Two modes of the V_k center decay in BaF_2:Ce

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BaF_2:Ce was found to be a reasonably fast and rugged scintillator that offers a decent stopping power, favorable refraction index and availability of large size crystals [1]. Although the scintillation mechanism in this material has been studied for many years it still remains open for discussion. Recently it has been proposed that this mechanism is based on consecutive capturing of charge carriers by the activator [2], which takes place either at once after the excitation or is postponed by self-trapping of holes. The process of self-trapping creates V_k centers (F_2^- molecules), that are stable at lower temperatures but become mobile when the temperature rises hence are able to decay at the recombination centers (Ce^{3+} + e-). The motion of the V_k centers in undoped alkaline earth fluorides was investigated by the EPR method by Beaumont el at. [3]. They observed that in general this motion can be either linear (0° jump) or random (90° jump) but in the case of BaF_2 they detected only the random one. In our investigation of recombination process in BaF_2:Ce we observe both mentioned modes of decay, labeled V_{kI} and V_{kII} [2]. The V_{kI} mode corresponds to the random motion detected by Beaumont while the mode V_{kII} to the linear motion.

In this note we present and discuss some new results of our continued and more detailed research concerning the role of the V_k centers in the kinetics of recombination process in BaF_2:Ce. Particularly we focus on time profiles measurements at temperatures between 100 and 320 K. The step between consecutive measurements was adjusted to emphasize the expected effects. The investigated samples were excited with the 75 nm light selected by the McPherson monochromator furnished with the platinum grating characterized by a very good reflectance coefficient for the short wavelength part of the spectrum (in comparison to the previously used aluminum one). The emission was detected at 320 nm (cerium emission). Our experiments have also been performed on the two newly grown samples (Optovac) with the concentrations of the activator at 0.01% and 0.05% (previously 0.2%). The smaller concentrations allowed us to decrease undesirable effects observed in these crystals at higher concentration of the activator.

As the 75 nm wavelength corresponds to the photon energy of 16.5 eV (Eg of BaF_2 ~ 10 eV) it leads to the creation of free charge carries analogously as in the case of the high-energy excitation (i.e. by gamma particles). Moreover, created electrons have relatively high energy and can easily escape from the attractive potential of holes and wander off to be captured by Ce^{3+} ions. In the meantime the holes quickly self-trap and become the V_k centers, which, as noted before, delays the recombination process and introduces temperature dependent V_k motion. The effects of this process can be seen in Fig. 1 where the time profiles are shown (points).

It is easy to see that the time profiles change with temperature. As the temperature rises the new and slow components are introduced and the rising parts of profiles are changed. Those components are due to the decay of the V_k centers recombining at [Ce^{3+}+e-] and their decay and rise time constants change with temperature according to the following formula:

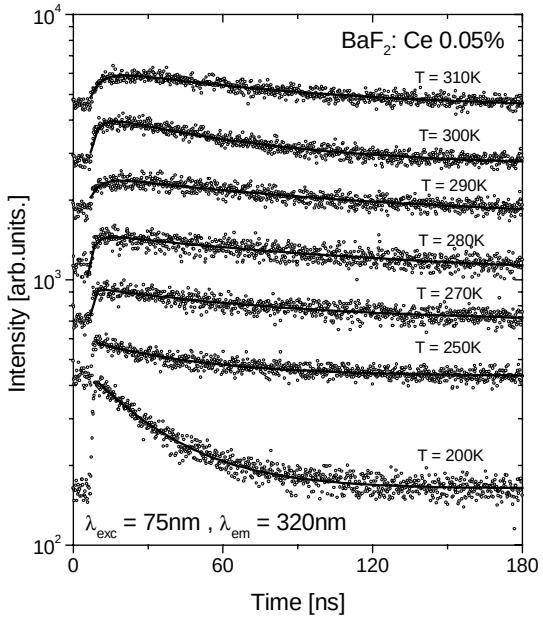
$$\tau = (s \exp(-E/kT))-1 \qquad (1)$$

where s is the frequency factor, E the activation energy, and k the Boltzmann constant.

These time constants can be extracted from the one-exponential fits to the experimental points (solid lines in Fig. 1). The values derived from fits to time profiles measured for both 0.05% and 0.01% samples are represented by squares and triangles, respectively, in the Arrhenius diagram shown in Fig. 2 where the logarithm of τ is plotted against the inverse temperature:

$$\ln(\tau) = E/kT - \ln(s) \qquad (2)$$

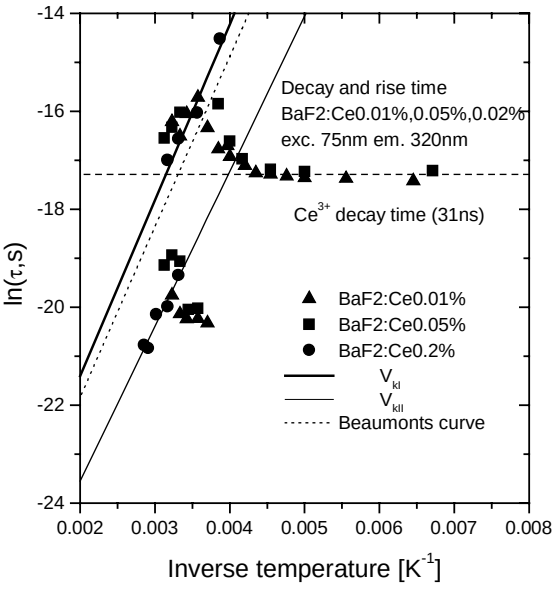
For comparison in the same diagram we also show the points extracted from the fits to the previously measured time profiles of BaF_2:0.2%Ce (circles). It is easy to see that new points extend our diagram and show the change in the decay time constants in the temperature range of 220 – 270 K.



In this diagram we also show four straight lines plotted according to formula (2) and indicating the expected changes in time constants of emission components associated with the V_k recombination. The parameters E and s used to calculate the solid lines were given by Wojtowicz et al. [4] and describe two modes of the decay of the V_k centers (V_{kI}: E = 0.31 eV, ln(s) = 28.59; V_{kII}: E = 0.273 eV, ln(s) = 29.88). The dotted line was calculated with parameters given by Beaumont el at. [3] (E = 0.3 eV, ln(s) = 28.79) that correspond to the random motion of the V_k centers. Note that the dotted and solid lines are reasonably close suggesting that the observed V_{kI} decay mode corresponds to the random motion. The horizontal dashed line indicates the radiative decay time of the excited Ce^{3+} ion (31 ns).

Figure 1. The emission time profile against temperature: $\bullet_{exc}$=75nm, $\bullet_{em}$=320nm

It is easy to see that the changes in time constants obtained from fits do follow the solid straight lines. First we observe the lengthening of decay time constant that coincides with the first solid thin line (V_{kII}). This indicates that as the temperature increases the lifetime of the V_{kII} centers approaches the radiative lifetime of the excited Ce^{3+} ion. Then the V_k lifetime shortens further and starts to modify the rise times (~260 K). At the same time the changes of decay time constants start to follow closely the solid thick line (V_{kI}) and the dotted line.



The observed behavior of time constants clearly shows that there are two components of emission generated by the recombination of the V_k and [Ce^{3+} + e-] centers and that these components can be associated with different modes of the decay of the V_k centers, linear and random.

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Figure 2. Arrhenius diagram (see text)

References

[1] C. Taylor, O.H. Nestor, and B. Utts, IEEE Trans.Nucl.Sci 33, 243 (1986)

[2] A.J. Wojtowicz, P. Szupryczynski, J. Glodo, W. Drozdowski, and D. Wisniewski, J.Phys. Condens.Matter 12**,** 4097 (2000)

[3] J.H. Beaumont, W. Hayes, D.L. Kirk, and G.P. Summers, Proc.Roy.Soc.Lond. A315, 69 (1970)

[4] A.J. Wojtowicz,W. Drozdowski, J. Glodo, and D. Wisniewski, Hasylab Annual Report 1999, pp. 351-352