Properties of Two-Photon Induced Emission from Dendritic Silver Nanoclusters

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Abstract. The emissive properties of dendritic silver thin films due to two-photon excitation are explored. Emission is observed to occur at points more than 40 μm from the focal spot, and is polarization dependent.

1. Introduction

Silver nanoparticles are known to enhance and amplify local electromagnetic fields. Reportedly strongest in aggregates of a fractal nature, this effect causes enhancements in nonlinear optical processes such as surface-enhanced Raman scattering (SERS), second harmonic generation (SHG), and multiphoton photoemission [1]. We report here on our observations of polarized two-photon induced emission from thin films of silver nanoparticle aggregates at distances over 40 μm from the focal spot of a femtosecond laser ("remote emission"), as well our ability to control such emission by altering the polarization and/or spectral phase of the excitation beam**.**

2. Experimental Methods

To prepare the thin films, silver nanoparticles are synthesized by the citrate reduction of $AgNO_3$. Cluster formation is then induced by the addition of fumaric acid. The clusters were allowed to precipitate onto quartz substrates for three days. A Ti:sapphire femtosecond laser (800 nm center wavelength, 13 fs pulse duration, 80 nm bandwidth) was used to carry out the experiments presented here. The beam was brought into an inverted microscope and focused onto the sample with a 60x/1.45 NA objective to a diameter of < 1 μm. Phase distortions in the pulse due to the objective and other optics were compensated with MIIPS [2]. Emitted light was imaged with a electron multiplying CCD camera.

3. Results and Discussion

Excitation of the thin films results in strong two-photon induced luminescence consistent with surface plasmon-mediated fluorescence of silver oxide [3, 4]. This emission occurs both at the focal spot (< 1μm diameter, "local") as well as at distances up to 40 μm (results not shown) from the focal spot ("remote"). The

remote emission is observed to be polarized, with the incident polarization not necessarily preserved. Additionally, the polarization of the incoming beam can be used to control the two-photon emission of various regions (see Fig. 1).

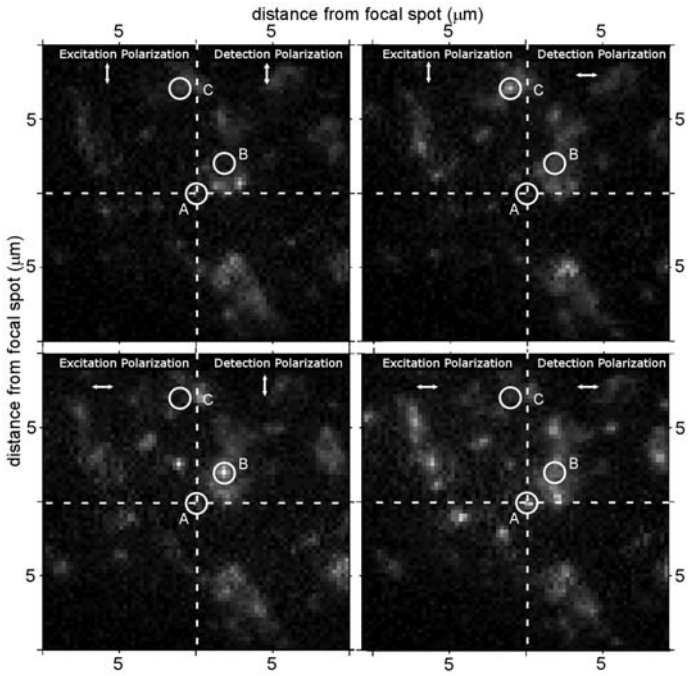

Fig. 1. Each panel is an image of the same region of silver (focal spot is noted by crosshairs and circle A) under different polarization conditions. The top panels were excited with vertically polarized light, while the bottom panels were excited with horizontally polarized light. The left panels show the vertically polarized component of emitted light, while the right panels show the horizontally polarized component of emitted light. A direct comparison between left and right panels (e.g. circle C, top) shows the polarized nature of the emitted light, while a direct comparison between top and bottom panels (e.g. circle B, left) shows how polarization can be used to control whether or not a particular region emits.

We have also explored the influence of phase-shaped pulses on the remote emission. By applying a sinusoidal phase function of the form $1.5\pi \sin(12\omega - \delta)$ across the spectrum of the laser pulse (the frequency, ω, is in fs^{-1}), and varying δ, we observe that regions of emission can be selectively and reproducibly turned on and off (see Fig. 2).

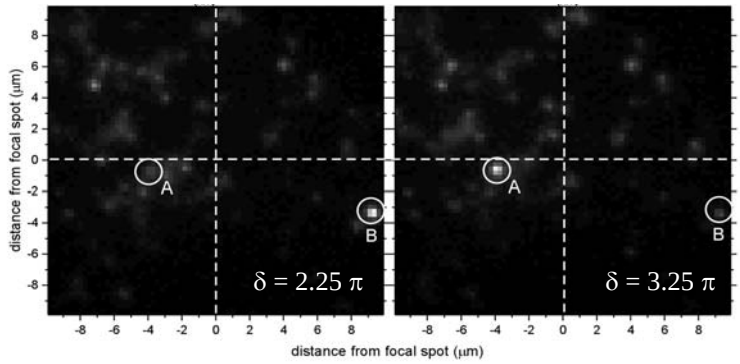

Fig. 2. Both panels are images of the same region of a silver sample (focal spot is denoted by crosshairs) in which the spectral phase has been modified by the application of a sine function with different δ values. It can be seen that the spectral phase of the laser pulse can be used to control emission from the silver (see circles A and B).

4. Conclusions

We have reported here that by altering the polarization or the spectral phase of a femtosecond laser we can control remote two-photon induced emission from dendritic silver nanoclusters. This has much potential in the field of plasmonics, which seeks to develop the ability to control the transportation of optical information along nanowires.

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