

# Ce<sup>3+</sup> and host emissions in VUV excited Lu<sub>2</sub>SiO<sub>5</sub>:Ce

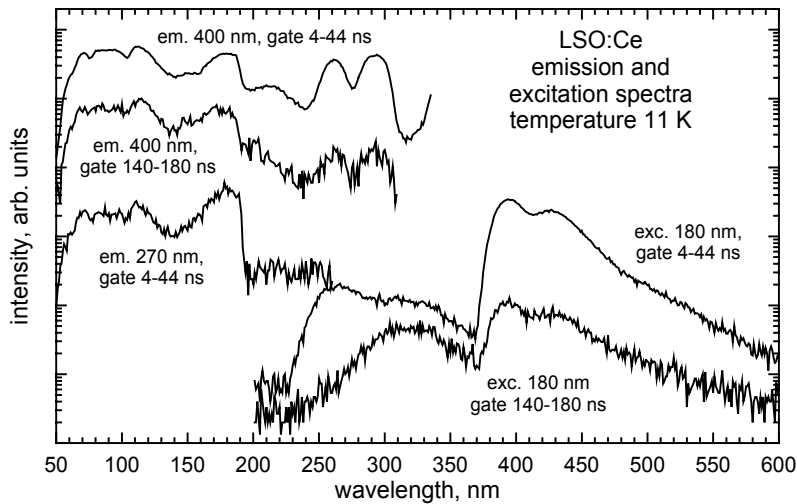
A.J. Wojtowicz<sup>1</sup>, W. Drozdowski<sup>1</sup>, P. Szupryczyński<sup>2</sup>, D. Wiśniewski<sup>1</sup>, and C.L. Melcher<sup>2</sup>

<sup>1</sup>Instytut Fizyki, Uniwersytet M. Kopernika, Grudziądzka 5, 87-100 Toruń, Poland

<sup>2</sup>CTI Molecular Imaging Inc., 810 Innovation Dr., Knoxville, TN 37932

Cerium activated lutetium oxyorthosilicate (Lu<sub>2</sub>SiO<sub>5</sub>:Ce, LSO:Ce) is a well known commercial scintillator material discovered more than a decade ago [1,2]. The material has been studied extensively and the dominant mechanism of scintillation light production in LSO:Ce was established to be due to a very efficient radiative recombination of charge carriers via two different Ce<sup>3+</sup> related centers, designated Ce1 and Ce2 [3-5]. The selective excitation of these two centers in LSO and other mixed silicates is possible by using a carefully wavelength tuned excitation (see W. Drozdowski et al [6]).

Almost all of the previous spectroscopic studies of LSO involved no VUV excitation wavelengths which are not readily available. Nevertheless there are processes involving excitons, traps, and uncontrolled defects producing effects such as slow components, room temperature afterglow and so-called "host" emissions that can be studied by using VUV. In this report we present results of VUV studies of the pixel 2x2x10 LSO sample grown by CTI MI Inc., performed at Superlumi station of Hasylab at DESY and aiming at revealing and characterizing the various host-to-ion (indirect) energy transfer channels that are active in LSO. In particular it would be interesting to identify and separate those indirect channels that selectively feed the two LSO centers, Ce1 and Ce2.



*Figure 1: Uncorrected emission (resolution 4.32 nm) and excitation (resolution 0.25 nm) spectra of LSO. The spectra were shifted vertically to aid presentation. The emission spectra were measured with the excitation wavelength set at 180 nm. The excitation spectra were measured with the emission wavelength set at 400 nm (maximum of Ce-emission) and at 270 nm (one of the two intrinsic "host" emissions). For details see text. Temperature was 11 K*

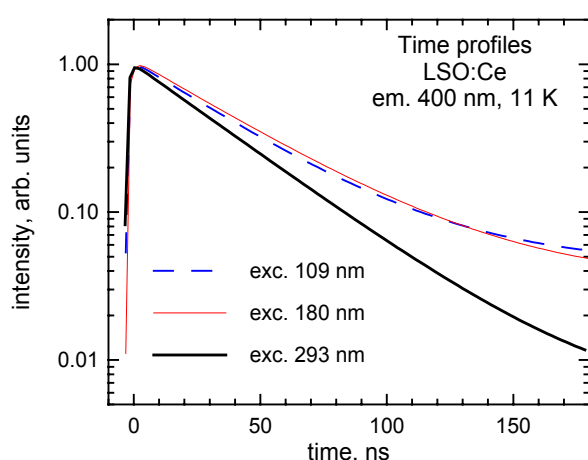
In Fig. 1 we present time resolved "gated" excitation spectra of the well known Ce<sup>3+</sup> emission (Ce1) and of the "host" emissions characteristic of the undoped LSO crystal. The excitation spectra of the Ce2 emission are presented in [6]. The emission spectra at 180 nm excitation are also shown in the same figure. The spectra were measured with the signal accumulated within a 40 ns time window triggered by synchrotron pulses (0.5 ns time width, 192 ns repetition time) with an appropriate delay (4 and 140 ns) to obtain "prompt" and "delayed" spectra, respectively. We note that the emission spectra show a characteristic structure between 370 and 480 nm corresponding to the well known spin-orbit split *d-f* transition at Ce<sup>3+</sup> (Ce1 center) and a complex band between 230 and 400 nm reflecting intrinsic so-called "host" emissions. The contribution of the Ce2 center to the emission spectra shown in Fig. 1, at the longer wavelength side of the Ce1 band, is relatively small despite the low temperature that reduces quenching of the Ce2 emission characteristic of room temperature. The "prompt" excitation spectrum (4-44 ns gate) shows the well known [3-5] *d-f* bands split by the crystal field (between 250 and 350 nm). Interestingly these bands are much weaker in the "delayed" spectrum indicating that all the processes of energy transfer, triggered by excitation below 200 nm and represented by structured bands between 50 and 140 nm and a "bandgap" peak at about 190 nm, are relatively slow.

The structure in the band below 140 nm (nearly the same in the two spectra) is due to spectral characteristics of the Al grating of the primary monochromator and does not reflect any real physical processes. The high

signal at these wavelengths is, nevertheless, indicative of strong sensitivity to VUV excitation and, since the VUV photons at these wavelengths provide over the bandgap excitation of the host material, the Ce ions must be efficient radiative recombination centers providing both fast and slow components in the scintillation time profiles. This notion is further supported by the emission spectra shown in Fig. 1 showing that in the Ce-activated sample under VUV excitation the emission is always dominated by Ce ions even for the 180 nm transition which only indirectly excites Ce ions.

The emission spectra shown in Fig. 1 under excitation at 180 nm show two additional bands at 270 and 330 nm that are characteristic of the undoped host material. These bands are also present in both “prompt” and “delayed” emission spectra under VUV excitation indicating that Ce-ions do encounter some competition from “intrinsic” emission centers in LSO.

The excitation spectrum of the 270 nm emission band, shown in Fig. 1, does not show any of the  $\text{Ce}^{3+}$   $f$ - $d$  bands. A characteristic asymmetrically shaped band steeply rising at the bandgap energy and peaking at about 190 nm, points to a large Stokes shift with no overlap between emission and absorption bands; features that are indicative of excitonic origin.



*Figure 2: Emission time profiles of LSO:Ce under direct  $f$ - $d$  (293 nm) and indirect (109 and 180 nm) excitations. The emission monochromator was set to 400 nm, temperature was 11 K.*

We have also measured a number of time profiles of Ce-emission under VUV and  $f$ - $d$  excitations at various temperatures. In Fig. 2 we show three representative profiles. The dashed and thin solid line profiles were obtained under VUV (109 nm) and excitonic (180 nm) excitations while the thick solid line shows the profile obtained under  $f$ - $d$  (293 nm). The  $f$ - $d$  excited profile shows no rise time and a decay time constant of 35.4 ns (within experimental error equal to the radiative lifetime of the excited  $\text{Ce}^{3+}$ , Ce1 center) while the indirectly excited profiles show a slightly longer decay time constants (39.3

and 40 ns) and significantly higher background that are indicative of slower indirect energy transfer processes.

In this report we have presented evidence that indirect VUV excitation in LSO provides a slower channel of energy that serves, almost exclusively, Ce1 centers. We have identified two competing intrinsic emission centers that are efficiently excited by wavelengths falling in the range of the “host” bandgap peak and are fed, therefore, by some exciton-related mechanism. We note that except for direct excitation (see [6]) we identify no indirect energy transfer channel that would efficiently feed Ce2 centers.

This work was supported by the IHP-Contract HPRI-CT-1999-00040/2001-00140 of the European Commission. The support and hospitality of Prof. G. Zimmerer, Dr M. Kirm and Dr S. Vielhauer of Hasylab is also gratefully acknowledged.

## References

- [1] C.L. Melcher, J.S. Schweitzer, Nucl. Instr. Meth. A 314, 212 (1992).
- [2] C.L. Melcher, J.S. Schweitzer, IEEE Trans. Nucl. Sci. NS-39, 502 (1992).
- [3] H. Suzuki, T.A. Tombrello, C.L. Melcher, J.S. Schweitzer, Nucl. Instr. Meth. A 320, 263 (1992).
- [4] H. Suzuki, T.A. Tombrello, C.L. Melcher, J.S. Schweitzer, IEEE Trans. Nucl. Sci. NS-40, 380 (1993).
- [5] D.W. Cooke, B.L. Bennett, K.J. McClellan, J.M. Roger, and M.T. Whittaker, Phys. Rev. B 18, 11973 (2000).
- [6] W. Drozdowski, A.J. Wojtowicz, D. Wisniewski, S. Janus, C.L. Melcher and P. Szupryczynski, this volume