Improving Plasmonic Nanoantennas

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Abstract: Improvements in energy damage threshold of gold nanoantennas were studied using stabilizing dielectric films. Annealed nanoantennas show a stronger plasmonic resonance, but have a decreased damage threshold.

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Nanoantennas have gathered great attention in recent years. The nanoantenna array has been widely used in near-field optics, nonlinear optics, and fluorescence research due to its enhanced localized electrical field [1, 2]. In applications using high intensity pulsed lasers, especially for nonlinear optical studies, high intensity damage may occur. Several studies have noted that under pulsed laser irradiance, the nanostructures will change shape and possibly be ejected from the substrate [3-5]. In order to stabilize the nanoantennas on the substrate, and to improve the damage threshold of gold nanoantenna under picosecond laser irradiance, we use a dielectric film coating which has a high melting temperature.

There have been numerous studies dealing with the electron-phonon coupling thermal dynamics in pulsed laser interaction with nanoparticles [5-7]. For example, laser cleaning is a promising method to achieve efficient cleaning of a semiconductor surface [8-13]. Laser cleaning forces smaller particles (< 100 nm), which can not be removed through conventional ultra-sonic cleaning, to be removed when the particles/substrate system interacts with the short laser pulses, resulting in thermal expansion. However, most of the existing theories and experiments focus on how efficiently remove particles. There has thus far been relatively little research considering how to stabilize nanoparticles from pulsed laser irradiance.

The mechanism of laser cleaning is shown in Fig. 1 (a), where the removal of particles is mainly due to the rapid thermal expansion induced by laser heating. Although thermal expansion of the particles is very small ($x' - x_0$ is within nanometers), it is sufficient to provide a very large acceleration due to the very short expansion time scale [12]. Nanoantenna ejection occurs when the final kinetic and elastic potential energy acquired by the nanoantennas is greater then the total adhesion energy (van der Waals force and the hydrogen bond energy), i.e., $E_{\text{kin}} + E_{\text{elast}} \geq E_{\text{vDW}} + E_{\text{H-Bonds}}$. E_1, in Figure 1, is the sum of kinetic and elastic energy, which is positively correlated to the energy absorbed by nanoantennas, $E_{\text{abs}} = F\sigma_{\text{abs}}$ (F is the laser fluence value and σ_{abs} is the nanoantenna absorption cross-section). F_0 at which nanoantenna ejection begins is described as the damage threshold value. In order to increase F_0, a dielectric layer with thickness d is coated on top of the nanoantennas as shown in Figure 1 (b). This dielectric film supplements the existing adhesion energy with E_2, which is related to the film thickness and material properties. Therefore the required energy for ejection in the coated case (b) is larger than in the uncoated case (a). Since more energy may be absorbed (greater E_{abs}), this directly corresponds to an increased damage threshold.

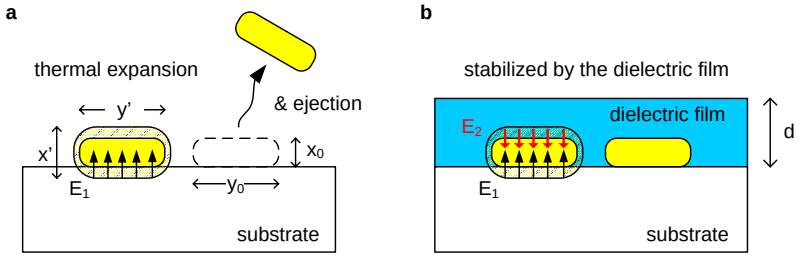

Fig. 1. A schematic of laser heating and thermal expansion of nanoantennas. (a) nanoantennas/substrate in air; (b) nanoantennas covered with a dielectric film.

To test this hypothesis, gold nanoantennas were fabricated using E-beam lithography on an indium-tin-oxide (ITO)-coating glass substrate which has a 15 nm ITO layer. The antennas consist of coupled square particles of 100 nm by 100 nm, and a thickness of 30 nm. The gap between the particles is 30nm, and the array periodicity is 400 nm in both the x and y directions. The FE-SEM image in Figure 2 (a) shows the resulting nanoantenna structure. From

far-field spectra measurements, the x-polarization resonance wavelength (λ_r) is observed at 710 nm, and the transmittance (T), reflectance (R), and absorption value (A=1-R-T) at λ_r are [0.16, 0.39, 0.45] respectively. An identical nanoantenna sample was coated with a 100 nm silica film using an E-beam evaporator at 10^{-6} torr . The x-polarization resonance wavelength of the nanoantennas with the 100 nm silica film is red-shifted to 800 nm. The [T, R, A] value at λ_r = 800 nm is [0.11, 0.41. 0.48] respectively. Films of PMMA and organic molecules and damage threshold of nanoantennas with different electron relaxation rates were also studied.

A Ti:sapphire based OPA laser system was used to test the damage threshold of both samples. Each sample is placed at the focal plane of the laser, which is tuned to the respective resonance wavelength of each sample. The laser has a pulse duration is 2 ps, repetition rate of 10 Hz, and a focused beam waist (Φ) of 35 um. The incident electric field polarization is set to the primary polarization (across the gap, x-polarization). By varying neutral density filters before the sample, the irradiance intensity is increased from 0.5 mJ/cm^2 to 14 mJ/cm^2 . The damage threshold is found when a sudden transmission increase occurs, indicating nanoantenna ejection. Table 1 shows the damage thresholds for nanoantennas with and without a silica film as 12.11 mJ/cm^2 and 2.52 mJ/cm^2 . Images of the resulting nanoantenna damage, with and without the silica film, are shown in Fig. 2 (c) and (b).

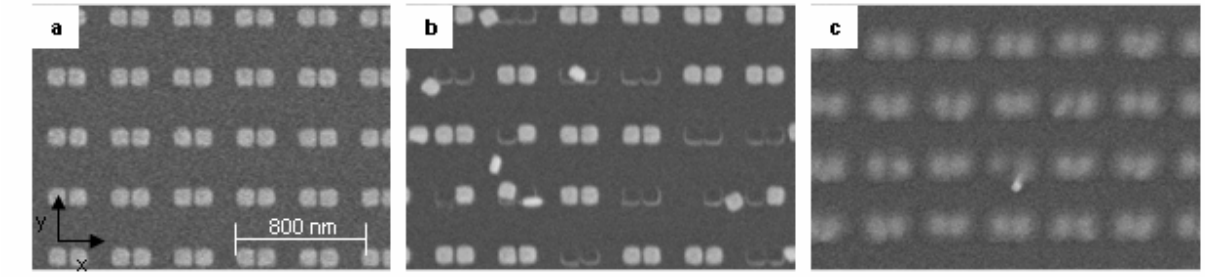

Fig. 2. (a) An SEM image of the initial nanoantennas. (b) Damage to nanoantennas in air: ~50 % of nanoantennas are ejected at 2.52 mJ/cm^2 . (c) Damage to nanoantennas covered with 100 nm silica: only few nanoantenna is ejected at 12.11 mJ/cm^2 .

Table 1. Comparison of nanoantennas with and without a 100 nm silica film.

Nanoantennas	λ_r (nm)	Absorption	Damage threshold F_0 (mJ/cm^2)	E_{abs} (J) per nanoantenna
in air	710	0.45 ± 0.02	2.52	1.81×10^{-12}
in 100 nm Silica	800	0.48 ± 0.05	12.11	9×10^{-12}

We have demonstrated that covering nanoantennas with a 100 nm silica film can increase the damage threshold by five times for picoseconds laser irradiance. This study is necessary in order to design and use nanoantennas in nonlinear optics, and other high-intensity applications.

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