This is the published version of:


Access to the published version:
http://dx.doi.org/10.1364/OL.34.003740

Copyright:

This paper was published in Optics Letters and is made available as an electronic reprint with the permission of OSA. The paper can be found at the following URL on the OSA website:
http://www.opticsinfobase.org/abstract.cfm?URI=ol-34-23-3740. Systematic or multiple reproduction or distribution to multiple locations via electronic or other means is prohibited and is subject to penalties under law.
Vibrations of microspheres probed with ultrashort optical pulses

T. Dehoux,1 T. A. Kelf,1 M. Tomoda,1 O. Matsuda,1 O. B. Wright,1,2 K. Ueno,2 Y. Nishijima,2 S. Juodkazis,2 H. Misawa,2 Y. Tournat,3 and V. E. Gusev4

1Department of Applied Physics, Faculty of Engineering, Hokkaido University, Sapporo 060-8628, Japan
2Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0021, Japan
3LAUM, CNRS, Université du Maine, F-72085 Le Mans, France
4Laboratoire de Physique de l’Etat Condensé, UMR CNRS 6087, Le Mans 72085, France

Received July 30, 2009; accepted October 14, 2009; posted November 9, 2009 (Doc. ID 114981); published November 30, 2009

We use ultrashort optical pulses to excite and detect vibrations of single silica spheres with a diameter of 5 μm placed at the surface of an acoustically mismatched substrate. In addition to the photoelastic detection of picosecond longitudinal acoustic pulses propagating inside the bulk, we detect gigahertz acoustic resonances of the sphere through probe beam defocusing. The mode frequencies are in close accord with those calculated from the elastic vibrations of a free sphere. We also record a resonant enhancement in the amplitude of specific modes of two touching spheres. © 2009 Optical Society of America

OCIS codes: 320.5390, 110.5125, 110.7170.

Understanding vibrations in microscale systems is important given the trends for the miniaturization of mechanical resonators and actuators, with applications in filtering and mass sensing [1,2]. Having typical resonant frequencies in the gigahertz range, the vibrations of microstructures can be conveniently probed in the time domain using ultrashort optical pulses for noncontact excitation and detection [3,4]. These experiments have been carried out in one- and two-dimensional periodic microstructures. Single microstructures such as isolated spheres [5], disks [1], or cantilevers [2] are of particular interest for sensing applications, because their vibrational spectra are well known. Experiments on such structures have generally been conducted in the frequency domain. It is, however, very useful to carry out measurements in the time domain, because phase information is easier to access and frequency domain data are available by Fourier transforming. Despite the attention paid to the vibrations of uniform isotropic microspheres—a useful model system for vibrational analysis—from centimeter down to nanometer dimensions [5–10], there have been no time domain studies of the vibrations of isolated spheres with a diameter of ~1 μm. In this Letter we present such measurements on single microscopic silica spheres using ultrashort laser pulse excitation and detection. We also examine a system of two spheres in contact.

The samples are prepared by exploiting the adhesion of silica spheres (median 2) of diameter $D = 5\pm0.05$ μm to a polydimethylsiloxane (PDMS, medium 3) coated silicon substrate. During the contacting process, the spheres penetrate the PDMS to a depth of $\delta = 0.34$ μm, measured by scanning electron microscopy (SEM). A single-sphere site and a SEM image are shown in Fig. 1(a). The small $\delta$, combined with the large mismatch between the acoustic impedances of the PDMS [11] $Z_3 = \rho_3 v_{L3} = 1.1$ MPa m s$^{-1}$ and that of silica [12] $Z_2 = \rho_2 v_{L2} = 13$ MPa m s$^{-1}$ (longitudinal velocities $v_{L3} = 1.0$ and $v_{L2} = 6.0$ km s$^{-1}$; densities $\rho_2 = 1.1$ and $\rho_3 = 2.2$ g cm$^{-3}$), provides vibrational Q factors $\gg 10$. ($Q = 11$ is found for a Si particle in water; see [9].) The low Young’s modulus of the PDMS, ~0.7 MPa [13], ensures <1% shift from the free-sphere resonance frequencies [7]. Finally a metal film (medium 1, 80% Pt, 20% Pd) with a thickness of ~160 nm is sputtered on the sample for photoelastic transduction. We determine the stiffness coefficients and the density $\rho_1 = 19.6$ g cm$^{-3}$ of the layer with an effective medium theory [14]. Using the values for Pt and Pd in [12], the effective stiffness coefficients are $C_{11} = 345$, $C_{12} = 258$, and $C_{44} = 75.5$ GPa. We calculate the longitudinal sound velocity $v_{L1} = 4.3$ km s$^{-1}$ using Voigt averaging.

We use the optical pump-probe technique shown schematically in Fig. 1(a). Pump pulses from a Ti:Sapphire mode-locked laser with a wavelength of 810 nm, a pulse duration of 200 fs, and a repetition frequency of 80 MHz excite acoustic vibrations. This light is chopped at 1.1 MHz for lock-in detection. Each pump pulse, of energy $E = 25$ pJ, is initially absorbed in the metal layer over a depth comparable to the optical skin depth of 13 nm. Conduction band electrons are excited and diffuse over a depth of $(k/L)^{1/2} = 8$ nm during their thermalization with the lattice, where $k = 72$ W m$^{-1}$ K$^{-1}$ is the thermal con-
ductivity [12] and \( g = 11 \times 10^{17} \, \text{W m}^{-3} \, \text{K}^{-1} \) is the electron-phonon coupling constant in Pt [15]. The subsequent laser-induced heating yields a transient temperature rise of \( \sim 90 \, \text{K} \), producing a thermoelastic expansion that couples to longitudinal acoustic pulses and sphere vibrations, and a maximum steady state temperature rise of \( \sim -2 \, \text{K} \) (avoiding damage to the PDMS substrate). The reflectivity change \( \delta R(t) \) is measured with frequency-doubled circularly polarized probe pulses with a wavelength of 405 nm and energy of 3 pJ as a function of the pump-probe time delay \( t \) using a balanced photodiode. (The unmodulated acoustic oscillations excited by the probe pulse are not detected by the lock-in detection system.) The delay is achieved by multiple passes through a motorized mechanical delay line. The coaxial pump and probe beams are focused onto the top of a sphere at normal incidence through a 100\( \times \) objective lens with an NA of 0.8 to Gaussian spots of diameters \( w_1 = 1 \, \mu\text{m} \) and \( w_2 = 0.5 \, \mu\text{m} \) FWHM, respectively. Assuming that the incident probe beam waist is exactly at the surface of the sphere, the reflected Gaussian probe beam has a radius of curvature of \( D/4 \) at this position. We measured a 60\% clipping of this divergent reflected beam by the finite apertures of the objective.

We plot the amplitude \( \delta R(t)/R_0 \) in Fig. 2(a) for two single-sphere sites, where \( R_0 = 0.6 \) is the probe reflectivity, demonstrating a reproducible response. Ultrafast electron diffusion produces a sharp spike at \( t = 0 \). A longitudinal acoustic strain pulse of center frequency \( f_0 = 35 \, \text{GHz} \) propagates in the metal film and reflects off the film/sphere interface. This pulse produces a first series of echoes [see the inset of Fig. 2(a)]. The amplitude of successive echoes, of interval \( \Delta t_1 = 76 \, \text{ps} \), decreases in time owing to attenuation in the film and strain transmission to the sphere with a coefficient of \( 2Z_0/\left(\rho_1 v_{L1} + Z_2\right) = 0.28 \). We determine the film thickness from \( v_{L1} \Delta t_1/2 = 160 \, \text{nm} \) (ignoring small corrections from Gouy phase shifts). The tri-polar shape of the echoes is characteristic of the strain detection through the photoelastic effect in the metal film [16]. The transmitted strain pulse reflects off the SiO\(_2\)/PDMS interface and propagates back to the metal layer, where it produces a second series of echoes [downward arrow in Fig. 2(a)]. The interval between the two series of echoes, \( \Delta t_2 = 1.88 \, \text{ns} \), yields a reasonable value for the longitudinal sound velocity in fused silica, \( v_{L2} = 2D/\Delta t_2 = 5.3 \pm 0.3 \, \text{km/s} \) [12].

\( \delta R(t) \) shows oscillations from the sphere vibration, superimposed on a slow thermal relaxation. These oscillations are long-lived as is evident from their presence at \( t < 0 \) owing to previous pump pulses. We plot in Fig. 2(b) the amplitudes and phases from temporal Fourier analysis, with the 11 ns window giving a 94 MHz frequency resolution. Six resonances [see arrows in Fig. 2(b)] are detected at frequencies of \( f_1 = 0.57, f_2 = 0.73, f_3 = 0.93, f_4 = 1.12, f_5 = 1.30, \) and \( f_6 = 1.58 \, \text{GHz} \). Reproducible phase data are obtained as shown in the inset of Fig. 2(b). The phase leads of the resonances \( f_1 \) and \( f_4 \) are \((-100 \pm 25)^\circ \) and \((-155 \pm 15)^\circ \) with respect to a cosine, respectively.

To interpret these results, consider the vibration of a free homogeneous isotropic sphere [17]. Owing to the axial symmetry of the excitation, only spheroidal modes of azimuthal mode number \( m = 0 \) are generated. Modes associated with bodily motion of the whole sphere such as bouncing are too low in frequency to be detected with the present set-up [18]. Using the classical elastic dispersion equation for spheroidal modes [19], we determine the angular frequencies \( \omega_{nl}^0 \), with \( n \) and \( l \) being the radial and angular mode numbers, respectively. Knowledge of the longitudinal and shear sound velocities \( v_{L2} \) and \( v_{T2} \) in silica is not required for comparison with the data. It is sufficient to use \( D/v_{L2} = \Delta t_2/2 \) and \( D/v_{T2} = [(1 - 2\nu)/(1 - \nu)]^{1/2} D/v_{L2} \), where \( \nu = 0.17 \) is the Poisson’s ratio in silica [12]. We plot \( \omega_{nl}^0 D/2v_{T2} \) versus \( l \) in Fig. 1(b) for both theory and experiment. As \( m = 0, l \) defines the number of nodes on the surface of the sphere along a fixed-azimuth-\( \phi \) perimeter \( nD \). One can define the acoustic wavelength to be \( \lambda = \pi D/l \) in the high frequency limit and consider \( \omega_{nl}^0 D/2v_{T2} \) versus \( l \) as the dispersion curve for surface waves. Frequencies associated with Rayleigh or whispering gallery modes (WGs) are connected with dotted lines [20].

Assuming that \( \delta R \) arises solely from the photoelastic effect, the results of Fig. 2(a) imply that the amplitudes \( \eta_1 \) and \( \eta_2 \) of the longitudinal strain in the in-depth direction due to the acoustic pulse of center frequency \( f_0 \) and to the sphere vibration, respectively, are of similar order: \( \eta_2/\eta_1 \approx 1 \). The dispersion curves indicate that the measured vibrational frequencies can only correspond to modes \( n \leq 2 \) for which the surface displacement is \( u_2 \sim \eta_2 D \). (The radial displacement varies on a length scale \( \sim D \) for sufficiently small \( l \) mode numbers; see [20].) So the ratio of the surface displacement \( u_2 \) to that due to the acoustic

![Fig. 2.](image-url)
pulse $u_1 \sim \eta L / 2f_p$ is $u_2 / u_1 \sim 2D f_p / v_{L1} \sim 10^2$. This high ratio suggests that a nonnegligible surface displacement should contribute to $\Delta R$ from probe beam defocusing [21]. Even though we did not introduce an iris in our setup to maximize this effect, the finite apertures in the 100× objective act as one. If we assume that the lowest-order modes $n=0$ yield the highest surface displacement, they are likely to produce the highest amplitudes in $\Delta R$. So we identify the measured frequencies $f_1-f_0$ as the following $(n,l)$ modes: $(0,2)$, $(0,3)$, $(0,4)$, $(0,5)$, and $(0,6)$. We compare the measured normalized frequencies with the theory in Fig. 1(b). The good agreement suggests that we are indeed detecting $n=0$ modes. The acoustic wavelength $\lambda \lesssim 2w_1$ is limited by the pump spot di- ameter $w_1$, so only modes with $l \leq \pi D / 2w_1 < 8$ are detected.

As a further experiment we pumped and probed on the top of one of the spheres of a contacting pair, $\Delta R(t)/R_0$ and the corresponding Fourier spectra for two different such sites are shown in Figs. 2(c) and 2(d), respectively. The amplitudes of modes $(0,2)$ and $(0,4)$ (of frequencies $f_1$ and $f_2$, respectively) are particularly enhanced compared to the single-sphere case, and their phase leads become $-(75\pm25)^{\circ}$ and $-(110\pm5)^{\circ}$, respectively.

To elucidate this behavior, we solve the equation of motion for a free isotropic sphere and calculate the radial displacement $u_{nl}^r$ of each mode $(n,l)$ in spherical coordinates $(r,\theta,\phi)$,

$$u_{nl}^r(r, \theta, \phi) = A_{nl} \left[ j_l(\xi_l^r r) \frac{\partial j_l(\eta_l^r r)}{\partial r} + l(l+1) \frac{j_l(\eta_l^r r)}{r} \right] P_l(\cos \theta),$$

where $\xi_l^r = \omega_{l2}^r / v_{L2}$ and $\eta_l^r = \omega_{lT}^r / v_{T2}$ are the longitudinal and transverse wavenumbers, respectively. The spherical Bessel function of the first kind $j_l$ describes the $r$ dependence of $u_{nl}^r$. Each mode $(n,l)$ of amplitude $A_{nl}^r$ produces a radial surface displacement along a fixed-azimuth-$\phi$ perimeter defined by the Legendre polynomial $P_l(\cos \theta)$. We illustrate in Fig. 3 the calculated deformation of the surface of the sphere, $D/2 + \alpha P_l(\cos \theta)$, subjected to a radial displacement of arbitrary amplitude $\alpha$ for mode $(0,4)$. From the definition of $P_l$ we see that for modes of even $l$—notably modes $(0,2)$ and $(0,4)$—the displacement along the equatorial perimeter ($\theta = \pi/2$) is nonzero, and the contact between the two spheres is subject to an oscillating stress that should couple their resonances. A resulting shift in the frequencies [22] of modes $(0,2)$ and $(0,4)$ closer to a harmonic of the laser repetition rate could explain the enhancements observed [23]. A detailed analysis should allow the phase and relative amplitudes of the modes to be extracted.

In conclusion, we have demonstrated for the first time, to our knowledge, that—in spite of the high surface curvature—the acoustic modes of individual micrometer-sized spheres can be efficiently generated and detected using focused ultrashort laser pulses. The measurement of the mode frequencies should prove to be a useful tool for the quality control of the sphere elastic properties. The measured phase of the vibrational modes, when combined with a solution of the elastic wave equation that accounts for the periodic pulsed laser excitation and the spatiotemporal thermoelastic source, should allow the investigation of the vibration generation mechanism and elucidate the dynamics of the mechanical coupling between spheres.

References


Fig. 3. (Color online) Left, radial deformation of the sphere due to mode (0,4). Right, fixed-\phi cross-section (side- view). Dotted line, initial position of the surface.