

Large spectral tuning of liquid microdroplets standing on a superhydrophobic surface using optical scattering force

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(Received 18 October 2007; accepted 13 November 2007; published online 3 December 2007)

We demonstrate large spectral tuning of glycerol/water microdroplets standing on a superhydrophobic surface using the optical scattering force exerted by a 1064 nm Nd³⁺:YVO₄ solid-state laser. Spectral tuning up to 30 nm is presented in the whispering gallery modes as a result of the deformation of the microdroplets toward a truncated prolate spheroid geometry. Observed large spectral tuning is also reported to be highly reversible. This demonstration can inspire novel, largely tunable optical switches or filters based on liquid microdroplets kept in a sealed chamber.
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Large deformations can easily be introduced in liquid microdroplets by applying relatively small external forces or controlling the evaporation/condensation kinetics.¹ Liquid microdroplets have therefore the potential to function as largely tunable optical microcavities. It is possible to imagine liquid microdroplets kept in a sealed chamber as the building blocks of tunable switches or filters that are essential in optical communication systems based on wavelength division multiplexing.² Solid optical microcavities have not found large use in these applications, mainly due to their rigid nature which limits the spectral range of tunability.³ The fact that liquid microdroplets are low cost and disposable can also prove to be important in mass production of these photonic devices.

Ashkin has first demonstrated the acceleration and trapping of dielectric particles using the radiation pressure of an incident laser beam.⁴ Since this demonstration, optical forces have proven to be indispensable in many applications providing noninvasive manipulation of mesoscopic particles in the Rayleigh (particle size $< \lambda$) and the ray optics (particle size $> \lambda$) regimes.⁵ Liquid microdroplets have been suspended in air using optical levitation,⁶ or recently using optical tweezing.⁷ In this letter, we use the optical scattering force to largely tune the whispering gallery modes (WGMs) of glycerol/water microdroplets standing on a superhydrophobic surface. We achieve spectral tuning by adjusting the power of a 1064 nm, continuous-wave (cw) Nd³⁺:YVO₄ laser focused approximately at the center of individual microdroplets. The applied optical scattering force results in the deformation of the microdroplets to a truncated prolate spheroid geometry. Microscope images revealed up to 40% reduction in the equatorial diameter of a microdroplet upon excitation with 43 mW of laser power measured at the focus of the microscope objective. For smaller deformations, spectral tuning of a specific WGM up to 30 nm is demonstrated. Observed large spectral tuning is also demonstrated to be highly reversible.

The effect of deformation on the WGMs has been previously studied using chain of microdroplets.⁸ Spectral shifts of less than 1 nm were observed in the WGMs along the entire rim in slightly deformed microdroplets positioned at different distances from the microdroplet generator. How-

ever, in these experiments the deformation of individual microdroplets was not controlled.

Figure 1 shows the sketch of the employed spectral tuning mechanism. A liquid microdroplet is stabilized on a superhydrophobic surface, and a trapping laser beam propagating in the upward vertical direction (z direction in Fig. 1) is focused near the center of the microdroplet. Upon scattering, a net scattering force is applied on the microdroplet along the propagation direction of the trapping laser beam.⁹ The gradient force on the microdroplet is zero in the idealized case when the laser is focused to the center of a microdroplet suspended in air. Since the microdroplet is held by the surface, the net upward force results in the deformation of the microdroplet toward a truncated prolate spheroid as shown in Fig. 1(b). The resulting equatorial radius (r_2) of the microdroplet becomes smaller than the initial radius (r_1), leading to a blueshift in the observed resonance frequencies of the WGMs which circulate around the circumference of the microdroplet in the equatorial plane, parallel to the surface.

The presence of the superhydrophobic surface is essential for the reported spectral tuning mechanism. The surface holds the microdroplet against the impulse generated by the optical scattering force, enabling its elongation in vertical direction. Optical scattering force would not lead to such a deformation for microdroplets suspended in air by optical levitation or optical tweezing. In the case of optical levitation, increased trapping laser power would result in the upward motion of the microdroplet. In optical tweezing, the

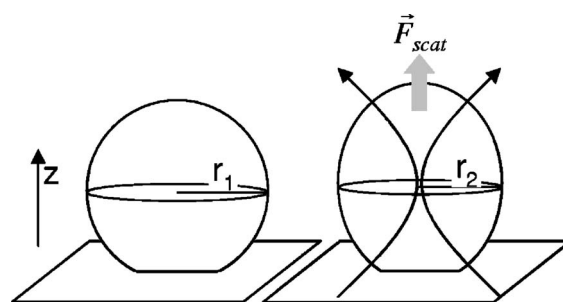


FIG. 1. Illustration of the spectral tuning mechanism. The scattering force of an infrared laser, that is focused to near the center of the microdroplet, is in the z direction. This causes a deformation in the truncated spherical shape of the microdroplet (left) toward a truncated prolate spheroid geometry (right). As a result, the equatorial radius drops from r_1 to r_2 .

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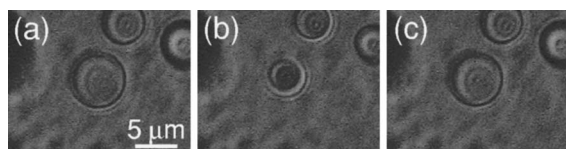


FIG. 2. Consecutive microscope images of a microdroplet under the [(a) and (c)] absence and (b) presence of 43 mW infrared laser. In the presence of the infrared laser, the diameter of the microdroplet drops from (a) 6.5 μm to (b) 4.1 μm . When the laser is once more blocked, the diameter of the microdroplet recovers to (c) 6.1 μm .

trapped microdroplet is stabilized at a position in which the scattering and gradient forces are equal to each other. Since both of these forces are linearly proportional to the trapping laser power, the equilibrium force applied to the microdroplet would remain to be 0 with increased trapping laser power, hence no size deformation would be observed.

Superhydrophobic surfaces were prepared by spin coating 50 mg/ml ethanol dispersions of hydrophobically coated silica nanoparticles (Aeroxide LE2, Degussa AG) on cover glasses, as reported previously.¹ For millimeter-sized water droplets an average contact angle larger than 150° was measured on these surfaces. Glycerol/water microdroplets were sprayed onto the superhydrophobic surface from a 7.5/92.5 glycerol/water solution containing 50 μM Rhodamine B molecules using an ultrasonic nebulizer within a humidity chamber. Size stabilization was achieved with gradual evaporation of the water content in the microdroplets. Microscope images revealed the glycerol content to be $\sim 10\%$ when the size equilibrium was reached approximately 1 h after the generation of the microdroplets. The achieved size stabilization was determined to be ~ 10 nm/60 s in the microdroplet radius (~ 1.5 nm/60 s in the spectral position of the WGMs).

A cw solid state green laser ($\lambda=532$ nm, power =20 mW) and a home-built cw $\text{Nd}^{3+}:\text{YVO}_4$ infrared laser ($\lambda=1064$ nm, power adjustable up to 1 W) were reflected off a dichroic mirror and focused using a high numerical aperture (NA) microscope objective (NA=1.40, 60 \times) in the inverted geometry. The same microscope objective was used to collect the fluorescence from individual microdroplets. WGMs were observed by exciting the microdroplets with the green laser in the vicinity of their rims in the equatorial plane (excitation power=1 μW at the focus of the microscope objective), and by recording the fluorescence spectra with a $\frac{1}{2}$ m monochromator (600 grooves/mm grating) and a charge coupled device (CCD) camera providing a spectral resolution of 0.17 nm. A mechanical shutter was used to unblock the green laser during the exposure times of the CCD camera. Microdroplets were constantly exposed to the infrared laser at varying powers during the experiments. All infrared laser powers reported in this letter refer to the powers at the focus of the microscope objective. Total transmission from the output of the infrared laser to the focus of the microscope objective was measured to be 10%.

Using the presented technique, very large deformations can easily be observed in the microdroplets at moderate infrared laser powers. In Fig. 2, three consecutive microscope images of a microdroplet taken in the [Figs. 2(a) and 2(c)] absence and [Fig. 2(b)] presence of the 43 mW infrared laser illumination are shown. Between images (a) and (b), the diameter of the microdroplet drops from 6.5 to 4.1 μm . When the infrared laser is once more blocked, the diameter of the

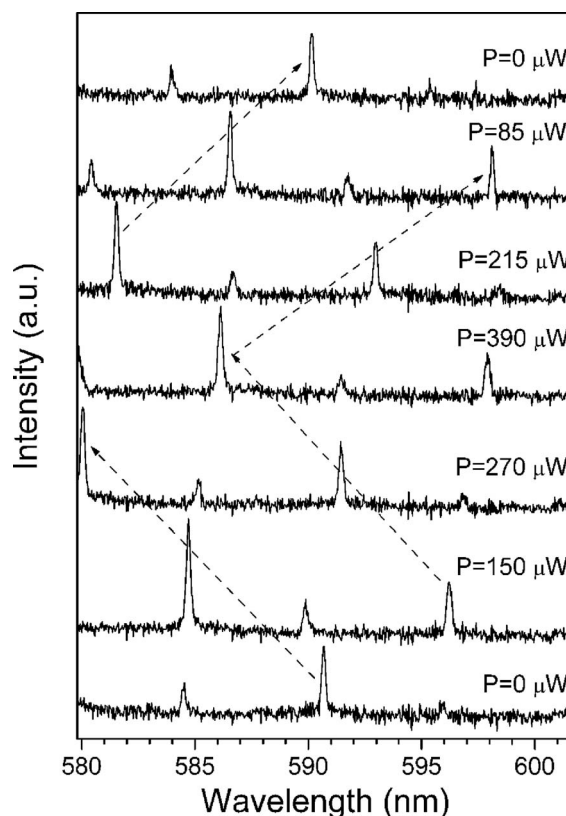


FIG. 3. Emission spectra recorded from an 8.2- μm -diameter microdroplet while the infrared laser power is first increased from and then decreased to zero. Arrows indicate the spectral drift direction of the high quality WGMs belonging to the same mode set. At the maximum infrared laser power of 390 μW , total spectral drift is 16 nm.

microdroplet recovers to 6.1 μm as seen in the image (c), showing the reversibility of the tuning mechanism. The observed large, reversible deformation corresponds to spectral tuning of the WGMs for at least >350 nm (>20 free spectral ranges) for this microdroplet.

We were able to follow the spectral drift of the WGMs under smaller incremental deformations. In Fig. 3, we plot the emission spectra obtained from an 8.2- μm -diameter microdroplet while the infrared laser power is first increased from zero to a maximum of 390 μW , and then decreased back to zero. Arrows show the spectral changes of the sharp WGMs which have a resolution limited spectral width of 0.17 nm ($Q=3500$) that belong to the same mode set. WGMs are first observed to drift to blue wavelengths while the infrared laser power is gradually increased. A total spectral drift of 16 nm is observed at the maximum infrared laser power of 390 μW . Following the maximum blueshift, redshift is observed in the WGMs with decreasing laser power. When the infrared laser is once more blocked, WGMs are observed to be blueshifted by 0.37 nm in comparison to their initial spectral positions. The observed blueshift is mainly attributed to evaporation in the microdroplet due to the heating of the green and the infrared lasers, and instabilities in the evaporation/condensation kinetics in the humidity chamber.

In Fig. 4, we plot the hysteresis curves of the spectral drift in the WGMs as a function of the infrared laser power while the power is first increased and then decreased to zero. The microdroplet discussed in Fig. 3 (microdroplet A) as well as two other microdroplets (microdroplets B and C)

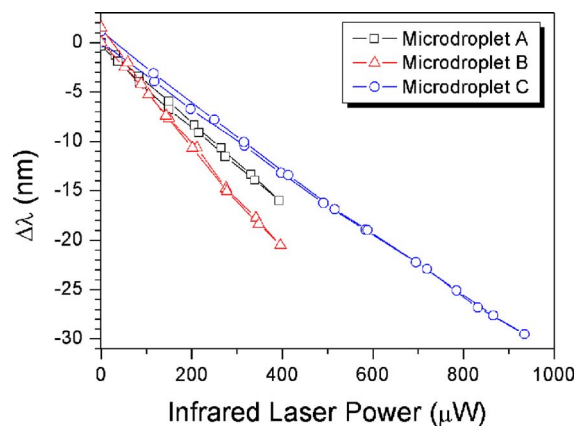


FIG. 4. (Color online) Hysteresis curves of the spectral drift in the WGMs as a function of the infrared laser power for three different microdroplets with diameters of (microdroplet A) $8.2 \mu\text{m}$, (microdroplet B) $9.0 \mu\text{m}$, and (microdroplet C) $8.2 \mu\text{m}$. At the infrared laser power of $935 \mu\text{W}$, a maximum spectral drift of 30 nm is observed in microdroplet C.

were used. Almost no hysteresis is observed in these curves, showing the high level of reversibility of the presented spectral tuning method within maximum total spectral ranges of 16 , 21 , and 30 nm for microdroplets A, B, and C, respectively. The observed high linearities in the hysteresis plots are due to the relatively small deformations introduced to the microdroplets. The difference in the slopes of the three hysteresis curves can be due to different viscosities caused by slightly different glycerol percentages or different geometries (contact angles) of the microdroplets.

We have demonstrated a method which enables large (up to 30 nm) and highly reversible tuning of the WGMs of microdroplets standing on a superhydrophobic surface. The presented method poses no fundamental limitation on increasing the maximum possible tuning range of 30 nm when required. Our demonstration can inspire novel applications in optical communications systems. Tunable optical switches or filters

based on liquid microdroplets kept in a sealed chamber can readily be envisioned. Such devices can also benefit from the fact that liquid microdroplets are disposable. For future applications, other superhydrophobic surfaces that can provide larger contact angles can be used in order to obtain ultrahigh quality factors in the observed WGMs.¹⁰ The presented method can also be used in characterizing the deformation of a microdroplet under larger scattering forces, or studying the mechanical oscillations of liquid microdroplets with high resolution.¹¹

This work was supported by the Scientific and Technological Research Council of Turkey (Grant No. TÜBİTAK-105T500). The authors thank the Alexander von Humboldt Foundation for equipment donation, and Vahid Sandoghdar for fruitful discussions. A. Kiraz acknowledges the financial support of the Turkish Academy of Sciences in the framework of the Young Scientist Award program (Grant No. A.K/TÜBA-GEBİP/2006-19).

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