

## **Er<sup>3+</sup> excited state absorption and the low fraction of nanocluster-excitabile Er<sup>3+</sup> in SiO<sub>x</sub>**

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Silicon nanoclusters (Si-nc) can be efficient sensitizers of surrounding Er<sup>3+</sup> ions, making Er-doped silicon-rich silica material very appealing for 1.5 $\mu$ m amplifiers and lasers pumped by broadband sources. Recent reports of optical signal enhancement in waveguides based in this system have increased the interest even more. However, there is currently a debate on how many surrounding Er ions can one single nanocrystal excite. In fact, there is considerable experimental evidence that the fraction of Er ions excited by Si-nc is actually very low, of the order of few percent. We point out that excited state absorption (ESA) is a major cause for this low fraction of indirectly excitable Er.

We base our argument on the fact that in most reports of Si to Er transfer, the emission of the Si-nc was centered at 800nm, and recent works show spectral hole-burning in the Si-nc emission band, attributed to resonant transfer to the <sup>4</sup>I<sub>9/2</sub> Er level. However, the metastable level of erbium <sup>4</sup>I<sub>13/2</sub> can also absorb a 800nm photon resonantly to the higher levels (<sup>4</sup>S<sub>3/2</sub>, <sup>2</sup>H<sub>11/2</sub>). We have estimated that this process can be almost as efficient as the transfer to an unexcited Er.

When there is only one Er per nanocluster, ESA processes would be an energy drain that increases the pump threshold value of an amplifier or laser. However, when there is a large population of Er ions surrounding each Si-nc, ESA can prevent the excitation of most of the ions. This is because the transfer time to each Er ion depends on the distance, and when there are many ions at different distances, the nearest ion is likely to receive a very fast transfer. But ESA processes to that ion will be very fast too, making the transfer to further ions very unlikely. We have modelled different populations of Er ions and Si-nc according to this picture and our results agree with reported curves of photoluminescence versus pump power.

Another evidence of this effect is related with the nanocluster luminescence behaviour versus pump power. If ESA processes were not occurring, the transfer time to Er would decrease as long as Er population becomes excited. This would increase the lifetime of the Si-nc emission, leading to a superlinear behaviour with power. Since this behaviour has never been observed, this means that the transfer time to Er is the same regardless of the excitation state of the Er population, which is a consequence of ESA.

We conclude that ESA processes must be considered for future material designs that require Er population inversion, and we indicate possible approaches that can potentially avoid this mechanism.