# Optically Trapped Gold Nanoparticle Enables Listening at the Microscale

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We explore a new application of optical tweezers for ultrasensitive detection of sound waves in liquid media. Position tracking of a single gold nanoparticle confined in a three-dimensional optical trap is used to readout acoustic vibrations at a sound power level down to −60 dB, causing a ∼90 $\mu$eV increase in kinetic energy of the nanoparticle. The unprecedented sensitivity of such a nanoear is achieved by processing the nanoparticle's motion in the frequency domain. The concept developed here will enable us to access the interior of biological microorganisms and micromechanical machines not accessible by other microscopy types.

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The contactless nature of optical forces exerted by light on material objects enabled optical tweezers [1] to become a standard tool widely used in biology, physics, chemistry, and material sciences. There are three important classes of applications of optical tweezers: (i) noninvasively manipulating micro- and nano-objects in space and arranging them on substrates; [2] (ii) applying precisely adjustable forces [3] and torques; [4], and (iii) measuring forces at the scale of a single molecule. [5] In this Letter, we expand the list of optical tweezers' applications by exploring and demonstrating an ultrasensitive sound detection scheme based on motion analysis of a three-dimensionally optically trapped nanoparticle.

A small particle immersed in a liquid continuously experiences stochastic collisions with solvent molecules resulting in a random drifting of the particle, known as Brownian motion. Upon interaction with a tightly focused laser beam the particle can be trapped or confined in three dimensions within a certain volume [1]. Effectively, the light intensity gradient creates a three-dimensional potential well restricting particle's motion roughly to the focal volume of the focused beam. The particle diffusion is restricted, but not completely stopped. By analyzing this confined motion, some parameters of the trapping potential well can be deduced, offering a fairly simple approach to calibrating optical tweezers [6]. Essentially, this widely used calibration approach uses stochastically fluctuating solvent molecules in thermodynamic equilibrium to move a particle at the bottom of the trapping potential well, probing the characteristics of the well in an elegant way. Now we pose a question, whether and at which sensitivity an optically trapped particle can be used to readout nonequilibrium fluctuations of the particle's environment, e.g., acoustic waves or local solvent flows.

To address these questions we set up an experiment in an aqueous medium schematically illustrated in Fig. 1(a) and 1(c). The experiment consists of two essential parts: a three-dimensionally optically trapped gold nanoparticle, we name it a *nanoear*, and a sound source. The latter is realized either macroscopically or microscopically. The macroscopic sound source is a tungsten needle glued on a loudspeaker [Fig. 1(a)] and immersed into a water drop, which is placed between the microscope objective and the substrate. The microscopic sound source is mimicked by an aggregate of gold nanoparticles [Fig. 1(c)]. The aggregate is plasmonically heated by an intensity-modulated laser beam to generate acoustic waves due to the photoacoustic effect [7], which is widely exploited in photoacoustic spectroscopy [8].

The optical setup used in these experiments is schematically shown in Fig. 1(b) and is effectively the same as we used in our previous optical trapping studies [9]. In short, the setup is based on an upright dark-field microscope (Zeiss Axiotech 100) equipped with a Ti:sapphire laser (Tsunami) operating in continuous wave mode at 808 nm to trap individual gold nanoparticles and a frequency-doubled Nd:YAG laser (Millenia Vs, 532 nm, Spectra-Physics) to optothermally generate an acoustic wave. The trapping laser beam is expanded by a homebuilt telescope to slightly overfill the back aperture of a water-immersion microscope objective (Zeiss Achroplan 100x/1.0/Ph3). Its focal plane is set 3.5 $\mu$m above the substrate. The heating laser is modulated by a chopper in the frequency range of 10–20 Hz with a duty cycle of 50%. Stacked notch filters (Semrock, StopLine 808 and 532 nm) are used to filter out scattering from both lasers. A $XYZ$ translational stage (Linos) is used to move a substrate with respect to the objective. Images are recorded with a digital camera (Canon EOS 500D) at 50 frames per second.

In order to justify experimentally that an optically trapped nanoparticle can be used to optically readout acoustic waves, we perform experiments using sound waves generated by the macroscopic sound source. The loudspeaker the tungsten needle is glued onto is driven at a frequency of 300 Hz. After one of the freely diffusing gold nanoparticles (60 nm, citrate-stabilized, BBInternational,

 

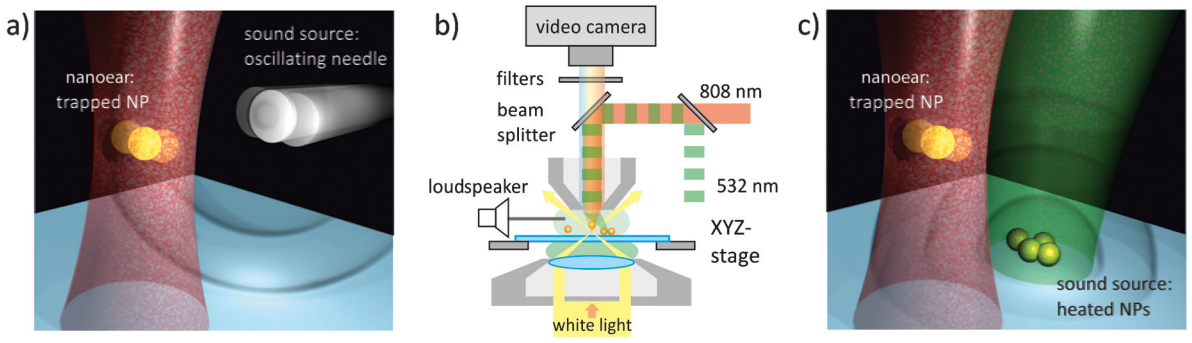


FIG. 1 (color). (a) Schematic of the listening to a macroscopic sound source with a nanoear—an optically trapped gold nanoparticle. (b) Sketch of the experimental setup used for the listening experiment, which includes simultaneous three-dimensional optical trapping, optothermal and mechanical sound generation, and imaging of trapped gold nanoparticles. (c) Schematic of the listening to optothermally generated sound.

$1.3 \times 10^7$ ml$^{-1}$) is trapped, the motion of the trapped nanoparticle is recorded at dark-field illumination with the video camera with sound switched off and on.

Videos of trapped nanoparticles are recorded for 30 s yielding 1500 video frames. A typical video frame of a trapped single 60 nm gold nanoparticle is shown in Fig. 2(a). Nanoparticle tracking is performed by localizing the nanoparticle at each individual frame using a two-dimensional Gaussian fit to the recorded diffraction-limited intensity patterns. Figure 2(b) displays the particle positions obtained in a typical measurement with no sound generated. It is worth noting that the position of a single

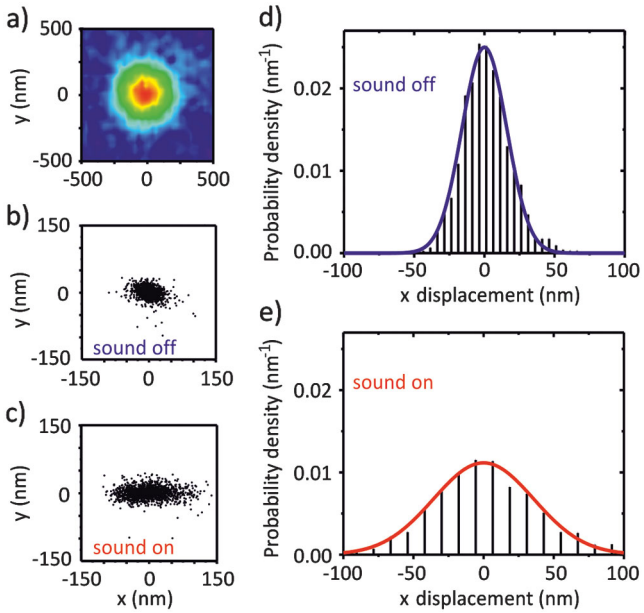


FIG. 2 (color). (a) False-color plot of a trapped gold nanoparticle as detected by the digital camera. (b)–(c) Results of particle tracking over 1500 frames with sound off and on, respectively. (d)–(e) Probability density distribution of particle positions fitted with a Gaussian function with sound off and on, respectively.

emitter or scatterer can be determined with an accuracy well beyond the diffraction limit if a sufficient number of photons is collected. [10] In our experiments we estimated the localization accuracy to be better than 1.5 nm. Figure 2(d) shows the probability distribution of the particle displacement inside the trap which is of Gaussian shape. The Gaussian shape of the probability distribution originates from the harmonicity of the optical trapping potential $U(x) = x^2\kappa/2$. Here, $\kappa$ is a spring constant, referred to as "trap stiffness" [11]. When the sound is switched on, an oscillating water motion is generated at the spatial position of the trapped nanoparticle, causing an additional driving force being exerted on the nanoparticle. The motion of an acoustically driven trapped nanoparticle is clearly seen in Fig. 2(c) and 2(e) as the elongation of the nanoparticle's position distribution in the direction of sound propagation.

In the experiment described above we have demonstrated that an optically trapped gold nanoparticle can be used to optically readout acoustic waves generated in liquid media. State-of-the-art piezocomposite transducers used in photoacoustic microscopy are extremely sensitive and would certainly also enable detection of the macroscopically generated sound waves [12]. The reasonable question arises, how sensitive such an optical readout by means of the trapped nanoparticle can be. To address the sensitivity aspect we have performed the experiment using the microscopic sound source as it is illustrated in Fig. 1(c). The experiment is performed in two steps. First, aggregates of gold nanoparticles are printed on a glass substrate from a concentrated colloidal suspension ($2.6 \times 10^9$ ml$^{-1}$) as it is reported elsewhere [13] using the unmodulated green heating laser. These aggregates are later used for optothermal sound generation. Then the substrate is rinsed with pure water reducing the particle concentration significantly to $1.3 \times 10^7$ ml$^{-1}$. The substrate is aligned so that one of the deposited aggregates is hit by the heating laser. The trapping laser focal plane is again set 3.5 $\mu$m above the substrate. After one of the freely diffusing gold nanoparticles is

trapped, the motion of the trapped nanoparticle is recorded at dark-field illumination with the video camera in three configurations: (i) heating laser on, (ii) heating laser off, and (iii) heating laser on, but focused on a bare substrate area with no gold aggregate. The latter two are used to exclude systematic errors and a direct influence of the heating laser on the trapped nanoparticle. A trapping laser power of 50 mW in the objective focal plane was sufficient to trap individual gold nanoparticles for more than 5 minutes. The heating laser power was set to 30 mW in the objective focal plane. The laser beams are adjusted to be focused by the objective lens laterally 11 $\mu$m away from each other in the focal plane.

The trapped nanoparticle motion is tracked and analyzed as explained above, yielding the position ($x$, $y$ displacement) of the nanoparticle recorded 50 times per second. The $x$ displacement of the particle over time with the heating laser turned off is shown in Fig. 3(a), illustrating random fluctuations of the nanoparticle position in time around its equilibrium point. In the harmonic approximation of the potential of the optical trap, the distribution of the particle position $P(x)$ can be fitted with the Gaussian function $P(x) = P_0 e^{-x^2/2\sigma^2}$ [black curve in inset in Fig. 3(b)]. From the width $\sigma$ of this distribution, we obtain the trapping stiffness $\kappa = k_B T/\sigma^2$ [6]. At typical

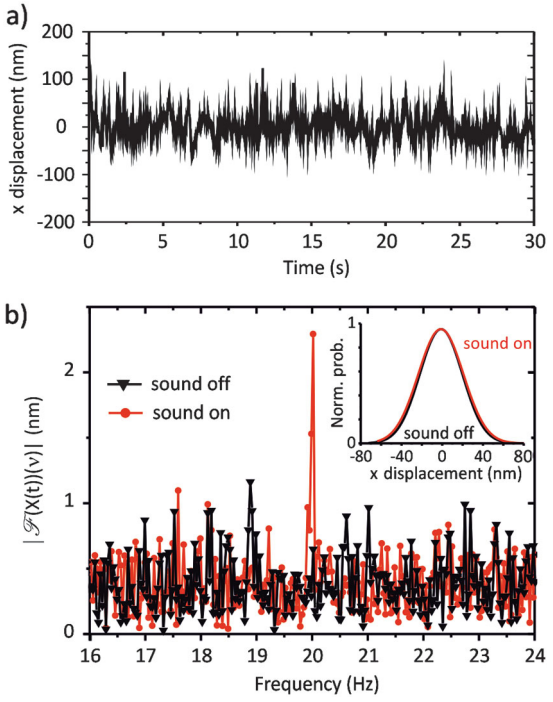


FIG. 3 (color). (a) Trajectory of the $x$ displacement of a tracked trapped particle. The $x$ coordinate in the analysis is chosen to be along a line connecting the sound source and the trapped nanoparticle. (b) Fourier transformation of the trajectory of a particle with sound switched off (black) and on (red). The inset shows the respective probability distributions of the particle displacement.

experimental settings we calculated $\kappa_{\text{typ.}} = (170 \pm 6)$ aN/nm/mW.

When the chopped heating laser focused on an aggregate of gold nanoparticles is turned on, water flows oscillating in time around the heated aggregate are induced [14]. The periodically modulated micro flows of liquid close to the aggregate result in an optically generated acoustic wave. Acoustic waves generated by the optical heating of a nanoparticle aggregate are significantly weaker compared to those produced with the macroscopic needle. In all the experiments with the optothermally generated acoustic waves, neither the trajectory nor the probability distribution of the displacement change significantly upon activation of the sound source [inset in Fig. 3(b)]. To probe the sensitivity limits of the optical readout, the discrete fast Fourier transform algorithm [15] implemented in IGOR PRO is applied to the measured trajectories. Further analysis is performed in frequency domain. In frequency domain the trajectories recorded with no sound, i.e., heating laser off, transform into flat, frequency independent spectra, revealing that all possible frequencies are present in the motion of a Brownian particle [16]. When the sound is switched on, a clearly recognizable single peak at the frequency of the sound source (20 Hz) appears superimposed with the spectrum of Brownian motion as it is shown in Fig. 3(b). Note, although the sound is clearly "seen" in the frequency spectrum, the width of the particle displacement probability density distribution [inset Fig. 3(b)] is increased insignificantly, within the experimental error margins, from $\sigma_{\text{off}} = (21.5 \pm 0.4)$ to $\sigma_{\text{on}} = (22.1 \pm 0.5)$ nm. Indeed, the probability density distribution is formed by a sum of amplitudes of all nanoparticle oscillation frequencies, while in frequency domain the amplitudes corresponding to different frequencies are split, enabling extremely high readout sensitivity at given frequencies.

To estimate the sound detection sensitivity we recall that the magnitude of a Fourier spectrum corresponds to the amplitude of a harmonic oscillation at a given frequency. In the experiment illustrated in Fig. 3 the height of the peak at 20 Hz above the constant contribution of the Brownian motion yields the amplitude $A = 1.85$ nm of the additional motion introduced by the optically driven sound source. Inside of the parabolic potential of an optical trap with stiffness $\kappa$, the energy $E(x) = x^2\kappa/2$ is required to displace the particle by a distance $x$. Thus, using the width $\sigma$ of the particle's displacement probability density distribution and the above formula for $\kappa$, we find the acoustic energy transferred to the particle as $E_{\text{transferred}} = 1/2(A/\sigma)^2 k_B T \approx 90$ $\mu$eV. The average power of a harmonic acoustic wave incident onto the nanoparticle's geometrical cross section is given by $P_{\text{avg}} = 1/2 c\rho v_{\text{max}}^2 \pi r^2$ [11], where $c$ is the phase velocity of sound, $\rho$ the density of water, $v_{\text{max}}$ the maximum velocity of the water molecules, and $r$ the particle radius. Assuming a laminar flow of liquid with viscosity $\eta$ around the nanoparticle, the Stokes

drag force is exerted on the nanoparticle, which is compensated at a certain maximum displacement $x_{\max}$ by the restoring force of the optical trap: $\kappa x_{\max} = 6\pi\eta r v_{\max}$. Combining last two expressions, the average acoustic power detected by the nanoear is $P_{\text{detected}} = \frac{1}{2\pi} c\rho(\frac{\kappa x_{\max}}{6\eta})^2$. This yields a detected sound power level of $L_W = -58$ dB. With these experimental settings we could confidently detect sounds with a sound power level down to $-60$ dB. For comparison, an empty room has a sound power level $L_{\text{emp.}} = 20$ dB and at very specific conditions humans are able to hear down to power levels of $L_{\text{hear}} = 0$ dB [17]. This means that a nanoear based on a single optically trapped gold nanoparticle serves as a sound detector with an acoustic power sensitivity 6 orders of magnitude higher than the human ear.

It is worth noting that, in principle, the vibrations seen in the motion of trapped nanoparticles could be caused not only by acoustic oscillations in the water drop, but also by additional heating of the trapped nanoparticle due to the heat transfer from the aggregate or by the radiation pressure forces exerted on the trapped nanoparticle by light scattered by the aggregate. To estimate the influence of both these effects we have calculated the temperature rise at the position of the trapped nanoparticle ($10^{-20}$ K) and the optical force exerted on the trapped nanoparticle (0.04 fN) in case the entire energy of the heating beam is scattered homogeneously in all directions. This optical force would result in a deflection of the trapped nanoparticle from its equilibrium position of 0.005 nm. Therefore, we conclude that the oscillating motion of the nanoparticle in this experiment is caused solely by sound waves.

To make sure that the optical listening method explored here is not specific to the chosen sound source, namely, optothermally heated aggregates of gold nanoparticles, we have performed a set of additional listening tests. In these experiments thin gold stripes (thickness 75 nm) are deposited on a glass substrate. The heating laser is focused on the stripe edge to generate acoustic waves and a gold nanoparticle is optically trapped as it is done in the previous experiment. As a result we have observed qualitatively the same behavior: acoustic vibration in water can be read out by tracking the motion of a trapped nanoparticle and analyzing it in the frequency domain. Additionally, the heating laser modulation frequency was varied revealing that the peak position precisely follows the sound frequency used in the experiment (Fig. 4).

It is worth noting that we find the peak in the frequency spectra manifesting a sound source only in the direction of the line connecting the sound source and the trapped nanoparticle. The motion of the nanoparticle in the direction orthogonal to this connecting line is found not to be affected by the sound. This experimental observation is easy to understand taking into account the longitudinal nature of acoustic waves, resulting in the liquid oscillation only in propagation direction. This natural anisotropy of

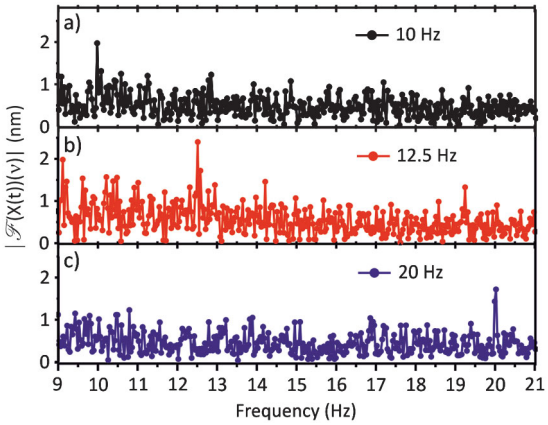


FIG. 4 (color). Fourier transformations of the trajectories of an optically trapped nanoparticle with sound switched on at frequencies (a) 10 Hz, (b) 12.5 Hz, and (c) 20 Hz. The heating laser modulation frequencies are kept below the sampling rate/2 = 25 Hz to fulfill requirements of the Nyquist—Shannon sampling theorem [23].

the nanoparticle's response to acoustic waves enables not only listening to ultraquiet sound sources, but also accurate determination of the direction towards the source of the acoustic waves. Recent advantages in designing multiple optical traps by means of scanning galvo mirrors [18], acousto-optic beam deflectors [19], or spatial light modulators [20] enable simultaneous three-dimensional trapping of multiple particles localized at different positions. Using a fast video camera ($> 100,000$ fps are available), motion of multiple optically trapped nanoparticles can be recorded and analyzed in real time [21], offering all necessary information for localization of sound sources be means of triangulation, similar to the principle used in the global positioning system [22].

In conclusion, we have performed proof-of-concept experiments demonstrating how a single optically trapped gold nanoparticle can serve as an ultrasensitive nanoear capable of detecting acoustic vibrations in aqueous media with the sound power level down to $-60$ dB. We have introduced a method based on Fourier transformation for analyzing the motion of trapped nanoparticles. This method allowed us to optically readout an acoustically induced vibration of the nanoparticle with an energy of $\sim 90$ $\mu$eV, which is only 0.1% of a nanoparticle's thermal energy. The concept introduced in this Letter opens new ways of obtaining useful information on live (bacteria, cells, or viruses) and artificial micro-objects that produce acoustic vibrations, but cannot be directly visualized in an optical microscope, i.e., due to strong absorption or scattering of light. Furthermore, a new type of acoustic microscopy with optical readout could be built based on a set of nanoears distributed in a sample, "listening" to acoustic signals and collecting amplitudes, phases, and propagation directions of acoustic waves to gain insight into the sources of these signals.

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