Oscillating bubbles at the tips of optical fibers in liquid nitrogen

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We report that a bubble with a radius of a few micrometers may be created at a precise location on a metal-coated optical fiber tip immersed in liquid nitrogen by microsecond optical pulses with peak powers of less than 20 mW. Dynamic optical measurements reveal that after termination of the optical pulse the bubble exhibits stable oscillations for several tens of microseconds, at frequencies up to several megahertz, as it slowly collapses.

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It is well established that bubbles can be created through optical breakdown of liquids, including cryogenic liquids, when very intense nanosecond laser pulses are focused into them [1–3]. For example, the generation of bubbles through optical breakdown of liquid nitrogen requires intensities of at least 5×10^{11} W/cm^2. In the course of our research on the nonlinear optical properties of nanoscale metallic films at cryogenic temperatures, we discovered that bubbles could be formed at the ends of optical fibers in liquid nitrogen using very low optical intensities—less than 3×10^4 W/cm^2. The reflectivity of a fiber tip on which a bubble is created shows large-amplitude oscillations, lasting several tens of cycles, with a frequency spectrum extending to several megahertz. The free oscillation of bubbles (especially air bubbles in water [4]) and forced oscillation of acoustically trapped bubbles [5] (including hemispherical bubbles on a solid surface [6]) have been the subject of extensive study, numerical simulation, and theoretical modeling [7]. However, to the best of our knowledge, stable high-frequency oscillations such as we observe have not previously been reported for laser-generated bubbles in liquid nitrogen. The generation of bubbles on the tips of optical fibers using a low-intensity laser source is an elegant method that allows for precise control of the bubble's location and accurate optical interrogation of its dynamics. We believe that our techniques and results may be of interest to those studying the dynamics of bubbles in liquids, in particular those researching the dynamics of bubble formation and collapse [8,9], sonoluminescence [10], and claims of "bubble fusion" [11].

In our experiments, bubbles were generated on the tips of single mode silica optical fibers coated with a semitransparent granular gallium film covered by a protective silicon oxide layer. The metallic films, which had a mass thickness of ~9 nm, were deposited using an atomic beam source onto fiber tips cooled to ~100 K under UHV conditions [12]. Following application of the oxide layer the tips typically had reflectivities of ~20% (in vacuum). When such a fiber is immersed in boiling liquid nitrogen and the tip is subjected to pulsed optical excitation (delivered via the fiber) bubble formation occurs as heat generated by absorption of laser radiation is released from the metallic film. The nitrogen was poured in air and was exposed to the atmosphere during the experiments so it will have contained a considerable concentration of dissolved oxygen. The optical pulses used were produced by a semiconductor diode laser operating at a wavelength of 1.55 μm, were 1 μs long with peak powers up to 17 mW, and had a repetition rate of ~1 kHz. Bubble dynamics were observed by monitoring the reflectivity of the fiber tip (see Fig. 1): the appearance of a bubble changes the medium in contact with the fiber tip from liquid to gas and the associated change in refractive index leads to a change in reflectivity. The magnitude of this change (derived from separate measurements of reflectivity in liquid and gaseous nitrogen atmospheres) is indicated in Fig. 2(a). The bubble's reflectivity dynamics were monitored in real time using a low-power (0.8 mW) cw diode laser light source operating at 1.31 μm and a digital storage oscilloscope [13]. The reflected signal at the 1.31 μm probe wavelength was isolated from reflected 1.55 μm light (from the pulses used to initiate bubble formation) prior to detection using a wavelength division multiplexer and spectral filters. Our experimental conditions are very different from those under which sonoluminescence has been observed in liquid nitrogen [2], and we therefore assume that our detection system does not receive any luminescence signal. The detection system had an overall bandwidth of 125 MHz.

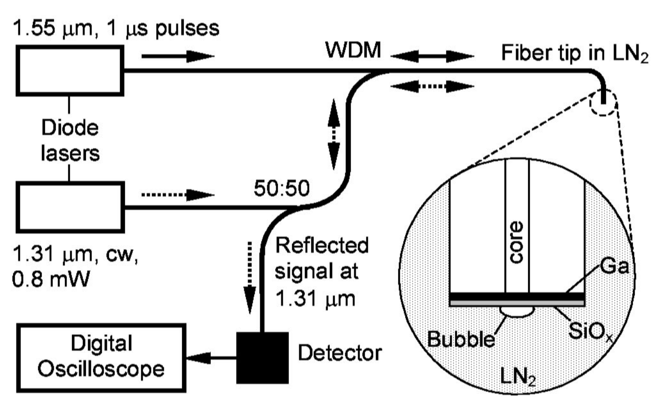

FIG. 1. Schematic of the diode-laser/fiber-optic arrangement for generation and optical study of bubbles in liquid nitrogen, with enlarged detail of the fiber tip.

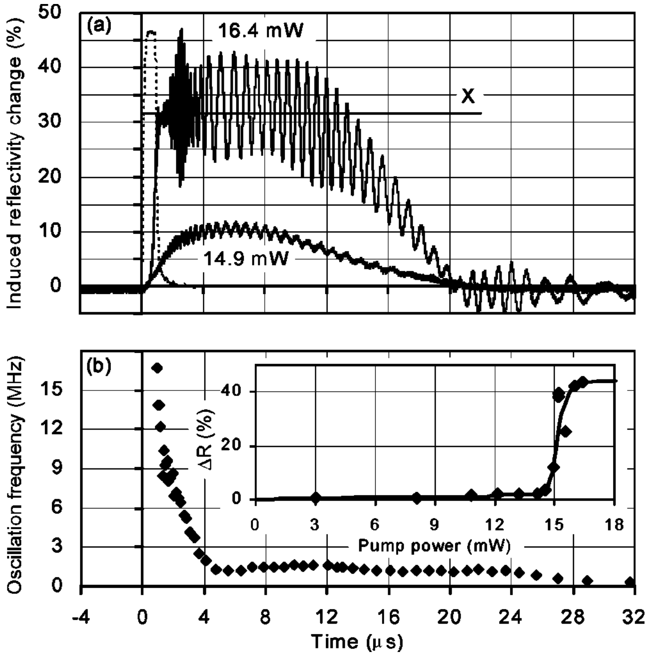

FIG. 2. Reflectivity oscillations resulting from bubble formation and collapse at the tip of an optical fiber immersed in liquid nitrogen. (a) Dynamics of reflectivity oscillations following excitation by 1 μs laser pulses (dashed line) with peak powers of 16.4 and 14.9 mW. The solid horizontal line (X) indicates the reflectivity change that arises when the fiber tip goes from a liquid to a gaseous environment. (b) Frequency of reflectivity oscillations as a function of time following a 1 μs pump pulse with a peak power of 16.4 mW. The inset shows maximum induced reflectivity change (ΔR) as a function of pump power for pulses of 1 μs duration.

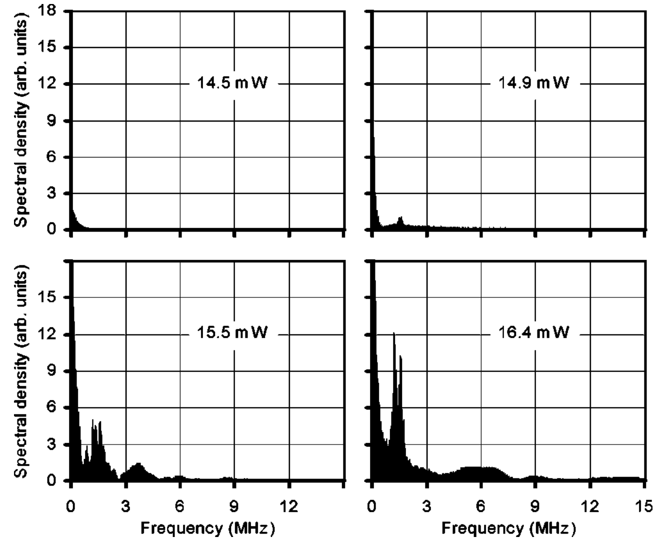

FIG. 3. Frequency spectra of reflectivity oscillations following the generation of a bubble on the tip of a fiber by 1 μs optical pulses with various peak powers.

We observed that with 1 μs pump pulses, bubble creation (detected as a large, rapid change in fiber tip reflectivity) requires a threshold pulse energy of ~ 15 nJ [see inset to Fig. 2(b)]. Following creation, large-amplitude oscillations are seen in the reflectivity of the fiber tip against the background of a nonoscillating reflectivity increase that peaks approximately 5 μs after the end of the pulse and then takes around ~ 15 μs to relax [see Fig. 2(a)]. The oscillations are fastest immediately after the pump pulse, when they have a frequency of ~ 17 MHz [Fig. 2(b)]. The oscillation frequency then drops rapidly, settles at ~ 1.35 MHz within a few microseconds, and remains almost constant for 25–30 cycles, after which it decreases slightly and becomes less stable—varying between 0.1 and 0.8 MHz. In total, the oscillations persist for ~ 200 μs after the laser pulse. Fourier analysis of the reflectivity's time dependence shows that the frequency spectrum of the oscillations broadens with increasing pump power but a characteristic peak at ~ 1.5 MHz is always present (see Fig. 3).

We believe that when a pulse of laser light is delivered to the fiber tip, a bubble forms, initially as a shallow meniscus, on the fiber's core (i.e., on that part of the surface which is exposed to the laser radiation), and that it then grows toward a more hemispherical shape during the pump pulse. After termination of the laser excitation, the bubble can be considered as a free (i.e., undriven) oscillator [7]. The natural frequency of a spherical gas bubble in a liquid, undergoing low-amplitude oscillation, was first calculated by Minnaert [14] (assuming adiabatic compression): $\omega_0 = \sqrt{3\gamma p_0 / \rho R_0^2}$, where ω_0 is the angular frequency, γ is the ratio of specific heats (c_p / c_v) for the gas, p_0 is the static pressure in the liquid, ρ is the liquid density, and R_0 is the bubble's equilibrium radius. Clearly, we do not have an isolated spherical bubble but, as discussed by Jones and Edwards [15], the spherical symmetry of such an equation means that it should be applicable to a hemispherical cavity on a semi-infinite solid surface. The adiabatic regime will apply only to the oxygen component of our bubbles because, as the bubble contracts, the nitrogen will simply condense on its surface [the process of condensation can be very rapid, occurring in a time of the order of the gas collision time ($\sim 10^{-10}$ s), which is much shorter than the bubble's oscillation period]. Oxygen does not condense because its boiling point is lower than that of nitrogen. Nevertheless, if Minnaert's equation is applied, the observed "steady-state" frequency of ~ 1.35 MHz ($= \omega_0 / 2\pi$) is found to correspond to a bubble radius of 2.8 μm ($\gamma = 1.453$, $p_0 = 1$ atm, and $\rho = 809$ kg/m^3). This value is consistent with an upper limit on the radius set by energy conservation considerations. Indeed, calculations based on a modified version [16] of Yamaguchi, Yoshida, and Kinbara's model for the optical properties of an aggregated metal film on a dielectric substrate [17] show that the film absorbs not more than 5% of the 15 nJ pulse energy. If all of this absorbed energy were expended in the evaporation of liquid nitrogen (latent heat of vaporization 2×10^5 J/kg) then a hemispherical bubble with a maximum radius of 7.4 μm would be formed (assuming atmospheric pressure inside the bubble).

We believe that changes in the bubble's size and shape affect the intensity of light reflected back into the fiber for subsequent detection by our measurement system, primarily because the surfaces of the fiber and the bubble act as the mirrors of a dynamic interferometer, akin to a Fabry-Pérot interferometer. An interferometer's reflectivity is increased and decreased by constructive and destructive interference between light reflected from the different surfaces. Thus, the continuous component of a bubble's movement (i.e., the overall expansion and contraction of the bubble) would generate a small number of reflectivity oscillations—roughly one for each half-wavelength change in the ''height'' of the bubble (where height is defined as the distance between the centerpoint of the fiber core's surface and the opposing bubble wall). Most of the reflectivity oscillations, however, would correspond to oscillatory movements of the bubble's surface. Interferometers are normally designed to minimize losses and are based on two plane parallel mirrors or a confocal arrangement of curved mirrors. So, to demonstrate that the interferometric mechanism can still produce significant reflectivity oscillations when one mirror (the bubble surface) is nonplanar and of variable curvature, and when account must be taken of the efficiency with which reflected light is coupled into the fiber, we performed a finite element numerical simulation of the electromagnetic field structure in and around a bubble on the surface of a fiber, and from there derived the intensity of reflected light. We considered a simplified structure wherein the bubble was located on the tip of an uncoated fiber and had the form of a ''spherical cap'' with a fixed base diameter equal to that of the fiber's core (9 μm) and a variable radius of curvature corresponding to heights of up to 4.5 μm (the core radius). The following values were used for the refractive indices of the structure's various components at the probe wavelength ($\lambda = 1.31$ μm): fiber core $= 1.4677$; fiber cladding $= 1.4624$; liquid nitrogen $= 1.4520$; gas inside bubble $= 1.0000$. This simulation showed that the smooth expansion or contraction of a bubble does indeed produce reflectivity variations with a period (in terms of bubble height) of ~ 0.68 μm (roughly $\lambda/2$ as expected) and that, in this simplified case, changes in height of less than $\lambda/4$ can lead to substantial reflectivity modulations that are more than sufficient to explain our experimental results.

In summary, we report that the absorption of low-power laser radiation by a nanoscale metallic film on the tip of an optical fiber immersed in boiling liquid nitrogen can lead to the formation of a freely oscillating bubble with a radius of just a few micrometers on the surface of the fiber's core. Reflectivity measurements, made via the same fiber, enable accurate studies of the bubble's dynamics to be performed. A potential advantage of this bubble nucleation technique is that it could be used with tapered fibers having optical apertures of just a few tens of nanometers (such as those widely used in near-field spectroscopy [18]). With these subwavelength apertures it may be possible to generate and study extremely small ''nano-bubbles,'' which are expected to show interesting thermodynamic properties [19] and could act as cavities to suppress or enhance the spontaneous emission of atoms [20]. We would expect a nanobubble in a cryogenic liquid to oscillate at frequencies of several tens of megahertz in highly controllable environments and could provide interesting opportunities for research on matter in extreme conditions and sonochemistry. Investigations of the optical properties of such nanobubbles would complement the burgeoning research on the optical properties of nanoparticles and their shape-oscillation dynamics [21].

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