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Mode mapping of plasmonic stars using TPL microscopy

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Abstract. We investigate the far-field and near-field optical properties of a complex plasmonic system made of gold disks forming a star-like pattern. Scattering spectroscopy first enabled us to monitor the resonant behavior of the structures. Two-photon luminescence (TPL) microscopy was then used to probe the local fields under different orientations of the incident linear polarization. Unlike the scattering spectrum, the TPL distribution over the structure is found to depend drastically on the incident polarization state, in good agreement with a full 3D theoretical simulation of the electric near-field intensity.

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1. Introduction

Resonant metal nanostructures supporting localized surface plasmons [1] potentially offer the opportunity of extending optics down to the subwavelength scale and contributing to the elaboration of future nano-optical devices. Beyond the intrinsic enhancement and confinement of fields nearby an isolated particle, significantly stronger optical energy concentration can be achieved when involving the electromagnetic coupling between several objects. In particular, it is now well known that pairs of particles (also known as coupled antennas) separated by a dielectric nanogap are capable of confining the local fields beyond the diffraction limit [2]–[6]. This property has already opened new opportunities in ultra sensitive sensing of biomolecules [7], enhanced spectroscopy [8] and in enhancing the efficiency of photon sources and detectors [9, 10]. More recently there has been a growing interest in exploiting such concepts to achieve dynamical spatial control of the subwavelength optical fields [11]–[14]. Dynamical control of electromagnetic fields at the nanoscale is anticipated to have applications in the domains of nano-optical multiplexing and selective optical addressing of isolated nano-objects.

In this paper, we study star-like plasmonic structures whose optical near-field features a strong dependence on the polarization of the illumination light. Recent two-photon photoemission electron microscopy (PEEM) measurements on similar structures have successfully demonstrated the adaptive control of the local optical fields distribution though a complex shaping of the incident polarization state [11]. Here, we combine far-field spectroscopy and two-photon luminescence (TPL) microscopy to investigate the local optical response of this geometry to different orientations of a linearly polarized excitation. The TPL distribution over the structure is found to be strongly dependent on the incident polarization state, in good agreement with the corresponding electric near-field intensity map computed using the Green dyadic method.

2. Experimental details and results

The sample consists of an arrangement of gold disks fabricated by e-beam lithography on an indium-tin-oxide (ITO)-coated glass substrate using a FEI-QUANTA 200 scanning electron microscope equipped with a Raith-Elphy+ lithography control. A poly (methyl methacrylate) (PMMA) film (150 nm thickness) was spin-coated onto the substrate and then baked for 2 h at 175 °C. The resist was patterned using the e-beam at an acceleration voltage of 30 kV. After exposure, the PMMA resist was developed using MIBK:IPA. A 2 nm Ti adhesion layer and a 40 nm Au layer were deposited using e-beam evaporation. The pattern was recovered onto the substrate using a lift-off process.

The fabricated structures consists of six 200 nm disks arranged in a star-like shape (figure 1). The gap between consecutive disks was measured to be about 30 nm. Several of these structures were fabricated on the substrate in the form of a matrix. A period of 2 µm prevents significant electromagnetic interaction between adjacent structures.

2.1. Far-field scattering spectroscopy

In order to study the resonance characteristics of the structures, far-field scattering spectroscopy measurements were performed under dark-field illumination using a spectrometer coupled
to an optical microscope through a multimode fiber. A polarizer placed in the illumination path allowed us to control the orientation of the incident linear polarization in the sample plane. In order to account for the spectral characteristic of our white light source, each of the scattering spectra was normalized by the illumination spectrum. The results, including reference measurements on isolated disks, dimers and trimers, are summarized in figure 2 for different orientations of the incident polarization.

For a polarization along the vertical axis ($\theta = 0^\circ$), the star features a broad resonance centered around 800 nm, significantly red shifted with respect to the respective resonances of the single disk, dimer and trimer (figure 2(b)). This tends to indicate that, under this polarization, the structure supports a resonant mode, resulting from the complex near-field coupling between the constitutive disks, which is governed neither by dimers nor the trimer contained within the arrangement. Interestingly, very similar resonances are found for $\theta = 60^\circ$ (figure 2(d)) and $\theta = 90^\circ$ (figure 2(c)) revealing no significant dependence on the incident polarization of the scattering spectroscopy from the star.

2.2. TPL microscopy

While far-field spectroscopy measurements have provided an insight into the resonant behavior of the structure, we are now interested in investigating the associated local field distribution.

Different techniques have been used in the past to map the field around coupled metal nanostructures. Imura et al [15] have applied two-photon and Raman near-field imaging for mode mapping of gold agglomerates and recently antennas have been imaged with scattering-type scanning near-field optical microscopy (SNOM) [16, 17]. The high resolution snapshots of the mode distribution are in fair agreement with theoretical predictions, although the field magnitude in the gap appears substantially weaker than predicted. Indeed the SNOM probe itself can interfere with the intrinsic properties of the structure under study and affect the resonance conditions [18, 19]. Therefore approaches free of scanning tips are preferred, despite
the intrinsic limited spatial resolution [20]. Confocal nonlinear microscopy is particularly promising as the higher power dependence on the electric field makes the nonlinear response preferentially sensitive to the most intense fields close to the metal. In particular, two-photon induced luminescence (TPL) through inter-band transitions of gold provides a powerful method to study gold antennas [21]. Indeed TPL microscopy has recently been shown to be well suited to probe plasmonic fields around gold nanostructures [5, 6], [22]–[25]. Despite the observed TPL enhancement, the low spatial resolution so far has not allowed a direct correlation of the TPL signal with the near-field distribution across the structures.

On the basis of our far-field spectroscopy data, we performed TPL microscopy on single stars. In our microscope setup, the structure was illuminated by a pulsed Ti:sapphire laser (150 fs pulses, 76 MHz repetition rate, average incident power 30 µW). The linearly polarized beam was tightly focused by an immersion oil 100x objective with NA = 1.25. The resulting intense fields induce locally a two-photon absorption process in gold which leads to a wide-band photoluminescence emission [21]. The luminescence was collected back through the same objective and sent to an avalanche photodiode diode after being filtered from the excitation light. Scanning the sample with respect to the illumination spot enabled us to map the distribution of TPL intensity around the structure with an optical resolution of about 200 nm, as determined by

Figure 2. (a) SEM micrographs (scale bars = 200 nm) and (b)–(d) normalized scattering spectra of single disk, dimers, trimers and stars for different orientations of the incident linear polarization.
the combination of two photon excitation and high NA. A λ/2 waveplate in the incident beam path allowed to control the polarization angle of the incident beam. The incident intensity of the light was always kept constant for all the polarization angles.

Figure 3 shows the TPL scans recorded over a single structure at an incident wavelength of 730 nm for four different orientations of the incident field. For an incident field polarized along the vertical axis (θ = 0°) the TPL intensity is found to be stronger at the extremities of the three arms (figure 3(a)). On rotating the polarization by 60° (counter clockwise), a drastic change in the TPL distribution is observed (figure 3(b)). Now, the signal dominates along the left bottom arm. This tends to indicate the local response is here determined by some gap effects where the incident field would create strong surface charge gradients resulting in strong field magnitudes. Indeed a symmetrical pattern is achieved when aligning the polarization along the right bottom arm (130°) (figure 3(d)). Finally, a much stronger TPL emission concentrated at the top arm is measured when the polarization is orthogonal to the top arm (90°) (see figure 3(c)).

At this stage, we are interested in comparing our experimental data with numerical simulations of the electric near-field intensity distribution based on the Green dyadic method [26]. In our model, the structure is illuminated through a glass substrate under normal incidence by a linearly polarized plane wave at 730 nm wavelength. Although the calculations of the local mode-field distributions are in general not equivalent to the experimental configuration, the excitation of the structure can be expected to be related to the strength of the corresponding local mode field. In order to account for the finite resolution of our microscope and enable better comparison with our experimental data, we applied a Gaussian convolution (200 nm waist) to the calculated near-field distribution. Figure 4 shows the theoretical maps for two directions of the incident polarization, θ = 0° and θ = 90°. A comparison with the corresponding experimental maps of figure 3(a) and 3(c) shows that the distribution of the TPL emission recorded in our experiment provides direct insight into the actual near-field mode distribution across the structure. In particular the field concentration in the upper branch for 90° polarization and at both lower branches under 0° polarization, respectively, is well restituted. Further
Figure 4. Numerical simulations of the normalized E-field intensity computed 20 nm above the structure for two different orientations of the incident polarization and an incident wavelength of 730 nm. A convolution with a 200 nm Gaussian profile was performed.

Figure 5. Polarization dependence of the TPL intensity over a star-like structure. The red, blue and green data points represent the maximal TPL signal at the top, left and the right arms, respectively.

discrepancies in the relative intensities and exact positions of maxima are attributed to deviations in the structural parameters compared with the ideal object considered in our model.

Relying on the correlation between TPL maps and local field distribution, a further analysis of our experimental data enables the actual ability to control the confinement of fields in the star geometry to be assessed through a simple rotation of the incident linear polarization. To illustrate this concept, we plot in figure 5 the polarization dependence of the maximum TPL intensity at each of the three arms. Each data point corresponds to an average TPL intensity over an area of $5 \times 5$ pixels. This graph clearly depicts how the confinement of the local field at determined locations of the structures can be controlled by the polarization state. A maximum field concentration is achieved at the top arm when the polarization is perpendicular to the
vertical axis (90°). Conversely, the signal is maximal at the bottom-left and bottom-right arms for a polarization at 60° and 130°, respectively. We believe such behavior opens possibilities for selective optical addressing of different objects located nearby the extremity of each of the star arms.

3. Conclusions

In this paper, we present a far-field and near-field study of star-like gold nanostructures. While the far-field spectroscopic data do not show significant dependence on the orientation of the incident linear polarization, TPL measurements reveal a drastic redistribution of the local field across the structure. We believe that the resulting local reconfiguration of the field at the structure extremities, well corroborated by full 3D numerical simulations, could enable the optical addressing of isolated adjacent nanobjects to be dynamically controlled.

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