Supplemental Information

Influence of temperature-dependent dielectric constant on the photoacoustic effect of gold nanospheres

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The results of the absorption characteristics of gold nanospheres immersed in water are compared when the refractive index of water is $n(T, \lambda)$ or $n = 1.33$, as shown in Fig. S1. The value of $n(T, \lambda)$ is taken from Ref. 1. It is found that the difference between the two conditions can be ignored.

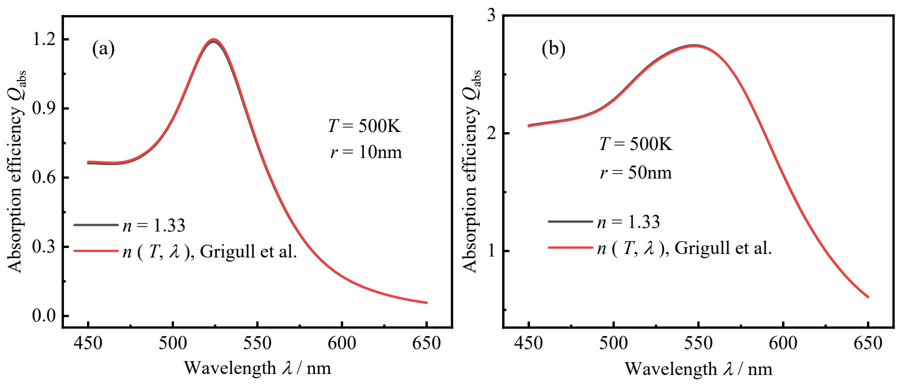

Fig. S1 The absorption efficiencies of gold nanospheres at water refractive indices of $n(T, \lambda)$ and $n = 1.33$: (a) and (b) are gold nanospheres with radii of 10 nm and 50 nm, respectively.

Fig. S2(a) shows that the absolute value of ΔQ_{abs} around the short wavelength (LSRP region) is larger than that of the long wavelength region (pink region). In addition, the maximum and minimum values of ΔQ_{abs} for gold nanospheres with a radius of 30 nm are always larger than those for gold nanospheres with a radius of 20 nm, which is due to the Q_{abs} positively correlating with the volume of gold nanospheres[2]. In contrast, as can be seen in Fig. S2(b), the relative rates of change around the short wavelength (LSRP region) (< 20%) are less than at the long wavelength region (pink region) (even larger than 100%). Moreover, the relative rates of change of gold nanospheres with a radius of 30 nm are smaller than those of gold nanospheres with a radius of 20 nm near the LSPR wavelength and the pink region.

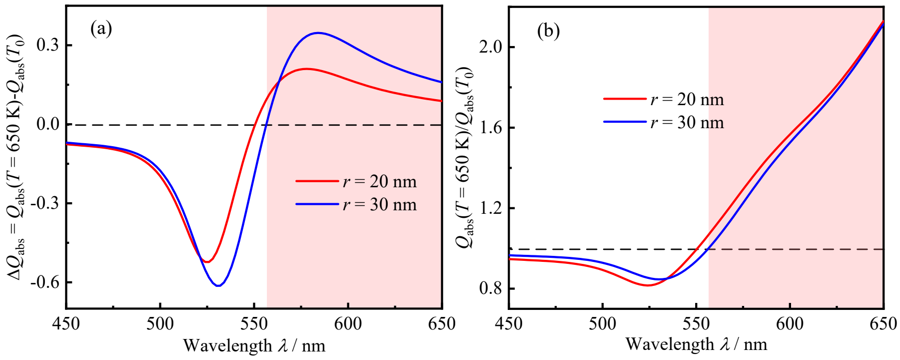

Fig. S2 The change of absorption efficiency of gold nanospheres at 650 K and 293.15 K: (a) ΔQ_{abs}; (b) $Q_{abs}(T = 650\ K)/Q_{abs}(T_0)$.

Because of the broadening of the spectra and the decrease of resonance energy, the absorption efficiency of 20 nm gold nanospheres decreases at the temperature increasing near the short wavelength, while increases at the long wavelength region. A "crossover point" coincidentally appears around 550 nm, implying that the absorption efficiency at this point is not affected by temperature. The "crossover points" are investigated for different radius gold nanospheres, as shown in Fig. S3 (the wavelength interval is 1 nm). The maximum change percentage χ in Q_{abs} at increasing temperature is defined as

$$\chi = \max\left(\left| \frac{Q_{abs}(T) - Q_{abs}(T_0)}{Q_{abs}(T_0)} \right|_{T:T_0 \sim 650\mathrm{K}} \times 100\% \right) \tag{S1}$$

The corresponding wavelength at the "crossover point", χ should be 0. It can be seen that a perfect "crossover point" does not exist. However, in practice it is still valuable to indicate wavelengths where the rate of change is sufficiently small (<1%). For gold nanospheres smaller than 50 nm radius, these wavelengths are around 550 nm, which gradually red shift with larger radius due to the red shift of LSPR wavelength.

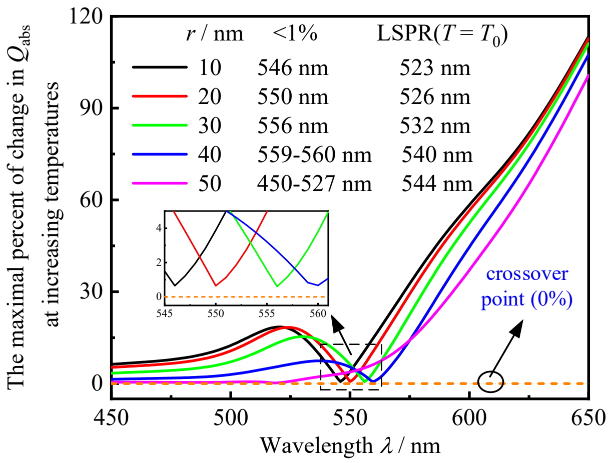

Fig. S3 The maximum change percentage of Q_{abs} at temperature increasing as a function of wavelength for gold nanospheres of different radii.

In this work, we consider the thermal resistance of the interface between gold and water (see Eq. 10), which makes the maximum temperature rise of gold greater than that of the surrounding water, as shown in Figs. S4(a) and S4(b). However, it can be seen that even if the two are different in value, the relative relationship between the maximum temperature rise of the gold nanospheres is the same as that of the surrounding water. In addition, for gold nanospheres of the same radius, the maximum temperature rise of gold nanospheres has exactly the same trend as that of the surrounding water as a function of wavelength, as shown in Figs.S4(c) and S4(d). Furthermore, the effect of temperature rise on the absorption efficiency of gold nanospheres is different at different wavelengths, but under the same pulse irradiation fluence, the relative size of $Q_{abs}(T_0)$ of different wavelengths is the same as that of temperature rise, as shown in Figs.S4(e) and S4(f). Therefore, the relative magnitude of maximum temperature rise of gold nanospheres and surrounding water can be obtained by comparing the relative magnitude of absorption efficiency $Q_{abs}(T_0)$ of gold nanospheres at reference temperature, even if the temperature dependence of dielectric constant is taken into account.

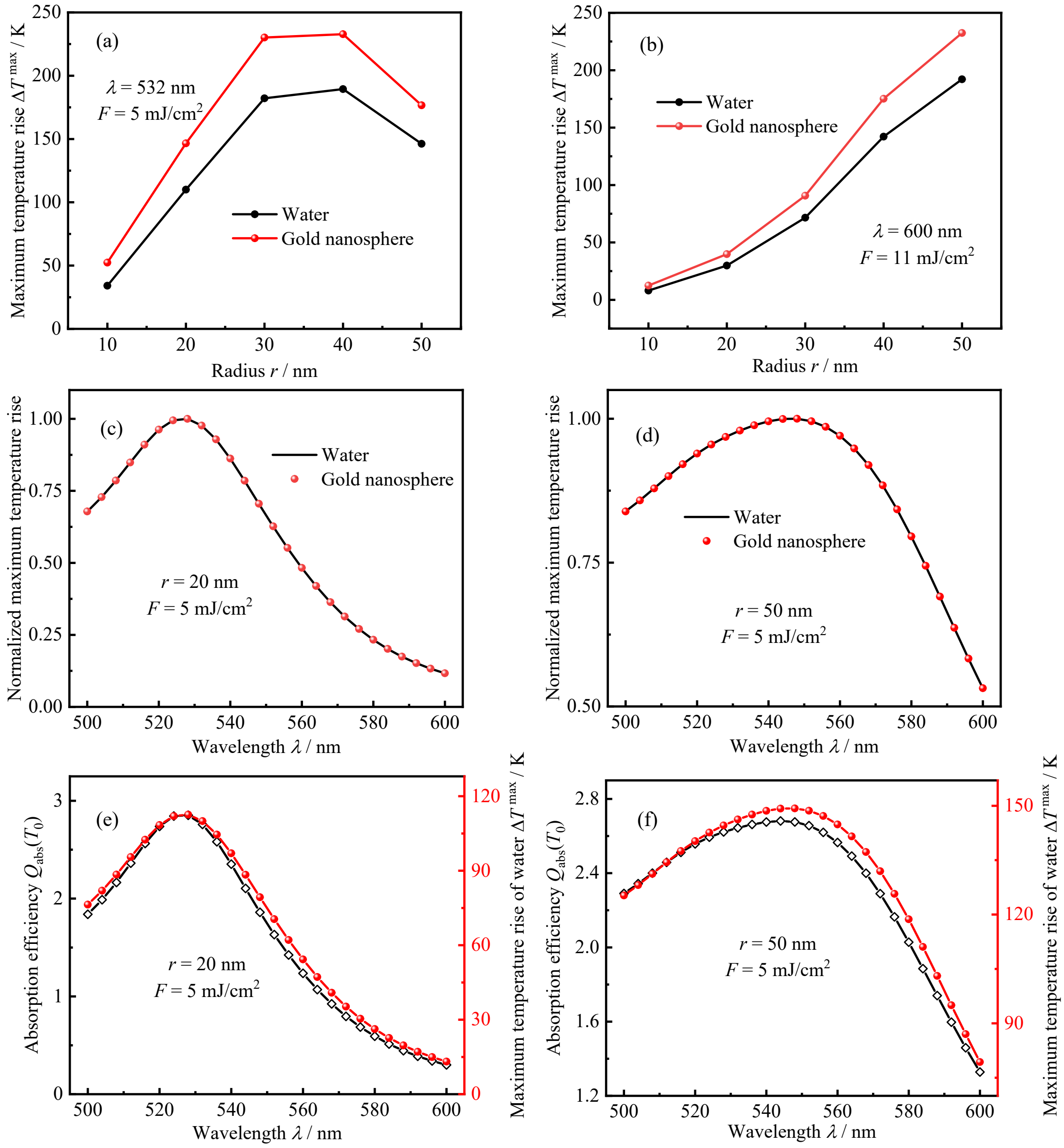

Fig. S4 The relationship between the maximum temperature rise of gold nanospheres and the maximum temperature rise of the surrounding water considering the temperature dependence of the dielectric constant: (a) and (b) are the contrast of the two maximum temperature rise of gold nanospheres of different radii at 532 nm and 600 nm, respectively; (c)-(f) show the agreement between the absorption efficiency $Q_{abs}(T_0)$, the maximum temperature rise of water and the maximum temperature rise of gold nanospheres.

Reference

1. Thormahlen, I.; Straub, J.; Grigull, U., Refractive Index of Water and Its Dependence on Wavelength, Temperature, and Density. *J. Phys. Chem. Ref. Data* **1985**, *14*, 933-946.

2. Metwally, K.; Mensah, S.; Baffou, G., Fluence Threshold for Photothermal Bubble Generation Using Plasmonic Nanoparticles. *J. Phys. Chem. C* **2015**, *119*, 28586-28596.