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Abstract: Second-harmonic generation from gold nanoisland films increases with the dielectric TiO_2 coating thickness. This occurs although the plasmon resonance shifts away from the second-harmonic wavelength, due to enhanced non-resonant local fields at the fundamental wavelength.

OCIS codes: (190.0190) Nonlinear optics; (190.4400) Nonlinear optics, materials; (310.6628) Subwavelength structures, nanostructures

1. Introduction

The optical properties of metal nanostructures are governed by their localized surface-plasmon resonances (LSPR), giving rise to strong local fields. This resonant behavior is especially important for nonlinear optical phenomena, which depend on higher powers of the excitation field. The tuning of LSPRs has been widely utilized to enhance the optical nonlinearity of metal nanostructures, e.g., for second harmonic generation [1,2]. However, the prior studies were mainly focused on tuning of the LSPR peak position to match the fundamental or harmonic wavelengths while neglecting the non-resonant part of the optical response. In this paper, we demonstrate enhancement of second-harmonic generation (SHG) from metal nanoparticles based only on the non-resonant response. We study gold nanoisland films coated with layers of amorphous titanium dioxide (TiO_2) with varying thickness. When the TiO_2 thickness increases, the strength of plasmon resonance grows and the LSPR peak shifts away from the SHG wavelength. As a result, the tail of the resonance at the fundamental wavelength becomes stronger, and the contribution of the optical response at the fundamental wavelength dominates over that at the second harmonic wavelength to the total local-field factor (LFF) for the SHG process.

2. Sample preparation and experiments

The samples were fabricated by air annealing of gold films deposited onto fused silica substrates by evaporation. The resulting films consist of separate nanoislands of 10-20 nm in size with a LSPR around 520 nm wavelength (Fig. 1a). The films were subsequently coated with a layer of amorphous TiO_2 using atomic-layer deposition. We fabricated several nanoisland films with identical parameters and varying thickness of the TiO_2 layer.

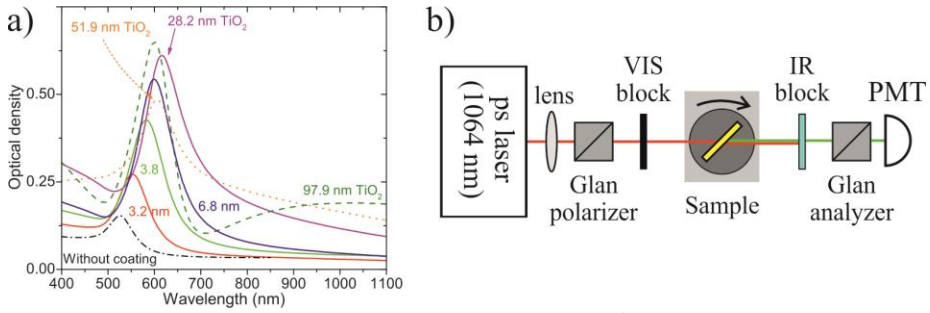

Fig 1. a) Measured absorption spectra. b) Experimental setup for nonlinear experiments.

The absorption spectra of the samples are shown in (Fig. 1a). The SHG experiments were carried out using a Maker-fringe technique (Fig. 1b) [3]. A picosecond laser (70 ps, 1064nm) was used as the source of fundamental light. In order to verify the source of the enhancement, experiments were carried out on an uncoated nanoisland film, nanoisland films coated with varying thickness of TiO_2, and bare substrates coated with varying thicknesses of TiO_2 (Fig. 2a). Due to the SHG response from the film being much stronger than that from the back surface of the substrate, the effective SHG signal strength can be estimated from the average of the neighboring extrema near the envelope maximum of the Maker fringes. The SHG signals from coated nanoisland films normalized to the response from the uncoated film are shown in Fig 2b.

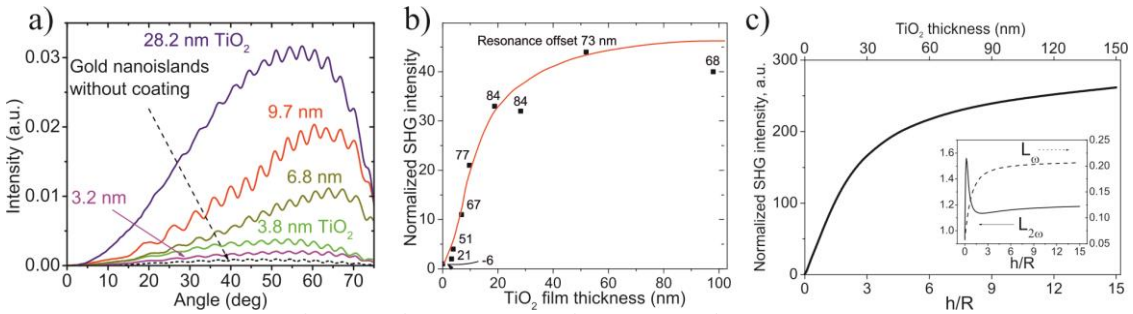

Fig 2. a) SHG irradiance versus angle of incidence for several samples from the Maker-fringe experiment. b) Estimated SHG signal level as a function of film thickness. Numbers next to data points indicate distance between the resonance and SHG wavelength. Line is guide for eye only. c) Calculated total SHG enhancement due to LFFs (particle radius 10 nm). Inset: LFFs for fundamental and SHG frequencies.

3. Discussion

To interpret the experimental results, we assume that the SHG susceptibility tensor component normal to the particle surface dominates. Thus, the relevant LFF for frequency Ω can be written as

$$L_{\Omega} = \frac{\left\langle \left| E_{\Omega}\left(r = R\right) \right| \right\rangle_{\theta,\phi}}{\left| E_{\Omega}^{0} \right|}, \tag{1}$$

where E_{ω}^{0} is the incident electric field, $\left|E_{\omega}\left(r = R\right)\right|$ is the normal component of the field at the particle surface, and the average is calculated over spherical angular coordinates θ and ϕ. We applied Eq. 1 for nanoparticles shaped like a truncated sphere and note that the SHG irradiance depends on the LFFs as $I_{SHG} \propto L_{2\omega}^{2} L_{\omega}^{4}$, where ω (2ω) is the fundamental (SHG) frequency [4]. The calculated SHG power enhancement values for different ratios between the coating thickness *h* and particle radius *R* for a truncated gold nanosphere (truncation angle 50°, particle radius 10 nm) are shown in Fig. 2c. The relative permittivities of TiO_2 and fused silica were taken to be 5.5 and 2.25, respectively, and the permittivity of gold was taken from Ref. 5 [5].

Our experimental results show that for increasing thickness of TiO_2, the SHG efficiency of the gold nanoisland films increases by as much as a factor of 45 while the LSPR shifts towards longer wavelengths away from the SHG wavelength. Moreover, the SHG from amorphous TiO_2 coating alone was observed to be negligible. This result is due to the increase of the non-resonant LFF at the fundamental wavelength dominating over the decrease of the LFF at the SHG wavelength. This result is also qualitatively supported by our model, and the quantitative difference of around 6 in the enhancement factor (Figs. 2b vs 2c) can be explained by size distribution of nanoparticles and Fresnel reflection effects at the interfaces of TiO_2.

4. Conclusions

In conclusion, we have studied the nonlinear response of metal nanoisland films coated with varying thickness of titanium dioxide. As a general result, the changes in dielectric environment give rise to an increase in the overall local-field factor for the SHG process, even though the plasmon resonance shifts away from the SHG wavelength. Experimentally, a 45-fold enhancement was observed. We believe that this phenomenon is of great importance and can be observed in a variety of contexts, independent of the particular shape or even the size of the nanoparticles. In addition, the effect is not limited to second-harmonic generation but should open new opportunities in all cases where the tailoring of local fields can be used to advantage in photonic applications.

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