ADVANCED OPTICAL MATERIALS

Supporting Information

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Highly Efficient Photoacoustic Conversion by Facilitated Heat Transfer in Ultrathin Metal Film Sandwiched by Polymer Layers

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Derivation of heat transfer in thin film sandwiched by two polymer layers

The incident optical energy absorbed in the thin metal layer is represented by (considering optical reflection (R) and transmission (T), i.e., light absorption A = 1- R – T)

$$A\cdot F = F_{Metal} + F_{Polymer} \text{ or } 1 = \frac{F_{Metal}}{A\cdot F} + \frac{F_{Polymer}}{A\cdot F} = \gamma_M + \gamma_P, \quad (1)$$

where γ is the thermal energy at each layer, normalized to the input thermal energy (M: metal and P: polymer). The thermal energy in the metal layer is represented by

$$F_{Metal} = \rho C_P d\Delta T, \quad (2)$$

where ρ is the density, C_P is the heat capacity, d is the thickness, and ΔT is the maximum temperature increase. After the optical pulse excitation, the surface temperature increase (using Green's function approach) and the thermal energy in PDMS are represented by

$$\Delta T_s = \Delta T\left(z=0, t=0.55\tau\right) = 0.8876\frac{F_{PDMS}}{\sqrt{\rho C_p k \tau_l}}, \quad (3)$$

$$F_{PDMS} = 1.1266\sqrt{\rho_p C_{p,p} k_p \tau}\Delta T_s = 1.1266\rho_p C_{p,p} l_{th} \quad (4)$$

where the thermal diffusion length is $l_{th} = \sqrt{\alpha_p \tau}$. By combining the equations (1), (2), and (4), the thermal energy at each layer, normalized to the input thermal energy, can be represented by

$$\gamma_{metal} = \frac{\rho_m C_{p,m} d_m}{\rho_m C_{p,m} d_m + 1.1266\rho_g C_{p,g} l_{th}} \quad (5)$$

$$\gamma_{PDMS} = \frac{1.1266\rho_p C_{p,p} l_{th}}{\rho_m C_{p,m} d_m + 1.1266\rho_p C_{p,p} l_{th}} = \frac{1}{0.8876\dfrac{\rho_m C_{p,m} d_m}{\rho_p C_{p,p} l_{th}} + 1}. \quad (6)$$

For the sandwiched structure and modified heat penetration depth ($l_{th,s} = \left(1.269\alpha\tau\right)^{1/2}$), the formula (6) can be modified as

$$\gamma_{PDMS} = \frac{2}{\frac{\rho_m C_{p,m} d_m}{\rho_p C_{p,p} l_{th}} + 2}. \tag{7}$$

Figure S1 shows that the results obtained from the formula (7) are well matched with those calculated from the finite element method (COMSOL Multiphysics).

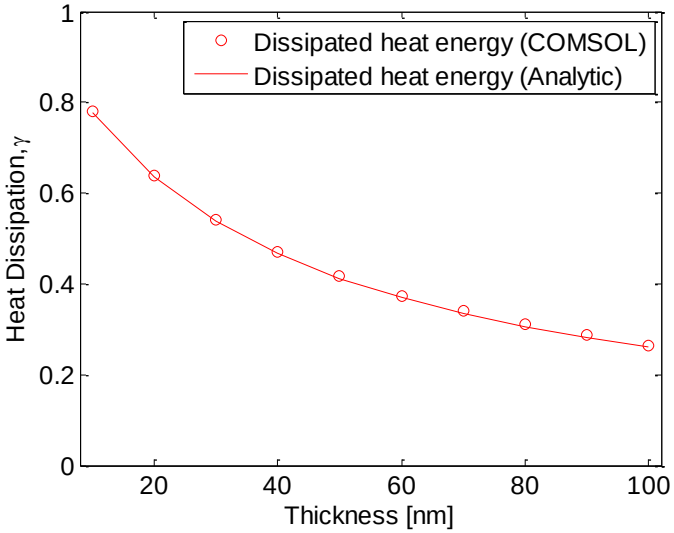

Fig. S1. Heat energy dissipated to the PDMS layers as a function of Ti thickness for the sandwiched structure (PDMS/Ti/PDMS). The symbol indicates results obtained from the finite element method (COMSOL Multiphysics), while the solid line represent results calculated from the formula (7).

Discussion 1. The effect of cavity roughness on the optical resonance

The roughness effect on the optical resonance was studied by using two different materials, PDMS and parylene. Thin PDMS layers prepared by spin coating were found to be rough, while parylene layers deposited by a thermal evaporator were very smooth. As shown in Fig. S2, the cavity effect in the structure based on a smooth parylene layer is well explained by the simulation result. On the other hand, the absorption spectrum of a rough PDMS layer is considerable different from the simulation. This discrepancy is possibly because the roughness of the PDMS layer could degrade cavity quality.

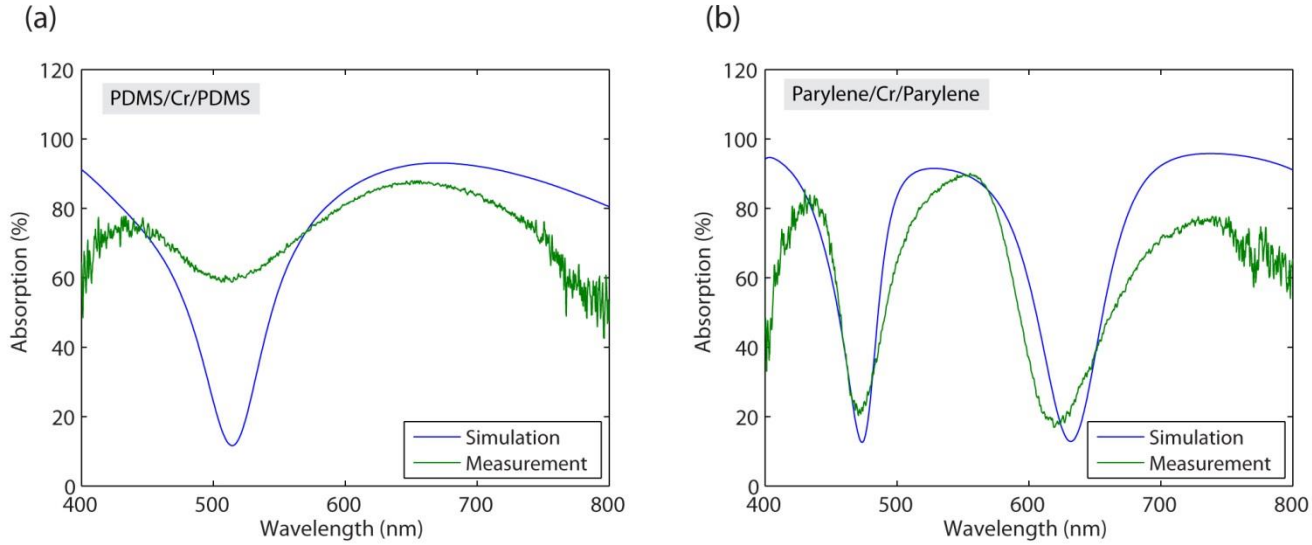

Fig. S2. The effect of surface roughness on the optical resonance. (a) Absorption spectrum for the PDMS-based sample. (b) Absorption spectrum for the parylene-based sample. The surface roughness of the spin-coated PDMS layer is much larger than that of the evaporated parylene layer.

Discussion 2. Nonlinear effect of PA generation

Although light absorption was increased two times, PA amplitudes were not increased as much as it is supposed to be, showing nonlinear effects. Such nonlinear effects may be caused by temperature-dependent material properties. Temperature in materials can increase linearly with laser pulse energy. If material properties remain the same, PA amplitudes are increased linearly. However, the material properties, more specifically Gruneisen parameter, are strongly related to temperature. Figure S3 shows the nonlinear effect. PA amplitudes in the PDMS-based structures saturate by increasing laser pulse energy. The result indicates that the Gruneisen parameter of PDMS decreases with temperature. In contrast, PA signals of the Cr (10 nm) in direct contact of water are greatly increased, meaning that the Gruneisen parameter of PDMS increases with temperature. Thus, the nonlinear properties of PDMS can explain the nonlinear increase in PA signals.

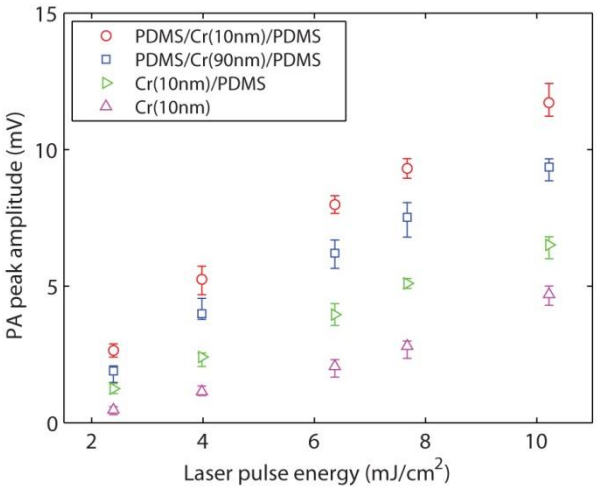

Fig. S3. Photoacoustic amplitude versus laser pulse energy. For the PDMS-based samples, the PA amplitudes show the amplitude saturation for higher laser pulse energies.