Quantitative assessment of energy transfer in upconverting nanoparticles grafted with organic dyes

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Lanthanide upconverting nanoparticles (UCNPs) are luminophores that have been investigated for a multitude of biological applications, notably cell imaging, high-sensitivity assays, and cancer theranostics. UCNPs exhibit the phenomenon of upconversion: sequential absorption of several low-energy photons followed by emission of a single high-energy photon.[1]

The principal advantage of using UCNPs as a luminophore for biological assays is prevention of sample autofluorescence due to IR excitation, drastically improving signal-to-noise ratio. In these applications, UCNPs are frequently used as a donor in resonance energy transfer (RET) pairs, due to extreme photostability, steady non-blinking emission, and several narrow bands of luminescence.[2,3]

However, because of the peculiarity and non-linearity of their luminescence mechanism, UCNP behavior as a RET pair component has been difficult to predict quantitatively, preventing their optimization for subsequent applications. To shed light on this question, we prepared water-dispersed polymer-coated UCNPs and investigated their luminescence decays and spectra, with varying UCNP size and quantity of acceptor dyes grafted on their surface. We observed an increase in RET efficiency with lower particle size and higher dye decoration (Fig. 1). We also observed several unexpected effects, notably a quenching of UCNP luminescence bands that are not resonant with the organic

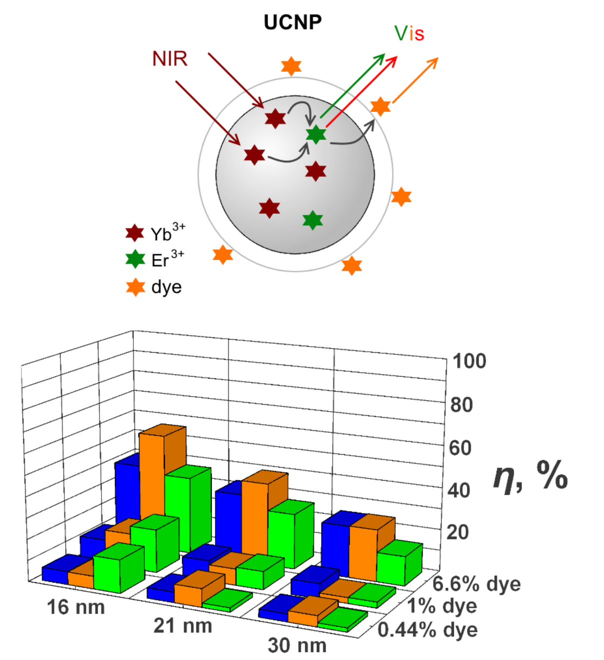

Figure 1. Top: scheme of the investigated system. Bottom: RET efficiency values, as calculated from luminescence decay lifetimes (green), spectra (orange), and Monte Carlo simulation (blue).

dye's absorption. We proposed a semi-empirical Monte Carlo model for predicting the behavior of UCNP-organic dye systems, and validated it by comparison with our experimental data.

These findings will be useful for development of more accurate UCNP-based assays, sensors, and imaging agents, as well as for optimization of UCNP-RET systems employed in cancer treatment and theranostics.

Funding: Labex NIE and COST CM 1403 «The European upconversion network».
O. Dukhno was supported by an Eiffel fellowship (Campus France) and a fellowship from the French Ministère de la Recherche.

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