On the Photoluminescence Linearity of Eu^{2+} based LED Phosphors upon High Excitation Density

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Background

The current standard architecture for SSL is the phosphor-converted light-emitting diode (pc-LED) in which high brightness LEDs based on (In,Ga)N are combined with one or more down-converting phosphors to produce composite white light of nearly any color temperature and color rendering quality.
Nonetheless their tremendous success in pc-LED design, blue LEDs have one well-known and central drawback: a non-thermal drop in efficacy with increasing power density. This "efficacy droop" restricts operation to reasonably low input power densities, disagreeing to the wish to extract more photons per unit area of the LED chip and thereby make SSL more affordable.
A solution to overcome the efficacy droop of blue LEDs, could be the use of LDs. Operated in stimulated emission, in principle high efficacies at much higher input densities than for LEDs can be reached. Certainly, at high input power densities state-of-the-art, high power blue, edge emitting LDs have already reasonably high (30-40%) power-conversion efficiencies, and a steady increase within the next years in efficiency can be expected.
A requirement of LDs for SSL is the ability to create white light. In principle, the same phosphors used in the pc-LED architecture can also be used with LDs. Indeed there are various reports dealing with phosphor-converted LDs (pc-LDs). Especially, Eu^{2+} activated phosphors are commonly used.
These phosphors, excited by high radiance pump sources, like high power LDs, offer considerable potential for high radiance conversion in SSL. Remarkably, theoretical arguments suggest that the radiance of the luminescent spot should increase linearly with the excitation density of the incoming light source up to 1 kW/mm^2. In practice, however, thermal quenching and (non-thermal) optical saturation limit the maximum attainable radiance of the luminescent material. In this work we present experimental data of the widely applied LED phosphors $(Sr,Ba)_2SiO_4:Eu^{2+}$ (OSE), $(Sr,Ca)_2Si_5N_8:Eu^{2+}$ (258:Eu), and $CaAlSiN_3:Eu^{2+}$ (CASN:Eu), in which these limits have been investigated. These systems are particularly good for high radiance conversion by virtue of their short luminescence lifetimes and little thermal quenching.

Experimental set-up

1. Pulsed laser diodes 465 nm (1000 mW) , 445 nm (80 mW) 405 nm (200 mW) or 375 nm (50 mW)
2. Focusing lens with a spot diameter of ~ 11 µm (1/e)
3. Infrared camera (7.5 … 14 µm)
4. Phosphor powder sample (layer thickness ~ 100 µm)
5. Passive cooling (Ag sample holder) or active cooling (He-Cryostate)
6. Emission monochromator
7. Detector (PMT red sensitive)

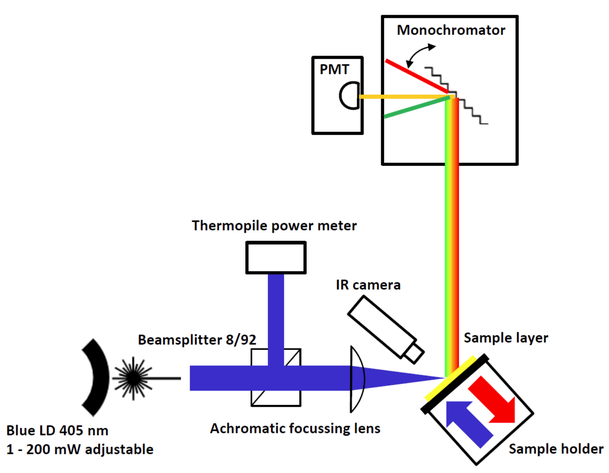


Fig. 1 Schematic of intensity saturation experiments

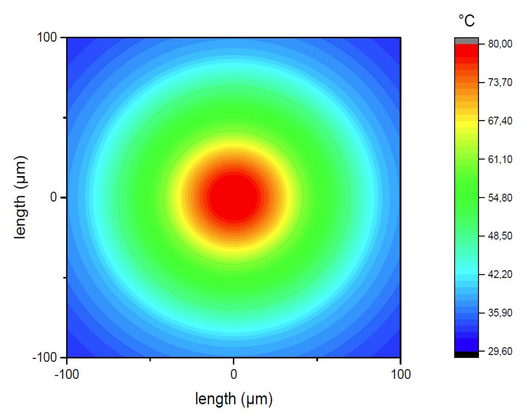


Fig. 2 Thermal distribution under 800 W/mm^2 excitation density to be exemplary for $(Ca,Ba)SiO_4:Eu^{2+}$

Results and Discussion

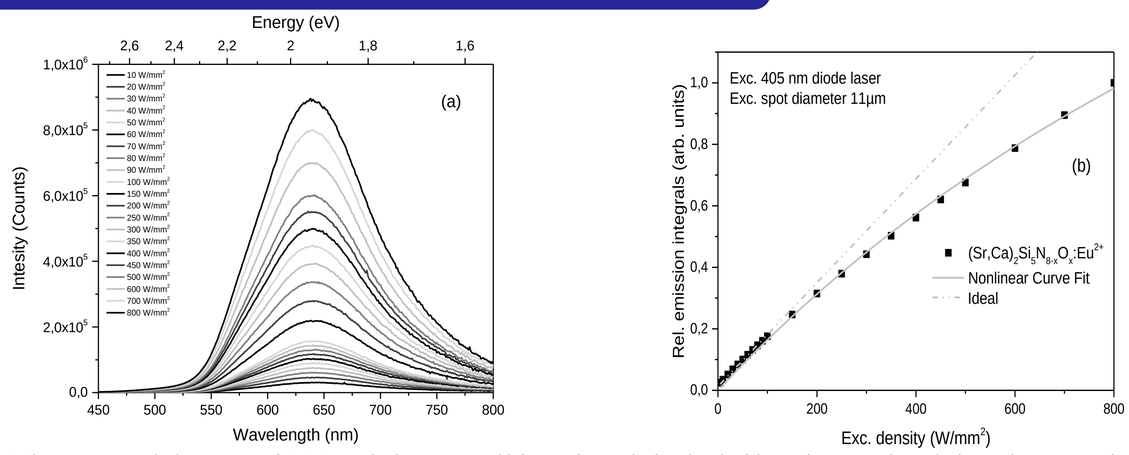


Fig. 3 Fluorescence emission spectra of 258:Eu excited at 445 nm with increasing excitation density (a). Luminescence intensity integrals versus excitation density (b).

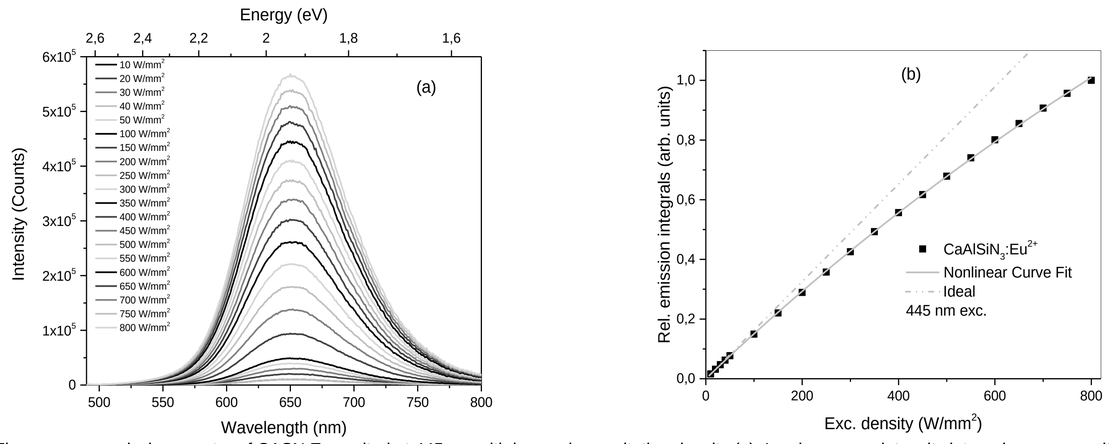


Fig. 4 Fluorescence emission spectra of CASN:Eu excited at 445 nm with increasing excitation density (a). Luminescence intensity integrals versus excitation density (b).

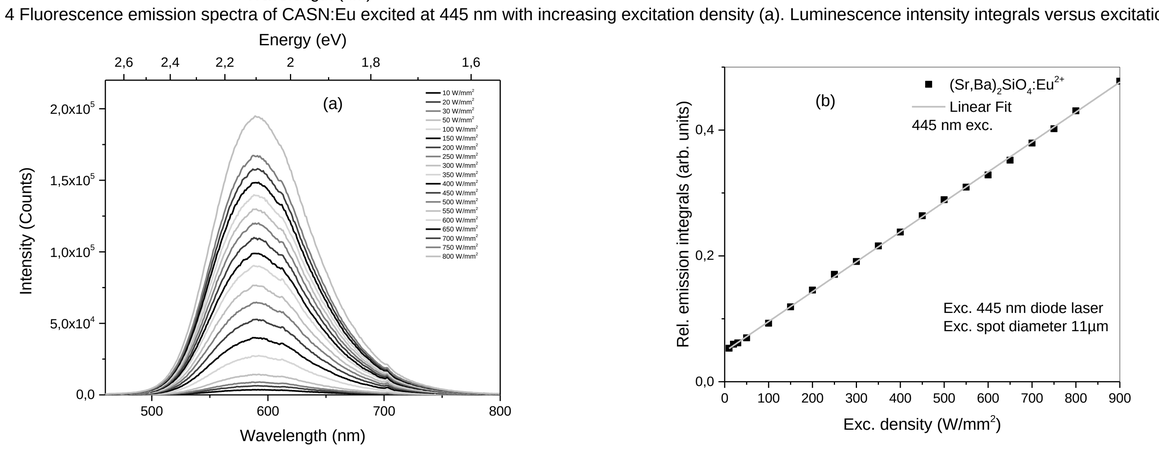
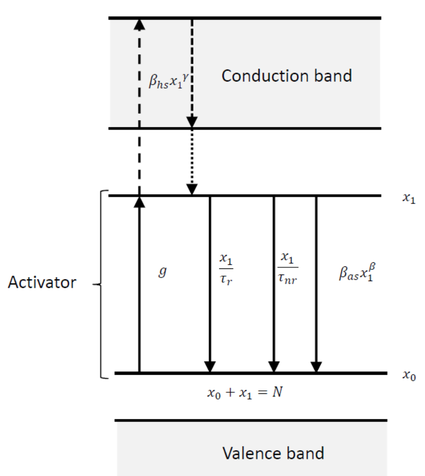


Fig. 5 Fluorescence emission spectra of OSE excited at 445 nm with increasing excitation density (a). Luminescence intensity integrals versus excitation density (b).

- Emission spectra as function of excitation density were recorded for λ_{exc} = 405 nm, which corresponds to the allowed transition of Eu^{2+} from the energy level $^8S_{7/2}$ of the [Xe]$4f^1$ configuration to the level $^7F_0$ of the [Xe]$4f^6 5d^1$
- In case of 258:Eu and CASN:Eu, optical pumping intensities have been shown to produce a significant efficiency depreciation already at low pump powers, which does not correlate with the relatively short Eu^{2+} luminescence lifetime (~ 1 µs)
- We assume a photoionization mechanism from excited state [Xe]$4f^6 5d^1$ into the conduction band of the host structure, which bottlenecks the limit maximum pump intensities
- We find that the investigated Eu^{2+} phosphors tend to show different efficiency depreciation
- This could be explained by the relative position of the excited state of Eu^{2+} to the bottom of the conduction band in the corresponding host material
- Finally, we use a rate equation model to simulate this nonlinear optical pumping caused by photoionization



Rate equations:

$$\frac{dx_1}{dt} = g - \frac{x_1}{\tau_r} - \frac{x_1}{\tau_{nr}} - \beta_{as} x_1^{\beta}$$

$$\frac{dx_1}{dt} = g - \frac{x_1}{\tau_r} - \frac{x_1}{\tau_{nr}} - \beta_{hs} x_1^{\gamma}$$

Fig. 6. Schematic diagram of the used model to describe the energy flow in a typical phosphor for fluorescent light sources.

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