VUV LUMINESCENCE OF BaF_2:Er AND (Ba,La)F_2:Er

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New applications such as fast scintillators, VUV solid state lasers and mercury free phosphors have generated a surge of interest in the VUV luminescence of rare earth activated wide bandgap materials. It is interesting to note that the so-called "extended Dieke's" diagram has been established only recently [1]. Lack of experimental and theoretical data on energies of the higher lying levels of the $4f^n$ and $4f^{n-1}d$ configurations and availability of excellent synchrotron facilities devoted to VUV (Superlumi and HIGITI stations, Hasylab, DESY, Hamburg) provide additional factors attracting attention of many researchers.

In this communication we present luminescence and luminescence excitation spectra of Er-activated BaF_2 and (Ba,La)F_2. Preliminary, low resolution results on BaF_2:Er have been published by Drozdowski *et al*. [2]. Since their interpretation did not take into account higher lying energy levels of the $4f^{11}$ configuration [2], we have planned and performed new experiments in August 2007. These experiments were designed to clarify the role of the $^2F_{5/2}$, $^2G_{7/2}$ and $^2G_{9/2}$ $4f^{11}$ states in the VUV and UV emissions following excitation into spin-allowed $4f^{10}5d$ levels of the Er^{3+} ion in BaF_2 and (Ba,La)F_2.

Surprisingly, the dominant VUV emissions in these two closely related materials, shown in Figs. 1 and 2, are different. The emission of BaF_2:Er, peaking at 163.5 nm, is slow, as reported earlier [2,3], and similar to VUV emissions from other Er-activated fluorides [4]. The emission of (Ba,La)F_2:Er, peaking at 162.5 nm is, unexpectedly, relatively fast (35 ns at room temperature and 45 ns at 10 K). Also, as showin in Figs. 1 and 2, the relative intensities of the longer wavelengths bands are different.

We explain these results by proposing that in BaF_2:Er the emitting level is, as expected, the lowest, high spin (HS) *d*-level, *(HS)*$4f^{10}5d(e)$. The transition to the lowest energy $4f^{11}$ level ($^4I_{15/2}$) is spin forbidden, hence slow. Fast relaxation from the higher energy low spin level *(LS)*$4f^{10}5d(e)$ is facilitated by an intermediate $4f^{11}$ state, $^2F_{5/2}$. In (Ba,La)F_2:Er *d*-levels are shifted and the corresponding, spin forbidden emission, at 170 nm, is much weaker. The likely reason is that the intermediate $^2F_{5/2}$ state levels in (Ba,La)F_2 overlap the LS *d*-band at 158.7 nm leaving a relatively large energy gap between this and the lowest energy HS level corresponding to the *d*-band at 165 nm. Since both of these bands correspond to the same electronic configuration $(4f^{10}5d(e)$, a change in configuration coordinate must be small and, consequently, the relaxation of the ion in the higher energy LS state to the lower energy HS spin state, must be relatively slow. Slow relaxation promotes build up of the population of the LS level, from which the spin-allowed transitions to the $^4I_{15/2}$ ground state originate, producing the dominant VUV band at 162.5 nm. The longer wavelengths VUV and UV bands, shown in Figs. 1 and 2, originate in spin-forbidden (BaF_2) and spin-allowed ((Ba,La)F_2) transitions terminating at the excited states of the $4f^{11}$ multiplet, ^4I_J. It is not clear why, in (Ba,La)F_2, the relative intensities of these bands are higher than in BaF_2.

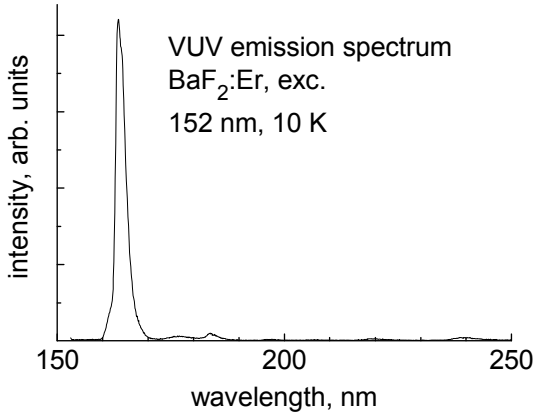

Figure 1. Time integrated VUV emission spectrum of BaF_2:Er. Excitation wavelength 152 nm (*d*-band), temperature 10 K.

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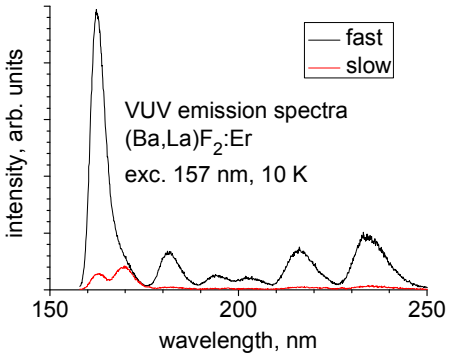

Figure 2. Time resolved emission spectra of (Ba,La)F_2:Er. Excitation wavelength 157 nm (*d*-band), temperature 10 K.

References

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