

## Decoupling bulk and surface radiation forces in a dielectric liquid

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The effect of radiation forces within a dielectric material and at its interface has been a long-standing debate for over a century. Yet there has been so far only limited experimental verification in complete accordance with the concurrent electrodynamic theories. Here, we use tightly focused pulsed laser beam to detect bulk and boundary optical forces in a dielectric fluid. From the optical convoluted signal, we decouple thermal and nonlinear optical effects from the radiation forces using a theoretical interpretation based on the Microscopic Ampère force density. It is shown, for the first time, that the time-dependent pressure distribution within the fluid chiefly originates from the electrostriction effects. Our results shed light on the contribution of optical forces to the surface displacements observed at the dielectric air-water interfaces, thus shedding light on the long-standing controversy surrounding the basic definition of electromagnetic momentum density in matter [1-4].

The effects of radiation pressure exerted on a dielectric surface parallel to the propagation of the incident electromagnetic radiation can be interpreted as the transfer of momentum from the photons at the surface. Radiation pressure effects were predicted by Maxwell in 1871 and experimentally observed by Lebedew in 1900. In 1905, Poynting presented a detailed geometrical calculation of the force by radiation pressure of light incident from free space on a transparent and non-dispersive dielectric medium, which predicted an outward force normal to the surface of the dielectric, opposite to the direction of propagation of the incident electromagnetic field. Conflicting theories for the energymomentum tensor were proposed by Minkowski in 1908 and Abraham in 1909 to explain this effect. These have subsequently been extensively debated in the literature over the past century. The mostused electrodynamic theories are the Abraham, Minkowski, Einstein-Laub, Chu, and Amperian formulations [3]. These theories can be used in conjunction with the elastodynamic theory to simulate the shape, amplitude, and speed of momentum-driven elastic waves using the elastic properties of the medium and the properties of the incident light. The absolute surface displacement measurements and the simulated displacements, based on first principles, would provide a highly rigorous method to correlate the elastic waves in an illuminated object to the electromagnetic momentum delivered by the incident light.

The measurements of the surface deformation at the air-liquid interface are performed using the photomechanical mirror technique [2,3]. In this method, the probe laser is reflected off the water surface



and the cylindrically symmetric surface deformation generated by the laser excitation causes focusing or defocusing of the central portion of the probe laser beam. A convex deformation is similar to a convex mirror in turn causing the intensity of the probe laser to decrease in the far field while a concave deformation focuses the probe and thereby increases the power that passes through the pinhole placed in front of the detector (Fig. 1a). In the continuous irradiation experiment, the calculated surface distortion is always convex and the corresponding signal shows a decrease in the probe power past the pinhole at all times. As illustrated in Fig. 1b, during pulsed irradiation, the surface first produces a convex column. The column subsequently collapses after irradiation causing a concave surface perturbation. This behavior corresponds to the probe laser power initially decreasing then increasing past the pinhole. The numerical calculations are in excellent agreement with our experimental results, in a test that is significantly more discerning than the earlier experiments by Ashkin and Dziedzic [5]. This demonstrates that this light-matter system is well modeled by our present understanding of radiation forces that lead to the momentum transfer. The Helmholtz force density is used to describe the imparted pressure on the surface of the liquid.

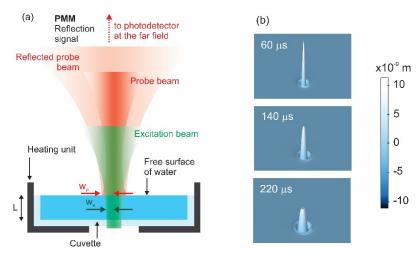
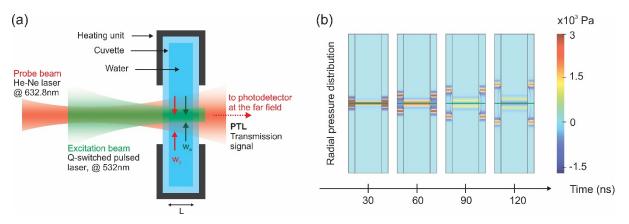


Fig. 1. (a) Schematic diagram of the apparatuses for the time-resolved photomechanical mirror used for pulsed excitation. The probe beam senses the entire region affected by the excitation laser. The complex reflection pattern of the probe beam just out of the sample propagates to the detector plane. The intensity variation measured at the center of the probe beam in the far field consists of complex contributions originating from all the surface waves created on the water. (b) The numerical simulation of the time evolution of the water surface deformation under pulsed excitation. A sharp peak appears a few microseconds after irradiation and is subsequently dispersed on the surface. The probe beam senses the entire region affected by the excitation laser. The complex reflection pattern of the probe beam just out of the sample propagates to the detector plane. The intensity variation measured at the center of the probe beam in the far field consists of complex contributions originating from all the surface waves created on the water.

To experimentally detect the dynamics of the pressure-induced acoustic waves by optical forces within water, we exploit a high sensitive detection of wavefront distortions by a time-dependent photo-induced lensing (PL) technique (Fig. 2). Nanosecond laser pulses irradiate the sample, changing the local pressure due to the radiation forces in addition to a small heat deposition. Nonlinear optical Kerr effect is also observed during the pulse duration. A low-irradiance laser beam traverses the sample thus probing the induced effects. Intensity of the probe beam is monitored in the far field by a fast photodetector.





**Fig. 2.** (a) Photo-induced lensing method - schematic of the time-dependent photo-induced lensing measurement set-up. Green and red routes represent pump and probe laser beams, respectively. (b) Radial pressure distribution built up in the sample over time.

The wavefront distortion sensed by the probe beam originates from the non-uniform excitation interaction with the sample leading to an increase in the internal energy, the latter being dispersed in two different modes of hydrodynamic relaxation. The increased internal energy results in a temperature change in the sample or the coupling material placed next to the sample. This temperature change results in a change in sample density. If the photothermally induced temperature alteration occurs faster than the time required for the fluid to expand (or contract, in some cases), then the rapid temperature change will result in a pressure change. The pressure perturbation then relaxes by emitting an acoustic wave. Once the pressure has relaxed to its equilibrium level, a density change proportional to the temperature will remain. The time-dependent intensity signal detected in the experiments shows only the center of the probe beam spot at the detector plane in the far-field region. The calculation of the PL signal requires the determination of the temperature and pressure fields considering all the effects of the radiation forces in the liquid and in the cuvette walls.

This work presents experimental methods to quantitatively measure the momentum coupling between the electromagnetic field and matter [1-4]. These methods can be applied to characterize materials, to further advance optical manipulation technology of deformable matter, and to provide the means to empirically validate differing electrodynamic formalisms, commonly known as the Abraham–Minkowski controversy. Even though substantial efforts have so far been devoted to addressing the question of the basic definition of electromagnetic momentum density in matter, this fundamental problem in classical and quantum mechanical theory of electrodynamics has not been unequivocally resolved. Our work represents a bold step in this direction, facilitating resolution of the century old Abraham-Minkowski controversy.

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