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CORRIGENDUM

Corrigendum: Photon momentum transfer at water/air interfaces under total internal reflection (2019 *New J. Phys.* **21** 033013)

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On page 3 of the manuscript there is a typo in the equation:

$$I_s = p_{\max}^2 / (\nu^2 \rho).$$

The correct form of this equation is:

$$I_s = p_{\max}^2 / (\nu \rho).$$

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PAPER

Photon momentum transfer at water/air interfaces under total internal reflection

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Abstract

The transfer of photon momentum at the water/air interface is important for optical manipulation of minute particles and is at the heart of the Minkowski–Abraham controversy. We use photoacoustic (PA) detection of ultrasound waves generated when pulsed laser light meets the water/air interface at 3.9 °C (zero thermal expansion), to distinguish momentum transfer from thermoelastic effects. The PA waves dependence on the angle of incidence reveals that momentum transfer maximizes at the critical angle. Momentum transfer is most efficient when the photons travel in water and remain in water after total reflection at the interface, rather than when they cross the interface between dielectric media.

Every time photons meet an air/water interface, they transfer momentum to that interface. Although this is a fundamental process in nature, conflicting theories on the momentum of a photon in a dielectric medium of refractive index n coexist since Minkowski [1] and Abraham [2] predicted the opposite change in momentum when light enters the optical material. Defining $p_0 = \hbar\omega/c$ as the photon momentum in the vacuum, Minkowski proposed a momentum $p_M = p_0 n$ that produces an outward force in the medium, whereas Abraham derived a momentum with the form $p_A = p_0/n$, which corresponds to an inward force in the medium. A compelling experiment in favor of $p_M = p_0 n$ was the observation that the pressure exerted by light on a mirror immersed in a liquid of refractive index n was n times higher than the pressure exerted by the same light in free space [3, 4]. An alternative argument, but with the opposite conclusion, is the thought experiment of a light pulse initially travelling in the vacuum and then intercepting a reflection-less planar slab, normal to its surface, where it slows down its velocity to c/n [5]. Conservation of center-of-mass velocity requires the slab of mass M to move with a velocity $(mc - mc/n)/(m + M)$, where m is the dynamical mass of a photon in vacuum with energy $E = mc^2$, and conservation of global momentum requires the pulse to have the Abraham momentum. Various interpretations and experiments continue to contribute to this lively debate [6–8].

Barnett recently proposed a resolution to this paradox [9, 10], recalling that the kinetic momentum of a body is the product of its mass and velocity but, in quantum mechanics, the canonical momentum is Planck's constant divided by its de Broglie wavelength, h/λ, following the canonical commutation relation $[\hat{x}, \hat{p}] = i\hbar$. Barnett identified p_A as the kinetic momentum and p_M as the canonical momentum of light. The kinetic (Abraham) momentum is associated with particlelike phenomena such as momentum transferred by forces, and readily explains the uniform motion of the center-of-mass energy. The canonical (Minkowski) momentum is related to the idea of translations within, or relative to, a host medium and to the wavelength of light, explaining the immersed mirror experiment and wavelike phenomena such as diffraction. However, interpreting the manifestations of photon momentum transfer remains problematic [7, 11–17].

Two major experimental challenges must be overcome to measure the transfer of momentum at the air/water interface. First, for optical powers below the onset of self-focusing, the surface distortions are in the picometer/nanometer scale and very sensitive detection methods are required. Second, even though the absorption coefficient of water can be very low at selected wavelengths [18], the heating due to residual light

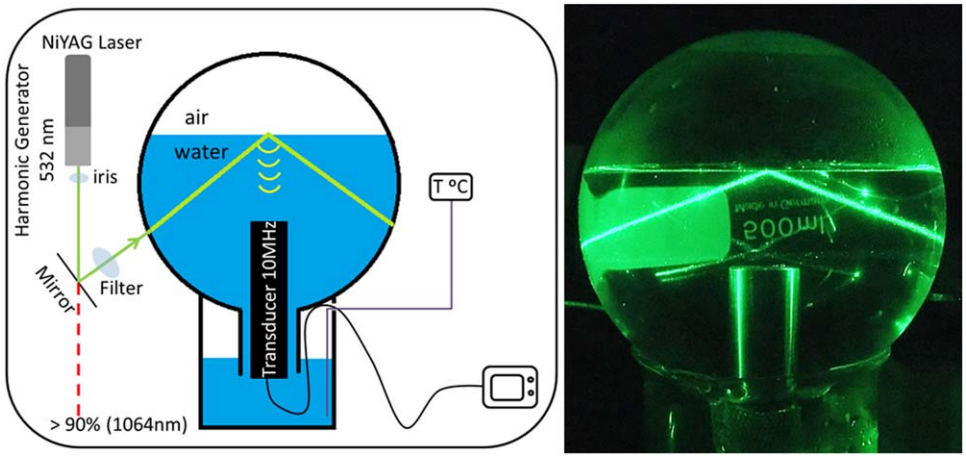

Figure 1. Experimental configuration. (Left) Schematic representation (filter: cutoff 1064 nm or cutoff 532 nm). (Right) Image obtained for $\theta_i = 68.6°$, where total internal reflection is observed.

absorption can nevertheless produce a thermoelastic expansion that obscures the transfer of photon momentum [6, 16, 19].

In this Letter, we investigate the perturbation induced by an unfocused laser pulse in the water/air interface using photoacoustic detection. Photoacoustic waves (PAs) generated by transient volume changes can be detected with high sensitivity using ultrasonic transducers [20–24]. This enables the measurement of the displacement of the surface molecules (inwards or outwards relative to the medium) either as a result of (inward or outward) pressure at the surface or of thermoelastic changes. Moreover, we take advantage of an exceptional property of water to distinguish between volume changes due to radiation forces at the water/air interface from volume changes due to thermal losses: water attains its maximum density at 3.9 °C. Hence, the coefficient of thermal expansion (α) changes from $\alpha > 0$ for $T > 3.9$ °C to $\alpha < 0$ for $T < 3.9$ °C. These changes in α make the amplitudes of PA waves of thermoelastic origin change signs, and go through zero, at 3.9 °C [25]. At $T = 3.9$ °C the PA waves report exclusively the volume changes due to surface distortions induced by momentum transfer. Additionally, the dependence of the PA waves on the angle of incidence of the laser pulse is also investigated. We show that photon momentum transfer is higher under total internal reflection (i.e. when the photons are reflected back into the dielectric medium), rather than when light crosses the interface between different dielectric media.

The experimental configuration is schematically presented in figure 1. The laser pulse reaches the water/air interface from below (water), with an angle of incidence θ_i relative to the normal. This angle was varied from nearly normal to the interface ($\theta_i = 0$), to larger than the angle of total internal reflection. According to Snell's law, using $n = 1.33$ for water at 20 °C, the critical angle is $\theta_c = 48.75°$. Water was initially cooled below 3.9 °C and its temperature was allowed to slowly increase to room temperature. At various temperatures, measured with a thermocouple thermometer (Delta OHM model HD 2108.1), laser pulses were directed at the selected angle to the water/air interface and the PA waves generated were detected with an A311S Panametrics (Olympus) immersion piezoelectric transducer (10 MHz), pre-amplified (Panametrics ultrasonic preamp) and connected to a Tektronix digital oscilloscope (model DPO7254, 2.5 GHz, 40 GS s^{-1}). Measurements of absolute pressures employed a calibrated membrane hydrophone Onda model HMB0500, 20 MHz. The laser pulses were generated with a Quantel Big Sky Ultra 50 Nd:YAG laser (pulse width $\tau = 8$ ns, TM polarization). The laser was set to deliver 532 nm laser pulses with proper choice of harmonic generators. However, 1064 nm light is also present in such pulses. We used a dielectric mirror (Unaxis BD10307805) ca. 90% transparent at 1064 nm, to reflect mostly the 532 nm to our setup. Additionally, we used Newport filters 10LWF-650-B and 10SWF-900-B to cutoff either 532 nm or 1064 nm laser pulses, respectively, from the beam directed to the setup. The spectral purity of the beam reaching the water/air interface was checked with an Avantis spectrophotometer. The laser pulse energies entering the setup were either 1.65 mJ at 532 nm or 0.165 mJ at 1064 nm, perfectly separated. The light spot area on the water/air interface was always smaller than transducer area ($A = 0.8$ cm^2).

Figure 2(a) shows PA waves collected at different temperatures using either 532 or 1064 nm pulses. The PA waves generated with 1064 nm laser pulses have the behavior expected for photothermal processes: as the temperature is allowed to increase from near 0 °C to room temperature, the amplitudes change sign at ca. $T = 3.9$ °C, from a thermoelastic contraction to a thermoelastic expansion (figure 2(b)). The amplitudes of the PA waves generated by 532 nm laser pulses are rather insensitive to the temperature (figure 2(c)) and must be assigned to transfer of momentum at the interface.

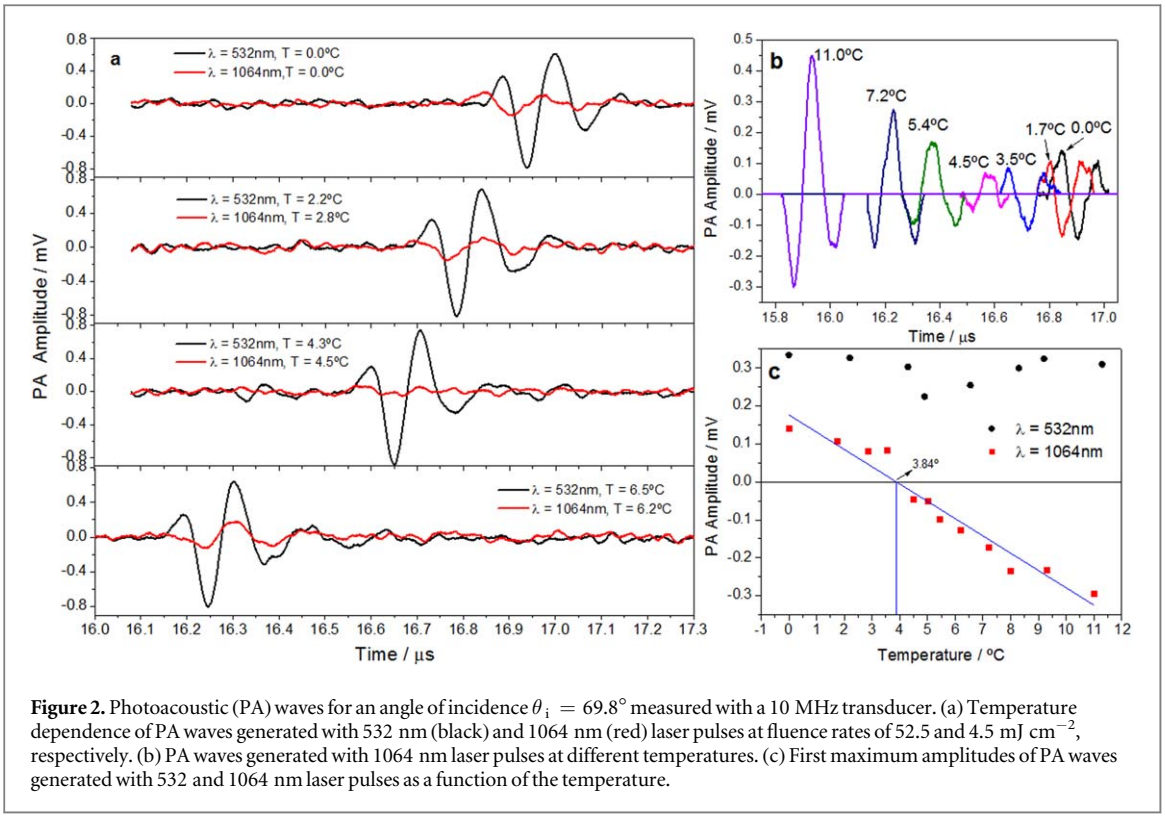

Figure 2. Photoacoustic (PA) waves for an angle of incidence $\theta_i = 69.8°$ measured with a 10 MHz transducer. (a) Temperature dependence of PA waves generated with 532 nm (black) and 1064 nm (red) laser pulses at fluence rates of 52.5 and 4.5 mJ cm^{-2}, respectively. (b) PA waves generated with 1064 nm laser pulses at different temperatures. (c) First maximum amplitudes of PA waves generated with 532 and 1064 nm laser pulses as a function of the temperature.

The absorptivity of water increases from ca. 0.0015 m^{-1} at 532 nm, where it is close to its minimum, to ca. 3 m^{-1} at 1064 nm [18]. As shown above, light absorption at 1064 nm is responsible for photothermal effects that can only be avoided near 3.9 °C. Considering the differences in fluence rates at 1064 and 532 nm, the temperature dependence of the PA waves and the light transmission over a 3 cm optical path in water, we estimate that, at room temperature, the thermoleastic expansion originated by a 1064 nm laser can generate a PA wave ca. 30 times more intense than the PA wave due to momentum transfer for the same laser pulse energy. The difference of absorptivities inverts this ratio to 1/100 when 532 nm laser pulses are employed. Care must be exercised in the separation of the Nd:YAG harmonics to avoid the contamination of momentum transfer by thermoleastic expansion at room temperature. A thermoelastic expansion will look like the transfer of Minkowski momentum.

The transfer of photon momentum at the water/air interface is related to the conversion of optical power in sound power. The incident laser optical power density on the interface is $I_L = E_L/(A\tau)$, which for $E_L/A = 52.5$ mJ cm^{-2} and $\tau = 8$ ns gives $I_L = 6.6$ MW cm^{-2}. The average sound power density can be calculated with $I_s = p_{max}^2/(v^2\rho)$, where v is the sound velocity and ρ the density, but requires the measurement of the absolute maximum pressure p_{max}. The PA waves generated by low laser fluences at the water/air interface are too weak to be detected by a hydrophone. On the other hand, piezophotonic materials are designed to produce very intense PA waves [26], which enable their measurement by a hydrophone and its indirect calibration with an immersion transducer. Hence, we used a calibrated hydrophone and 532 nm laser pulses with different energies to measure p_{max} generated by a piezophotonic film [26] floating on water, and repeated the measurement using the immersion transducer. The frequency range of the calibrated hydrophone (1–20 MHz) overlaps with the frequency range of the immersion transducer response (7.6–13 MHz, within −6 dB). The linear relation between p_{max} from hydrophone and voltage from immersion transducer allowed us to estimate $p_{max} = 11$ Pa for photon momentum transfer, and calculate $I_s = 5$ pW cm^{-2}. Hence, under the conditions of this experiment, the conversion of optical power into acoustic power by photon momentum transfer is only one part in 10^{18}.

Figure 3 shows the angle dependence of the PA waves generated by momentum transfer. Following Snell's law, the angles between $\theta_c = 48.75°$ and 90° correspond to total internal reflection. There is a very strong dependence of the amplitudes on the angle, with the maximum PA wave generated at θ_c. According to Minkowski formalism, the angular dependence of the momentum transfer has the form [27]

$$f(\theta_i) = \cos^2\theta_i[1 + R - (\tan\theta_i / \tan\theta_r)T], \quad (1)$$

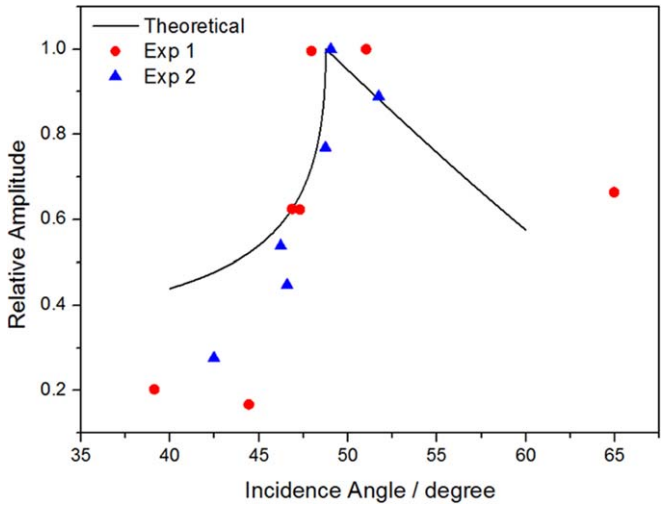

Figure 3. Incident angle dependence of PA waves generated by 532 nm laser pulses in independent experiments, and behavior expected from equation (1).

where θ_i and θ_r are respectively the incident and the refraction angles versus the interface normal, and R and T are the Fresnel coefficients of reflection and transmission in energy. Figure 3 shows that the Minkowski momentum should attain its maximum at θ_c in reasonable agreement with our observations.

The assignment of origin of the PA waves observed to the transfer of Minkowski momentum or of Abraham momentum depends on whether the water surface is pushed out towards air or pulled inwards towards the bulk, respectively. The first case should lead to a PA wave with a form similar to that of a thermoelastic expansion, whereas in the second case the PA wave should resemble that of a thermoelastic contraction. The expansion, or contraction, depends on the release of heat in the medium by the water molecules that absorbed the laser pulse. This process is independent of the direction of the laser pulse. Figure 4 shows that this is indeed the case for 1064 nm laser pulses incident at different angles on the water/air interface. On the other hand, directionality of momentum transfer sets different components in the acoustic waves that for angles very far from the normal ($\theta_i = 0°$ in our definition of angle of incidence) may change the transducer waveform. Figure 4 shows that for lager values of θ_i (i.e. for $\theta_i > 60°$), the transducer waveform changes. Ideally, the PA waves generated by photon momentum transfer (532 nm laser pulses) and by thermoelastic expansion at room temperature (1064 nm) should be compared for normal incidence ($\theta_i = 0°$) to avoid angular dependences. In practice, figure 3 shows that this is not possible because the intensity of the PA drops dramatically. Figure 4 shows that as θ_i decreases, the transducer waveform due from momentum transfer approaches more closely that originated by thermoelastic expansion. Hence, photon momentum transfer displaces surface water molecules outwards to air. This is a manifestation of an outward force at the interface, i.e. of the Minkowski momentum.

The transfer of photon momentum to water molecules at the water/air interface was unambiguously assigned at $T = 3.9°$ making use of the absence of thermal expansion of water at this temperature. The transfer of photon momentum at the water/air interface leads to the conversion of 1 part in 10^{18} of optical power into acoustic power. This minute conversion nevertheless produces PA waves that can be detected with ultrasonic transducers when nanosecond pulsed lasers with modest fluence rates are employed. The PA waves observed are consistent with the Minkowski momentum: the optical radiation pressure pushes the water/air interface outwards to air and the angular dependence of the PA waves follows the predictions of Minkowski. Far from the critical angle, where the momentum transfer is the highest, the PA waves decrease significantly in intensity and are difficult to measure. We did not succeed in detecting PA waves originated by photon momentum transfer at the air/water interface when laser pulses travel from air to water, possibly because of the lower conversion of optical power into acoustic power. Refraction is always present in this configuration. The fraction of photon momentum transferred is smaller when refraction is higher, i.e. when more photons cross the interface.

The thermoelastic properties of water and the sensitivity of the photoacoustic technique provided the conditions necessary to measure the transfer of photon momentum at the water/air interface. The thermoelastic component due to concurrent light absorption by water was suppressed by proper control of water temperature or selection of the wavelength. Photon momentum transfer was maximized at the critical angle, when the evanescent wave appears. The frustrated attempt of photons to cross the water/air boundary leads to a measurable loss of photon momentum. Minkowski and Abraham theories deal with changes in photon momentum when the boundary between different dielectric media is crossed. The total internal reflection of the

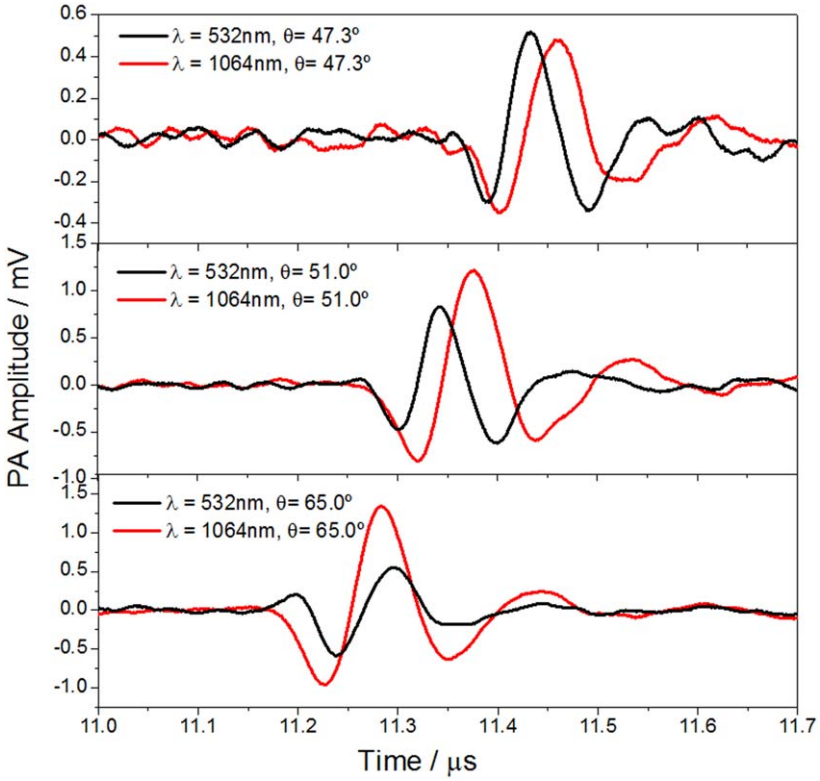

Figure 4. PA waves generated at 1064 nm (fluence rate 5 mJ cm^{-2}) and 532 nm (fluence rate 53 mJ cm^{-2}) at different angles of light incidence.

photons is also a case of photon momentum discontinuity at an interface, but our measurements shows that when light is reflected at the water/air interface and remains in water, a larger change in photon momentum occurs than when light crosses the interface. This observation unveils new opportunities in optical manipulations of fluids.

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