Modelling photothermal effect in a planar metamaterial absorber

Xi Chen, Jing Wang, Yiting Chen, Jiaming Hao, Min Yan, Min Qiu

Laboratory of Photonics and Microwave Engineering, Royal Institute of Technology (KTH)
Electrum 229, 16440 Kista, Sweden
Fax: + 46–87904090; email: min@kth.se

Abstract

The ultrasensitive photothermal heating in a plasmonic nanostructure irradiated by a pulsed white-light source is elaborated with a heat transfer model, which is subsequently solved with a finite-element method. The simulation results not only agree with our observation, but also provide more detailed temperature transition in the complex system.

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 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 batches with a great uniformity and with smooth particle surfaces for, e.g. enhanced plasmonic devices, sensing devices, or catalysts.

In a recent experiment, 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 thermal fusion. 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

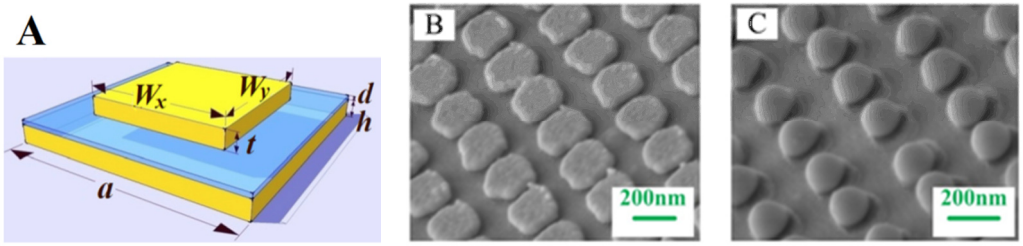

Fig. 1: (A) A unit of the metamaterial absorber. Yellow regions are gold, blue region is Al_2O_3. In the case of the fabricated sample (B), the geometrical parameters are: W_x=170nm; W_y=230nm; a=310nm, t=40nm; d=10nm; h=60nm. (C) The metamaterial after exposure to light pulses.

A unit of the metamaterial is shown in Fig. 1(A). 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]. 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

ISBN 978-952-67611-0-7

shown in Fig. 1(B). The pulsed white light source has a wavelength range of 0.5~2.4μm, a pulse width of 2.6ns, a repetition rate of 27kHz, and an average output power of 101mW. After attenuation and focus, the beam has a diameter of 46μm with a power of 36mW when reaching the sample. A region of the sample after exposure to the light source is shown in Fig. 1(C). It is shown that the top-layer particles have been melted and re-shaped to a uniform dome shape due to surface tension. This evidently suggests that the temperature within the nanoparticles has been elevated above the gold melting temperature, i.e. 1337K.

3. Theoretical modelling

Our simulation considers a unit cell of the metamaterial film. The top gold particle is modelled 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

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

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 influence the thermal conductivity of a gold film. Other material parameters in Eq. (1) are standard and are therefore omitted here for the space limit.

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, whose value depends on the unit's position relative to the incident beam center (r=0). For a unit at r=10μm, T_B=358K. We use a simplified model to estimate T_B: by using a structure with only the gold film and the silica substrate but with a large domain (greater than the incident beam size), we simulate heat transfer in this system for hundreds of heating cycles; the lowest metal temperature in a cycle is then obtained which serves the value for T_B.

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

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

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 r=10μm away from the light beam center; correspondingly we have E_u = 1.06×10^{-10}J. The 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. 2. In Fig. 2(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, above the gold melting temperature. Therefore we theoretically confirm the fusion of the particles and more importantly quantitatively show the time scale over which the melting occurs. The particles will gently cool down, almost close to room temperature, before the next pulse reaches (not shown). The loss of heat is mostly due to heat conduction via the silica substrate.

ISBN 978-952-67611-0-7

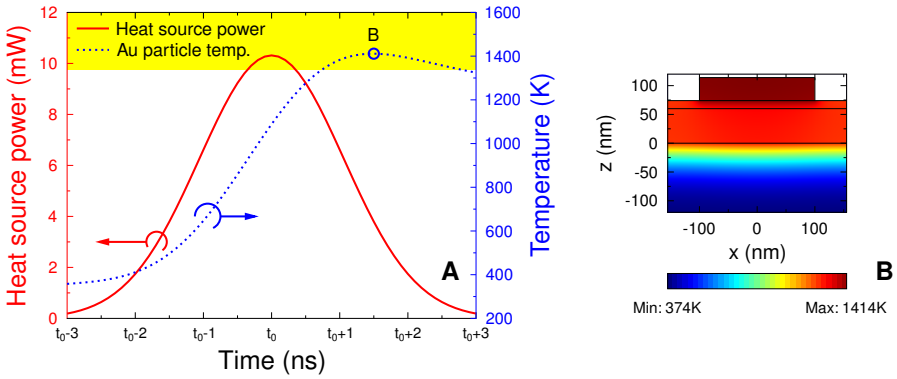

Fig. 2: (A) The transient heating of the top-layer gold nanoparticle. The trace is for the particle at r=10μm. The yellow-shaded region is for temperature>1337K, e.g. the melting point of gold. (B) The temperature distribution in the unit cell studied in (A) at $t=t_0+1.5$ns when the particle reaches its highest temperature.

In Fig. 2(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 100K. This suggests that the lower gold film may not be melted at this unit position.

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 ultra-sensitive photothermal phenomenon.

Acknowledgement: This work is supported by the Swedish Foundation for Strategic Research (SSF) and the Swedish Research Council (VR).

References

[1] S. Link, C. Burda, B. Nikoobakht, and M. A. El-Sayed, "Laser-induced shape changes of colloidal gold nanorods using femtosecond and nanosecond laser pulses," J. Phys. Chem. B, vol. 104, p.6152, 2000.

[2] A. O. Govorov and H. H. Richardson, "Generating heat with metal nanoparticles," Nanotoday, vol. 2, p.30, 2007.

[3] P. Zijstra, J.W.M. Chon, and M. Gu, "Five-dimensional optical recording mediated by surface plasmons in gold nanorods", Nature, vol. 459, p.410, 2009.

[4] W.S. Chang, J.W. Ha, L.S. Slaughter, and S. Link, "Plasmonic nanorod absorbers as orientation sensors", Proc. Natl Acad. Sci. USA, vol. 107, p. 2781, 2010.

[5] J. M. Hao, J. Wang, X. Liu, W. J. Padilla, L. Zhou, M. Qiu, "High performance optical absorber based on a plasmonic metamaterial," Appl. Phys. Lett., vol. 96, p. 251104, 2010.

