"Optimal detection angle in sub-diffraction resolution photothermal microscopy: application for high sensitivity imaging of biological tissues": reply

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Abstract: The differences between our model and existing models are rationalized in terms of the experimental conditions. The theory in [Opt. Express 22(16), 18833-18842 (2014)] is applicable when the temperature increase is moderate (~1 *K*) and the spatial extend of refractive index being modulated is comparable to or smaller than the wavelength, which are in accordance with our experiment.

OCIS codes: (170.5810) Scanning microscopy; (350.5340) Photothermal effects; (100.6640) Superresolution; (290.5825) Scattering theory.

References and links

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We are pleased that our previous work has aroused interest as well as debate [1,2]. Here is a reply and rebuttal.

#234983 - $15.00 USD
Received 18 Feb 2015; accepted 19 Feb 2015; published 3 Mar 2015

Reply and rebuttal

1. Regarding the comment "Applicability of previous models to finite modulation frequency"

We apologize for the misleading sentences described in Section 2. The correct expression is "However, their model is based on a steady-state calculation and is not applied yet to the case when (…)".

2. Regarding the comments "A signal where there should be none", "the scattering amplitude", and "the optical resolution"

Our model is based on the scattering theory with a Yukawa type potential under the Born approximation on an assumption that scattering amplitude is sufficiently small. To be more specific, the Born approximation requires [3]

$$(\Delta n / n_0) r_c^2 k^2 << 1, \tag{1}$$

where $\Delta n/n_0$ is the relative change of the refractive index around a nanoparticle, r_c is the spatial extent of the refractive index being modulated, and k is the wavenumber of the probe beam. We consider the case when only the pump beam is absorbed by a point absorber. This inequality is satisfied for a moderate temperature increase $\Delta n/n_0$ ~10^{-4} (generally corresponding to ΔT~1 K in many materials) and when r_c is comparable to or smaller than the focal spot size of the probe beam. In contrast, in the case of slow modulation ($r_\mathrm{c}k$?1) or a large $\Delta n/n_0$, GLMT or scattering theory with Coulomb type potential discussed by Selmke *et al.* is more appropriate [4–7].

This difference in the theoretical discussion is in accordance with the different conditions between our experiment and the experiment by Selmke *et al*. In our experiment, a balanced detection scheme is employed to eliminate the noise of probe beam intensity. This allows us to detect the relative change of transmitted probe intensity Φ as low as ~10^{-5} [8]. In this case, Eq. (1) is satisfied since $\Delta n/n_0$ ~10^{-4} and r_c is estimated to be ~0.2 μm assuming the thermal diffusivity of 1.5 x 10^{-7} m^2/s which is compatible or smaller than the focal spot size (~0.3μm). On the other hand, the relative change is Φ ~10^{-3} in the experiment by Selmke *et al* [4]. We consider that, in their case [4], the $\Delta n/n_0$ is as large as 10^{-2} - 10^{-3} even though the power level of the pump beam is not described. This difference in signal level may be related to the detection sensitivity. In our experimental setup, the noise level of the LD without balanced detection is ~5 x 10^{-5} when the time constant of lock-in amplifier is ~1 ms, which is comparable to the signal intensity resulting in the S/N level of 1. Balanced detection scheme is capable of reducing the intensity noise of a LD by 20-30 dB, and we successfully realized high-sensitivity imaging with close to the shot noise limit [8,9].

Regarding to the beam offset in the axial direction, we like to refer that it is difficult to adjust the two laser beams in such a way that they focus exactly at the same position in a sample due to several factors such as the chromatic and spherical aberrations of the objective lens. Therefore, it is hard to conclude whether signal appears or not under the condition of both of the two beams (pump and probe) are exactly focused at the position of nanoparticle. As was discussed by Selmke et al. their paper [4] showing in Fig. 2(e), there is some deviation of zero signal position in the axial direction because of local refractive index deviation from ideal one or due to aberration. It is difficult to determine the absolute position. Since relative position is practically more important in optical imaging than the absolute distance of the imaging point from for example from the surface of the sample. We consider that when r_c and/or $\Delta n/n_0$ are sufficiently large, PT signal from the area out of focus dominates and thus exhibits lens-like behavior as Selmke et al discussed. While when Eq. (1) is satisfied, signal from a focal point becomes non-negligible. In the case of small r_c and/or $\Delta n/n_0$, the point spread function in lateral plane is given by the product of the intensity profile of the pump and probe beam, and hence the spatial resolution is improved. In the case of large r_c and/or $\Delta n/n_0$, resolution is an interesting and important subject to be solved.

We appreciate Selmke *et al.* for pointing out the missing a factor $-2k^2$ in Eq. (6).

3. Regarding the comment "plane-wave spectrum Eq. (12)"

We appreciate Selmke *et al.* for pointing out the error in Eq. (12). The plane-wave decomposition of a focusing coherent field is based on geometrical optics [10] and the denominator $|\mathbf{r}-\mathbf{r}_0|$ should be deleted. In experiment, to make use of the full NA of the objective lens, the Gaussian pump and probe beams overfill the back aperture of the objective lens with the filling factor of 1.7. Thus, the amplitude factor is assumed to be uniform for simplicity. The difference between the two calculated results with and without the assumption is smaller than 5%. Therefore the assumption of uniform intensity distribution can be well rationalized.

4. Regarding the comment "the angle θ"

The reorientation of the solution is considered in the same manner as in [6,7].

#234983 - $15.00 USD Received 18 Feb 2015; accepted 19 Feb 2015; published 3 Mar 2015