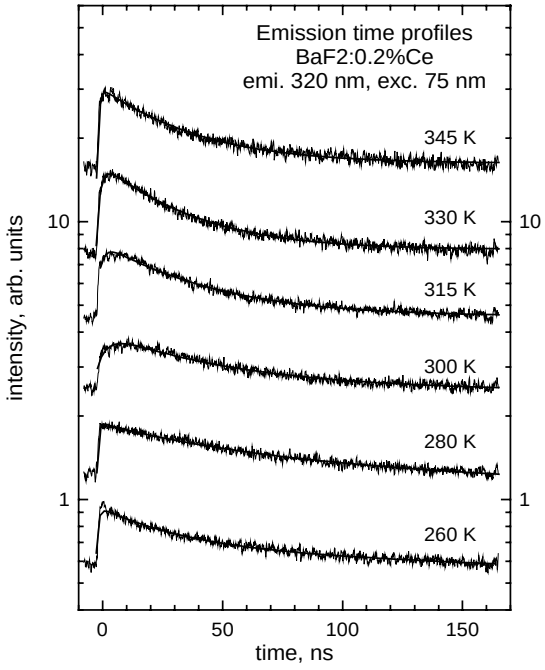


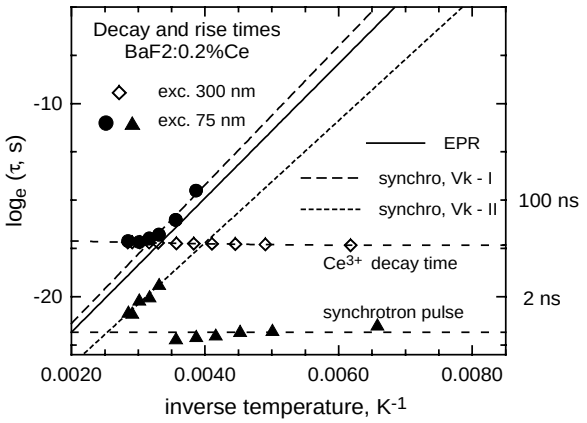
Figure 3: Emission time profiles of BaF_2:Ce under 75 nm excitation for various temperatures. The consecutive curves have been shifted vertically. The jagged thin solid lines represent experimental traces while the thick solid lines depict two-exponential fits with an additional fitting constant to correct for background and slower decay time components.

From a simple one-trap model we know that a trap can modify a rising or a decaying part of the time profile introducing a characteristic time τ (trap lifetime) [5],

$$\tau = (s \cdot \exp(-E/kT))^{-1},$$

where E is the trap depth, s the frequency factor, and k the Boltzmann constant. This formula can be used to extract trap parameters from the appropriate straight line fits as shown in Fig. 4. Since there are clearly two different straight lines that fit experimental points we conclude that there are two different traps responsible for longer decays (designated V_k-I, E = 0.31 eV, $s = 2.6 \cdot 10^{12} s^{-1}$) and longer rise times (designated V_k-II, E = 0.273 eV, $s = 9.5 \cdot 10^{12} s^{-1}$) at relevant temperatures.

Figure 4: Decay and rise times against the inverse temperature. Points in the diagram were obtained from fits to time profiles shown in Fig. 3. Long and short dashes lines depict straight line fits obtained for selected points. Horizontal dashed lines indicate rise and decay times determined by synchrotron pulse (or instrumental profile) and Ce^{3+} radiative lifetime. The solid line was calculated from trap parameters found from EPR measurements (see text).



The solid line in Fig. 4 was calculated from trap parameters characterizing the STH (V_k) random motion activation energy and frequency factor in BaF_2 (0.30 eV, $3.2 \cdot 10^{12} s^{-1}$) found by Beaumont et al. by monitoring the EPR signal from optically alined V_k centers [6]. Clearly this mode of the V_k motion (90° jump) is represented by one of the "traps" (V_k-I) that we found from time profiles of the VUV excited Ce^{3+} emission. The second "trap", V_k-II, must therefore correspond to the second mode, the so-called "linear" motion mode of the V_k center decay that could not be detected by EPR. We note also that the calculated trap lifetimes at 300K (61 and 4.4 ns) are close to decay and rise times found experimentally (64.2 and 3.9 ns).

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