Supplementary Material for the paper:

Silicon oxynitride nanofilms prepared by PLD with controlled Eu-local concentration

for broadband white light emitters

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S1. Excitation of the Eu-doped SiAION films: Comparison excitation at 355 nm vs 405 excitation

Photoluminescence excitation spectra using a 405 nm laser (EKSMA EO-50%-NL50). The films have been excited in a similar configuration than with the 355 nm. The photoluminescence signal has been collected at normal incidence using the same set up as described in the manuscript. For collection of the PL emission a 458 nm filter has been located in front of the monochromator. Figure 1 shows the resulting spectra both for the excitation.



Figure S1. Photoluminescence (PL) emission spectra corresponding to the SiAlON: Eu^{2+} films excited (a) at 355 nm and at (b) 405 nm. The inset in (b) shows the increase of the Integrated PL Intensity spectra of the samples at the highest emission, which was after annealing at 700 °C. The arrow shows spectral position of the the 458 nm filter used to block the tail of the 405 nm laser excitation

In Figure S2 we have plotted the comparison of the resulting normalized spectra for excitation at 355 nm and 405 nm for each Eu doping concentration. It can be seen that upon change in the excitation wavelength the shape and position of the maximum emission wavelength shows a negligible change. That the PL maxima do not change significantly is most likely due to the broad excitation band of the excited degenerated

Eu²⁺ :⁴f₆ ⁵d₁ levels combined with the broad range of different sites for the Eu²⁺ ions in our amorphous matrix. The only noticeable change is a slight decrease of the PL intensity for the 1.4 at. % Eu doped film in the low wavelength region (i.e. high energy region). This might be due to the differences in the PLE spectra compared to the other two films. The PLE spectra for 1.4 at. % Eu shows a double absorption band with two maxima at energies >300 nm and at 355 nm. Note that upon excitation at 405 nm we have significantly reduced the energy of the excitation, from 3.49 (355 nm) eV to 3.06 eV (405 nm) therefore we cannot any more excite the higher levels of the broad absorption band corresponding to the energy Eu²⁺ :⁴f₆ ⁵d₁ levels. With the excitation at 3.49 eV the high energy band can be excited partially, and therefore we observe the emission at lower wavelengths: whereas with the 3.06 eV mainly the low energy band is excited, and therefore the emission towards the lower wavelengths is favored.

Finally, the integrated luminescence as a function of the Eu^{2+} ions content when excited at 405 nm (Fig. S1. Inset) shows a similar behavior to that shown for the excitation at 355 nm (Fig. 4 inset in the manuscript). The integrated intensity shows a linear trend with the Eu content, therefore the PL intensity is mainly dominated by the increase in the number of Eu^{2+} emitters as the concentration in the films increases.

Altogether these results suggest that the broad distribution of Eu in different environments is quite homogeneous, and thus that excitation of these films in the technological relevant wavelength range of 380-420 nm is effective.



Figure S2. Comparison of the normalized photoluminescence (PL) emission spectra shown in Fig. S1 and corresponding to the SiAlON:Eu²⁺ films excited at 355 nm (continuous line) and at 405 nm (dotted line) for the different Eu concentrations (a) 1.4 at% Eu, (b) 2.8 at% Eu and (c) 5.6 at% Eu. The green line represents the 458 nm filter used to block the tail of the 405 nm laser excitation. Note this filter only slightly cuts the emission of the 1.4 at% sample.