Photostimulated trap filling in Lu_2SiO_5:Ce^{3+}

LETTER TO THE EDITOR

Photostimulated trap filling in Lu_2SiO_5:Ce^{3+}

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Abstract
Upon exposing Lu_2SiO_5:Ce^{3+} scintillators to daylight, the material shows
undesired intense afterglow lasting for several hours at room temperature.
This effect is studied by means of illuminating Lu_2SiO_5:Ce^{3+} with visible
and ultraviolet light. Conditions for trap filling and the relation with
photoconductivity phenomena and Ce^{3+} luminescence quenching will be
discussed. Autoionization of so-called Ce2 centres following the excitation
to the lowest 5d level seems to play an important role in the mechanism.

Since the discovery of the scintillation properties of Lu_2SiO_5:Ce^{3+} (LSO:Ce) in 1990 [1]
the material has found its way to application as a 511 keV gamma-ray detecting element in
positron emission tomography. LSO:Ce shows afterglow after exposure to ionizing radiation
or to daylight. In a preceding study [2] the afterglow was related to a specific peak in
the thermoluminescence (TL) glow curve. A model was proposed in which Ce^{3+} was
simultaneously the trap-creating defect and the luminescence centre. Furthermore, a centre-
to-centre recombination mechanism, i.e. not involving charge transport via the conduction
band, was suggested. Recent TL studies by Cooke and co-workers [3–5] have provided more
insight into the nature of the trapping centres. They demonstrate that the trap is intrinsic to
the oxy-ortho-silicate host crystal and that Ce^{3+} is not the trap-creating defect. The same traps
seem present [6] when rare-earth dopants other than Ce^{3+} or even other oxy-ortho-silicate host
lattices like Y_2SiO_5 and Gd_2SiO_5 are involved.

The current interest in the afterglow phenomenon in the oxy-ortho-silicates [3–5, 7] was
the motivation for analysing some hitherto unpublished data on the mechanism of trap filling
and afterglow in LSO:Ce^{3+} during illumination with ultraviolet and visible light. Despite the
fact that many questions will remain, the data presented will provide further clues as regards
the elucidation of the afterglow mechanism in LSO:Ce scintillators.

The sample used (sample LSO7-11) is the same as the one used in the experiments
described in [2]. The sample was illuminated by means of deuterium lamp light transmitted
subsequently through a quartz lens, a cut-off filter, and an interference filter. The lens ensures
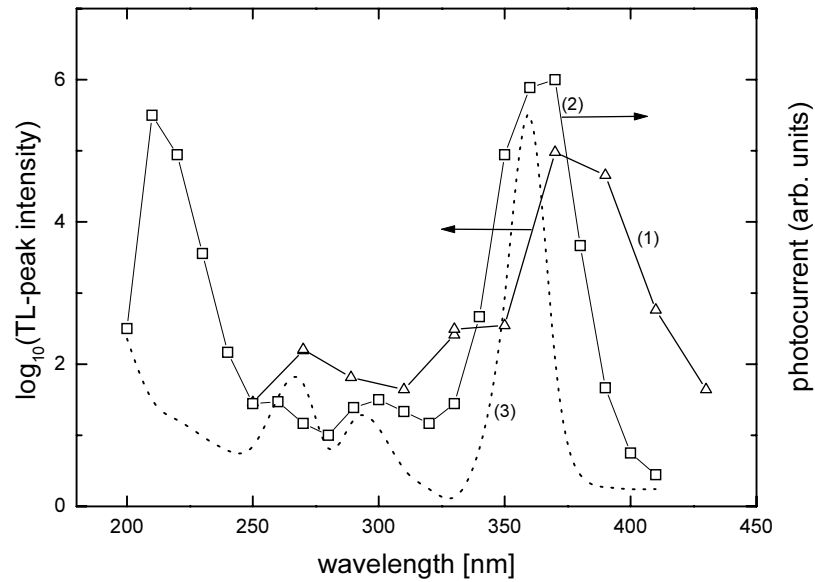
Figure 1. Curve (1): TL peak intensity (this work) against the wavelength of the trap-filling stimulating illumination light; curve (2): photoconductivity from [8]; and curve (3): optical absorption ($\log_{10}$(transmission)) from [2], for Lu_2SiO_5:Ce^{3+}.

that a parallel beam of light is incident on the interference filter. The cut-off filter prevents second-order transmission. The holder containing the sample could be immersed in liquid nitrogen. Prior to illumination, the sample was heated to 400 °C in order to empty all the traps. After illumination, the TL was recorded. This was repeated with a set of filters (FWHM = 10 nm) every 20 nm starting from 250 up to 430 nm. The experiment was done with the sample at room temperature and repeated with the sample at liquid nitrogen temperature. The transmission of the filter combination was measured separately. This, together with the known spectral profile of the lamp emission, provided the relative number of photons incident on the sample during the illumination period.

Figure 1 shows the integral intensity of the TL peak responsible for afterglow (see [2]) on a $\log_{10}$ scale as a function of the wavelength of the interference filter used. At wavelengths longer than 430 nm, the sample is totally insensitive to light and traps are not filled at all. At wavelengths shorter than 410 nm the trap filling occurs and increases rapidly with decreasing wavelength until it peaks at 370 nm. Going to 350 nm, it rapidly drops by more than two orders of magnitude. From 350 to 250 nm, trap filling does occur but it is, compared to that at 370 nm, rather inefficient and barely above the noise level. In contrast with these results, the same experiment repeated with the sample cooled to liquid nitrogen temperature did not yield any significant trap filling.

These results have a very close resemblance with those from photoconductivity experiments by Yen *et al* [8]. Figure 1 shows the photoconductivity on a linear scale as a function of wavelength at room temperature. Photoconductivity is maximal around 365 nm and for shorter wavelengths it drops markedly until it peaks again at 210 nm. Like for the trap-filling efficiency curve, photoconductivity is fully absent at liquid nitrogen temperature. According to Yen *et al* [8, 9], on excitation with 365 nm light, Ce^{3+} is excited to its lowest 5d level which is located just below the conduction band. At room temperature, thermally activated ionization takes place, leading to persisting photoconductivity. At liquid nitrogen temperature, thermal activation does not occur.

It is known that two different types of Ce centre, denoted as Ce1 and Ce2 by Suzuki *et al* [10], are present in Lu_2SiO_5. The optical absorption band around 355 nm—see figure 1—is mainly caused by the electronic transition to the lowest-energy 5d level in the Ce1 centres. The less abundant Ce2 centres yield much weaker absorption at 5–10 nm longer wavelengths [4,10]. Inspecting figure 1 reveals that both the photoconductivity curve and the trap-filling efficiency curve peak at wavelengths slightly longer than that for the fd absorption in the Ce1 centres. This suggests that the Ce2 centres play an essential role in photoconductivity and trap filling. Further evidence supporting this suggestion is provided by the thermal quenching behaviour of the Ce2 centres. The luminescence of Ce2 centres is very efficient at liquid nitrogen temperature [10] and apparently, instead of undergoing ionization to the conduction band, the 5d excited state returns radiatively to the 4f ground state. At room temperature the situation is reversed. Thermally activated ionization to the conduction band causes an efficient luminescence quenching of the df luminescence together with the simultaneous appearance of photoconductivity and trap filling. Since the luminescence of Ce1 centres is efficient at room temperature, the energy barrier for autoionization of the 5d state of Ce1 centres seems to be larger than that for Ce2 centres.

Summarizing, results from photoconductivity experiments, TL and afterglow, and luminescence quenching studies have been consistently explained by a model involving the autoionization of the 5d state of excited Ce2 centres. What remains puzzling is the fact that the TL recombination centre is still the Ce1 centre—as this is the only luminescing centre available at temperature above 300 K.

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