Invited Paper

Photothermal phenomena in plasmonics and metamaterials

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ABSTRACT

Our recent theoretical and experimental investigation of the photothermal effect in a planar metamaterial absorber is reviewed in the present paper. The observed ultrasensitive photothermal heating in such an absorber nanostructure irradiated by a pulsed white-light source is elaborated with a simple yet compelling heat transfer model, which is subsequently solved with a finite-element method. The simulation results not only agree with the experimental finding, but also provide more detailed understanding of the temperature transition in the complex system.

Keywords: photothermal effect, plasmonics, light absorber, heat transfer, melting temperature.

1. INTRODUCTION

Metallic nanoparticles exhibit arguably the most efficient photothermal response when excited by a laser close to their plasmonic resonance frequencies[1,2]. Such an ultrasensitive photothermal effect has already found applications in e.g. optical data storage[3], sensing[4], and even cancer therapy, etc. Considering the sub-wavelength resolution of the heating, the photothermal phenomenon can potentially play a critical role in designing remotely tunable nano-devices. One can also, as a by-product, utilize the photothermal process to fabricate gold nanoparticle arrays with a great uniformity and with smooth particle surfaces for, e.g. enhanced plasmonic devices, sensing devices, or catalysts.

In a recent experiment[6], we illuminated a metamaterial nanostructure with a pulsed broadband light source. We observed, most importantly, re-shaping of its top-layer gold particles from thin blocks to spherical domes due to surface melting. Here we model the process by taking both electromagnetic scattering and heat transfer dynamics into account. The results can qualitative explain the observations found from the experiment.

2. EXPERIMENTAL OBSERVATIONS

One element of the metamaterial absorber is shown in Fig. 1. Refer to the figure caption for information regarding material and geometrical parameters. The metamaterial has a peak absorption wavelength around 1.58μm and 2μm, depending on incidence plane and polarization[5].

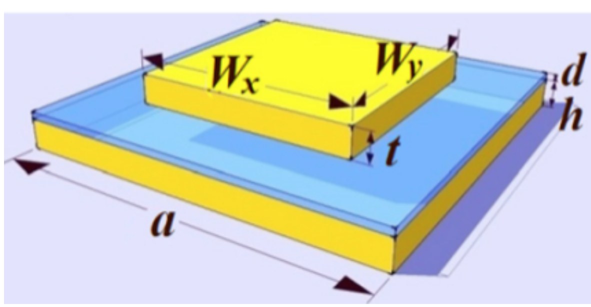

Fig. 1: A unit of the metamaterial absorber. Yellow regions are gold, blue region is Al2O3. the geometrical parameters are: Wx=170nm; Wy=230nm; a=310nm, t=40nm; d=10nm; h=60nm.

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The metamaterial is formed by repeating the unit over the xy plane. Although not shown in the figure, there is a silica substrate further beneath the gold film, which is important in our later heat transfer model. The fabricated sample is shown in Fig. 2.

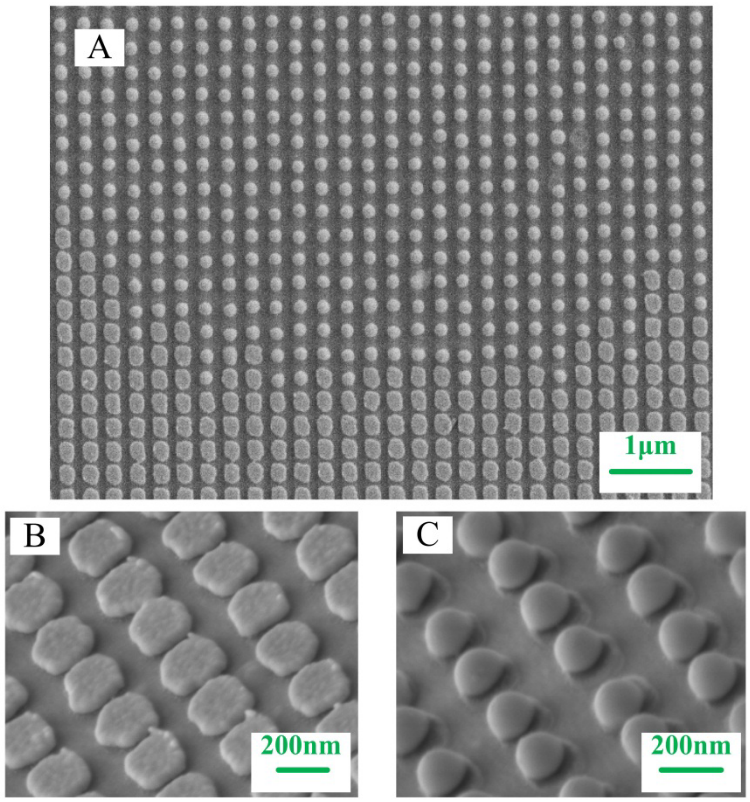

Fig. 2: (A) Top view of metamaterial absorber after laser Gaussian beam exposure. (B)Unchanged sample of metamaterial, (C) Reshaped sample of metamaterial due to sufficient laser pulse exposure.

We irradiate the fabricated gold nanostructure (Fig. 2B) by a focused broadband white light. The setup is illustrated in Fig. 3. After attenuation and focus, the beam has a diameter of 20μm with a power of 2.3mW when reaching the sample. The top gold particles are converted to spherical domes due to excessive heating and reshaping due to surface tension in liquid phase. Figure 2A shows a top-view scanning electron microscopy (SEM) image of the illuminated sample. One sees particles experience the reshaping from thin blocks to spherical domes inside a half-circle domain. While outside the half-circle domain, there are unchanged ones. The existence of a circular boundary separating the reshaped and unchanged regions is caused by the Gaussian beam shape of the incident light. Further increase the beam power we observe, of less technological interest, fragmented gold particles in the center and even severely burnt sample (not shown). Figures 2B and 2C compare the detailed oblique SEM views of the particles before and after irradiation. The particles in thin blocks in Fig. 2B have grainy top surfaces and rough side edges due to, respectively, the electron beam evaporation method used for metal deposition and the lift-off patterning process. In contrast, the gold particles after irradiation (Fig. 2C) have a much better surface quality. The spherical particles have an average radius of 80nm and a height around 110nm; their contacting surface to the Al_2O_3 layer has a radius about 70nm.

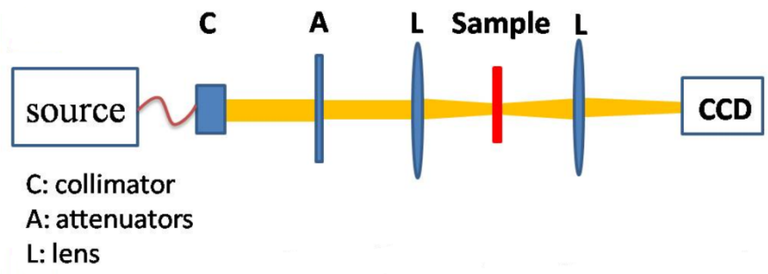

Figure 3. Schematic setup for the photothermal experiment.

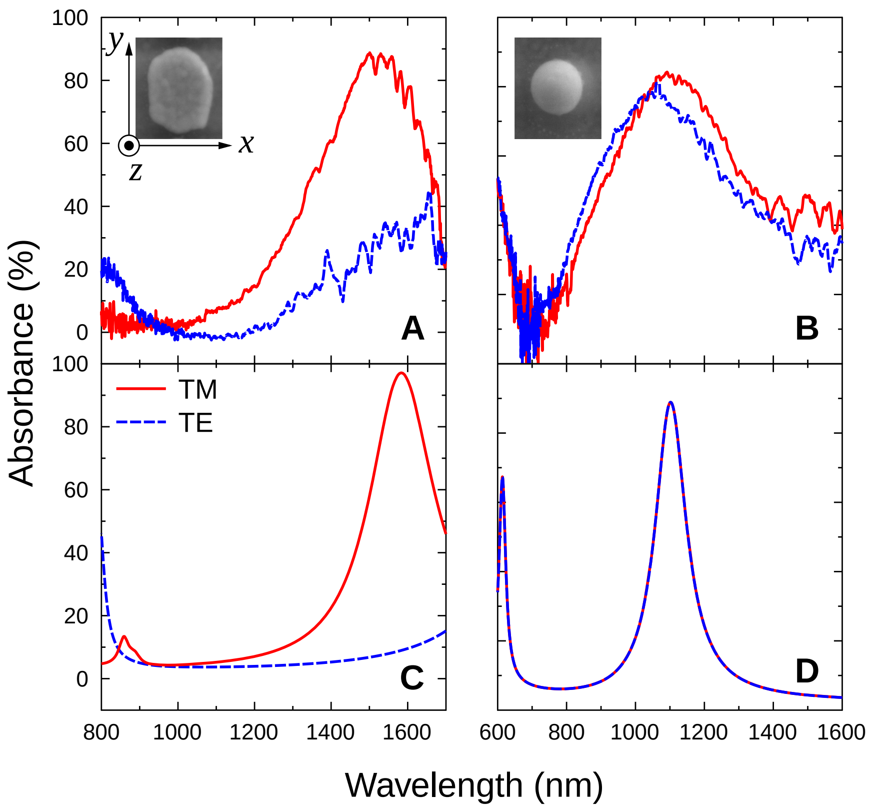

Fig. 4. Measured absorption spectra for (A) an unchanged sample region, and (B) a reshaped sample region at a 10º incidence angle. Plane of incidence: *xz*. Red-solid and blue-dashed curves are for TM and TE polarization respectively. (C,D) are simulated results for the situations described in (A,B) correspondingly. Insets: representative SEM images for a single sample unit.

The shape transformation and possibly crystalline of the gold nanoparticles should substantially influence the absorption characteristics of the metamaterial absorber. By using a homemade setup, we measure the absorbance of the sample at both rehaped and unchanged regions with a 10º incidence angle. We point out that the absorber in such a configuration in general has an absorption spectrum insensitive to the incidence angle. The plane of incidence intersects with the metamaterial in the direction along which the particles have a smaller size. Two measurement results are given in Figs. 4A and 4B for both the transverse-magnetic (TM) and the transverse-electric (TE) polarizations. For the sample region with rectangular nanoparticles (Fig. 4A), the absorption peak differs for the two polarizations, at 1.58μm for TM and ~2μm for TE[5]. For the region with dome-shaped nanoparticles (Fig. 4B), due to the higher symmetry of the unit cell, the absorption peaks for two polarizations almost overlap at 1.1μm. We carried out EM scattering simulations with structural parameters extracted from the experimental sample. The simulated absorption spectra for two sample regions are shown in Figs. 4C and 4D. Overall, the experimental results agree well with simulated results. This study suggests that an absorption spectrum can serve as a second signature for such a photothermal experiment.

3. THEORETICAL MODELLING

Our simulation considers a unit cell of the 3-layer metamaterial structure. The top gold particle is modeled with a square shape of size 200×200nm^2, which has a similar area compared to the fabricated ones. The simulation domain is 310×310×700nm^3. The heat transfer equation used is

$$C_s \rho \frac{\partial T}{\partial t} = k\nabla^2 T + Q_s \quad (1)$$

C_s, ρ, k is specific heat capacity, density and thermal conductivity of the material, respectively. Although bulk gold has a thermal conductivity of k_b=317 W/(mK), however we use a thermal conductivity of k_f=139 W/(mK) for the 60-nm thick gold film on the silica substrate. This is due to the fact that the sub-micron geometry as well as surface roughness drastically influences the thermal conductivity of a gold film. On the side walls of the model, the heat flux in horizontal direction is ignored. At the bottom of the domain, the silica boundary is set to a fixed temperature T_B=300K.

The heat source Q_s in Eq. (1) is calculated according to the absorbed light power[5], which also has a Gaussian temporal profile, after each light pulse. It has the expression of

$$Q_s(t) = \frac{R_{ab} E_u}{\Delta V \tau \sqrt{\pi}} \exp\left(-\frac{(t-t_0)^2}{\tau^2}\right). \quad (2)$$

where E_u is the position-dependent optical energy of a single pulse reaching a metamaterial unit, R_{ab}≈0.28 is the summed absorption ratio over the light source spectrum, ΔV is the volume of heat source, τ=1.5ns is time constant, t_0 is delay of the pulse. In our simulation model, we focus on a unit at the light beam center; correspondingly we have $E_u = 1.69\times10^{-10}$J. The photothermal heat source is assumed to be homogeneously distributed in the Au particle and in a portion of the Au film right beneath the Au particle.

The simulation is summarized in Fig. 5. In Fig. 5(A), we plot time variations of both the heat source and the temperature in the top nanoparticle. It is observed that at 1.5ns after the peak source power, the temperature of the nanoparticle reaches to its highest value 936K, which is well below the melting point of bulk gold 1337K. Therefore we theoretically confirm the temperature of the heated particles and more importantly quantitatively show the time scale over which the annealing occurs. The particles will gently cool down, almost close to room temperature, before the next pulse reaches. The heat dissipation is mostly due to heat conduction via the silica substrate. In Fig. 5(B), we show the temperature distribution in the unit when the nanoparticle reaches its highest temperature. It is seen that the gold film, although only 10nm away from the particle, has a lower temperature by around 50K. This suggests that the lower gold film is cooler and more difficult to be reformed thermally.

4. CONCLUSION

In conclusion, we have constructed a model for simulating the transient temperature variation in a metamaterial absorber when it is irradiated with a pulsed broadband light. Our numerical simulation not only confirms the experimental observations, but also provides quantitative insights to the ultrasensitive photothermal phenomenon. Our experimental and simulated results indicate that an artificially engineered metamaterial nanostructure can exhibit photothermal effects owing to a strong plasmonic resonance. After illuminating the absorber with a pulse broad band light, the top-layer gold particles reshape from thin blocks to spherical domes due to such strong plasmonic resonances. One can apply such a nanostructure to an area which can then be remotely heated optically. This can lead to new possibilities for all-optical thermal tuning, infrared imaging, micro-bolometer, thermal actuators, sensors, etc. At the same time, our observation of the gold nanoparticles reshaping promises a new route for fabrication of dome-shaped metallic nanoparticles and even other-shaped metallic components for improved plasmonic and metamaterial devices.

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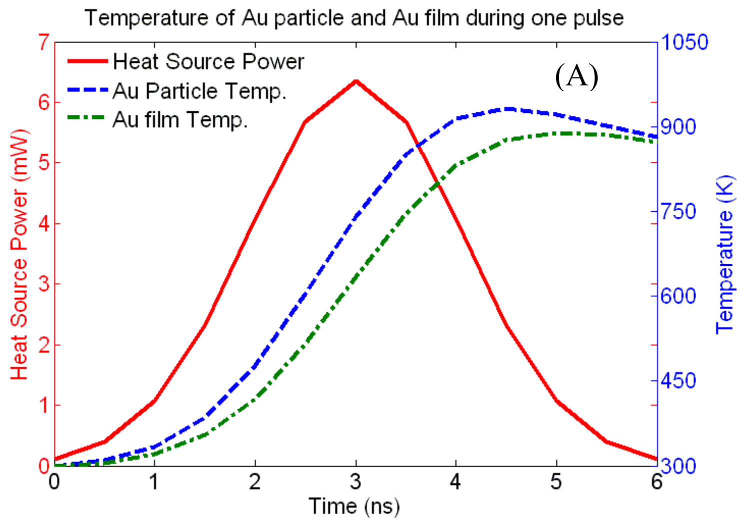

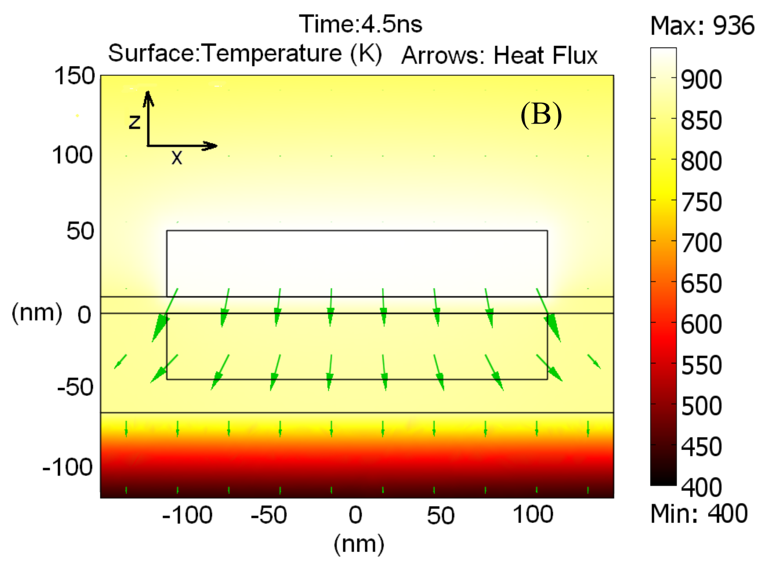

Fig. 5: (A) The transient heating of the top-layer gold nanoparticle. The trace is for the particle at center of the laser beam. (B) The temperature distribution in the unit cell studied in (A) at t=4.5ns when the particle reaches its highest temperature. The arrows indicate heat flux.

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