

Multi-photon luminescence of single gold nanoparticles: Exploring the dynamics of plasmons and electron hole pairs

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Abstract. We investigate the dynamics of the multi-photon excitation of single, isolated gold nanoparticles and show that the one/two/three emission can be actively switched by changing the excitation pulse duration.

Luminescence of bulk gold has first been reported in the pioneering experiments of Mooradian in 1969 [1] and was attributed to the radiative recombination of electron hole pairs. In the recent years it was pointed out that their optical properties, like elastic scattering [2] or one photon luminescence [3], are dictated by the particle plasmon.

Here we utilize multi-photon excitation to investigate the optical properties of single gold nanoparticles and report on efficient two and three photon luminescence emission based on the excitation of electron hole pairs. Furthermore, we show that the emission of the same gold nanoparticle strongly depends on the excitation conditions and can be actively switched by changing the excitation pulse duration from 500 fs (Fig.1(a)) to 100 fs (Fig.1(b)).

Fig. 1 (a) shows the emission spectrum of a single gold nanosphere excited with one (black line) and two (red line) photons exhibiting a single emission band at 550nm. This emission follows closely the plasmonic properties of the gold nanosphere. In Fig. 1(b) the gold nanosphere is excited with one (black line) and two (red line) photons and the green line displays a corresponding simulation. In this case a shorter excitation pulse duration was used, which drastically changes the emission properties. This emission can be described by the radiative recombination of electron hole pairs. Additionally, we are using single gold nanorods with different aspect ratios to investigate the interplay between the particle plasmon and electron hole pairs, which enables us to develop a quantitative model to fully describe the two and three photon luminescence emission of single gold nanoparticles.

These findings greatly increase the understanding of the physical processes underlying the luminescence of gold and hence help to increase the usefulness of gold nanoparticles in various application fields, e.g. in material science, bio imaging, microscopy and spectroscopy.

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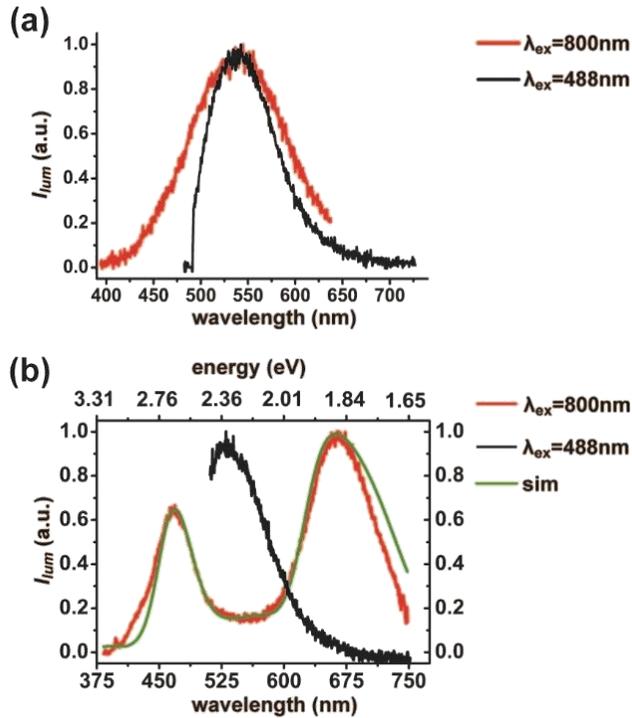


Fig. 1. (a) shows the typical plasmonic emission spectrum of a single gold nanosphere excited with one (black line) and two (red line) photons exhibiting a single emission band at 550nm. (b) displays the emission excited with one (black line) and two (red line) photons. In this case the pulse duration was reduced from 500 to 100fs causing a drastic change in the emission behaviour. The green line shows a corresponding simulated spectrum.

References

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