Prolonged Raman lasing in size-stabilized salt-water microdroplets on a superhydrophobic surface

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We demonstrate prolonged Raman lasing from individual salt-water microdroplets with 10–20 μm diameters located on a superhydrophobic surface. The mechanism is based on the absorption heating of a 1064 nm cw IR laser and the resonant heating of a 532 nm pulsed, pump laser. A clear hysteresis is observed in the lasing intensity as the droplet size is photothermally tuned by the IR laser, indicating a self-stabilization mechanism due to the resonant absorption of the pump laser. Using this mechanism, Raman lasing near 650 nm is sustained for up to 25 min, ~1000 times longer than lasing durations reported in previous studies. © 2010 Optical Society of America

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Liquid microdroplets are promising candidates as optical microcavities owing to their ideally smooth and nearly spherical geometry. They can possess optical resonances with ultrahigh quality factors (i.e., whispering gallery modes, WGMs) at the spectral region with low material absorption (visible wavelengths for most of the cases). Large optical intensities can be accumulated in their WGMs, making them very suitable for nonlinear optics applications [1]. In fact, single laser pulse excitation of liquid microdroplets generated by a vibrating-orifice aerosol generator was used in the very first nonlinear optics demonstrations using optical microcavities [2,3]. Since then, however, liquid microdroplets have not been widely used in photonic device applications, mainly because of their fragility against external disturbances, unlike their solid-state counterparts [4–7].

Recently, Raman lasing has been observed in microdroplets whose positions were stabilized by using a superhydrophobic surface [8] or electrodynamic levitation [9]. Achieved Raman lasing could not be sustained for long time periods because of frequent breaking of the resonance condition, which was caused mainly by thermal fluctuations induced in the liquid microdroplet during the buildup of the large WGM intensities at the Raman band.

In this Letter, we demonstrate Raman lasing from salt-water microdroplets over long time periods. The technique we use employs two lasers: a cw IR laser for photothermal tuning of the size [10], and a pulsed green pump laser for Raman excitation. The IR laser brings the droplet to a size in resonance with the green pump laser. At this size, the resonant absorption of green laser provides self-stabilization [11–13], which helps maintain the fixed size required for prolonged Raman lasing.

Superhydrophobic surface preparation was described previously [8,10–12]. 3 M NaCl-water solution is sprayed onto the superhydrophobic surface by an ultrasonic nebulizer. The sample containing the microdroplets is then attached to a humidity controlled chamber with relative humidity fixed at 84.34% by a saturated KCl-water solution (KCl-W) in a small extension to the main body of the chamber, as shown in Fig. 1. The humidity chamber is cooled to ~22°C by a thermoelectric cooler (TEC), with no further effort toward temperature control. Microdroplets are excited by a *Q*-switched, frequency-doubled Nd:YAG laser

operating at 532 nm (repetition rate 1 kHz, pulse width 100 ns). The excitation beam is focused to a ~10 μm spot centered at the rim of the microdroplets by using an oil-immersion objective (NA = 1.40, 60×) in the inverted geometry. The average power of the excitation beam is measured to be 8–10 mW at the focus of the objective. A cw IR laser (λ = 1064 nm, maximum output power 300 mW) focused at the center of a microdroplet is used for nonresonant photothermal tuning of the size. The IR laser power (P_{IR}) is controlled by using a half-wave plate (WP) rotated by a stepper motor (9200 steps per revolution) followed by a polarizing beam splitter (PBS). A power meter is placed at the other output of the PBS. The IR beam is combined with the excitation beam at a dichroic mirror (DM1) which then directs both beams to the input port of the microscope. The Raman signal is collected through the same objective, reflected off another dichroic mirror (DM2), and transmitted through a long-pass filter (LP). The signal is then dispersed by a monochromator (spectral resolution 0.07 nm) and detected either by a CCD camera with an exposure time of 50 ms or by a photomultiplier tube (PMT) placed behind an exit slit. The spectral window detected by the PMT is set at ~3 nm with the selected exit slit width. An oscilloscope is used to record the PMT signal.
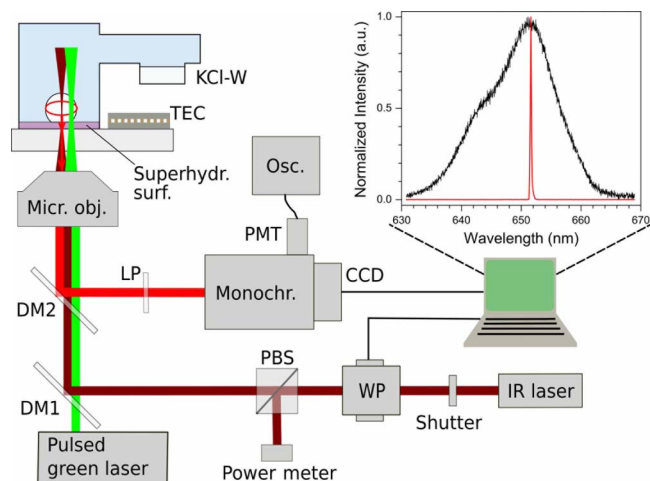

Fig. 1. (Color online) Experimental setup. Inset, normalized Raman spectra of a 5.9 M NaCl-water solution and that recorded from a microdroplet exhibiting Raman lasing.
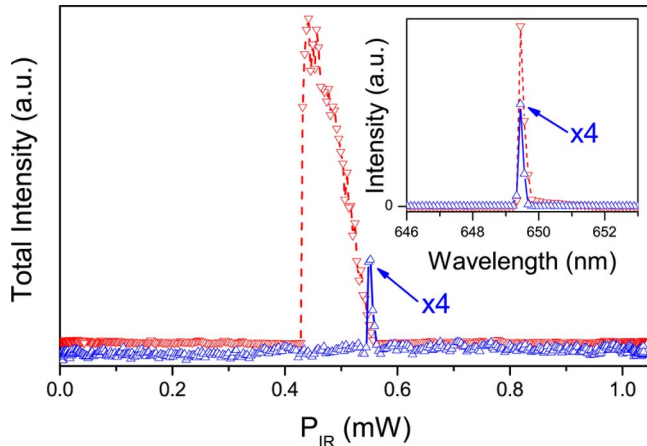
Fig. 2.   (Color online) Total intensity versus IR laser power (P_{IR}), collected from a 12 μm diameter NaCl-water droplet exhibiting Raman lasing in increasing (blue solid) and decreasing (red dashed) P_{IR} sweeps. Inset, Raman lasing spectra from increasing (blue solid) and decreasing (red dashed) P_{IR} sweeps. Intensity during increasing P_{IR} is multiplied by 4 for clarity.

The prolonged Raman lasing in the water microdroplets can be understood based on the photothermal tuning action induced by the IR laser. Photothermal tuning allows for precise tuning of the volume of binary microdroplets by adjusting P_{IR} [10]. For the case of a salt-water microdroplet, this tuning mechanism can be almost fully reversible owing to the nonvolatility of salt at room temperature [14]. Changing P_{IR} slowly ensures—to a good approximation—a one-to-one correspondence between this parameter and the droplet size.

Figure 2 shows the total intensity between 627–669 nm collected from a 12 μm diameter droplet plotted against P_{IR}. P_{IR} is varied smoothly, following one period (a period is 205 s) of a sinusoidal wave, starting from the minimum. Calibration experiments under these experimental conditions revealed a photothermal tuning range of ~4 nm at the maximum P_{IR} value of 1.04 mW for WGMs near 650 nm. Being heated by the IR laser, the droplet first shrinks down to a smaller volume and then swells back to its initial size again. Consequently, the droplet takes on a particular size twice; first during shrinking, then during swelling. These two sweeps over the accessible sizes are colored blue (solid curve) and red (dashed curve) for increasing and decreasing P_{IR}, respectively. During the sweep, the WGM resonance for the 532 nm pump light becomes satisfied at a certain size. This results in much more efficient pumping of the Raman gain medium, giving rise to lasing. Among the sizes swept, lasing occurs at the size corresponding to a P_{IR} of 0.55 mW in the increasing-P_{IR} direction. In the decreasing-P_{IR} direction lasing occurs from 0.55 to 0.43 mW, during a larger part of the sweep. The inset compares spectra corresponding to P_{IR} of 0.55 mW and 0.49 mW recorded during the increasing and decreasing P_{IR} sweeps, respectively. As expected, the peaks in the spectra coincide exactly at 649.45 nm, indicating that lasing occurs at the same size, independently of where this size is approached from. The WGM peak remains exactly at 649.45 nm while P_{IR} is swept from 0.55 to 0.43 mW in the decreasing direction, indicating self-stabilization of the microdroplet size (not shown).

A hysteresis is clearly seen in the P_{IR} dependence of the total Raman lasing intensity, shown in Fig. 2. We attribute this hysteresis to the resonant heating mechanism that occurs when a WGM absorption becomes in resonance with the green laser at a definite droplet size. As the resonant size is approached from above, the evaporation rate becomes accelerated by the resonant heating. This causes the droplet to pass over the resonant size too rapidly, resulting in an instantaneous lasing only. However, when the resonant size is approached from below, self-stable operation points can be found where Raman lasing can be sustained for long time periods. At such an operation point the resonant heating resists size changes by decelerating (accelerating) the condensation rate upon an increase (decrease) in the microdroplet size. It is therefore necessary to start from a smaller size and gradually decrease P_{IR} in bringing the droplet to a self-stable operation point.

Self-stabilization requires spectral tuning over a half-linewidth of the absorbing WGM at 532 nm. Assuming a quality factor of 10^4, this translates to a temperature change (ΔT) of approximately 1 mK in the microdroplet, for the relative humidity used in our experiments [14,15]. First-principle calculations assuming laser absorption through the diameter of the microdroplet [16] reveal green pump power necessary to change the microdroplet temperature by 1 mK to be 7 mW. Although this level of green laser power is available in our experiments, heat generated during the Raman conversion process should also be taken into account [17]. For a 14 μm diameter droplet, the Raman conversion rate necessary to increase the microdroplet temperature by 1 mK corresponds to ~20 nW of average power Raman lasing at 650 nm. Considering the total CCD counts detected from a WGM exhibiting Raman lasing (typically 10^6 counts/s), the low digital conversion gain used in the experiments, the overall detection efficiency of the experimental setup, and the low collection efficiency of the WGMs because of detection in the inverted geometry, we think that heating due to Raman conversion cannot be ignored in the present resonant heating mechanism, especially at large lasing intensities.

Figure 3(a) shows the consecutive spectra of 25 min of Raman lasing recorded from a 14 μm diameter droplet. Lasing occurs at the intense WGM peak at 651.6 nm that lies within the Raman band of water for 532 nm excitation. Recording spectra covering the whole Raman band enabled us to check whether lasing occurs at other spectral locations, and, as is the case with the droplet of Fig. 3(a), we observed lasing mostly at a single wavelength. The stable spectral position of the lasing wavelength shows that the droplet remained in its resonant size during the whole measurement. The maximum intensity of the consecutive spectra and the P_{IR} are plotted against time on the left and right vertical axes of Fig. 3(b), respectively. The fast decrease of P_{IR} appearing in the beginning of the measurement is part of the sinusoidal sweep, where no lasing size has been met yet. Once lasing is obtained, the sweep of P_{IR} is automatically stopped. From that point on, P_{IR} is adjusted manually to compensate for the long time fluctuations due to temperature variations of the chamber. After recording sufficient data, the experiment is stopped manually by blocking the pump laser. The
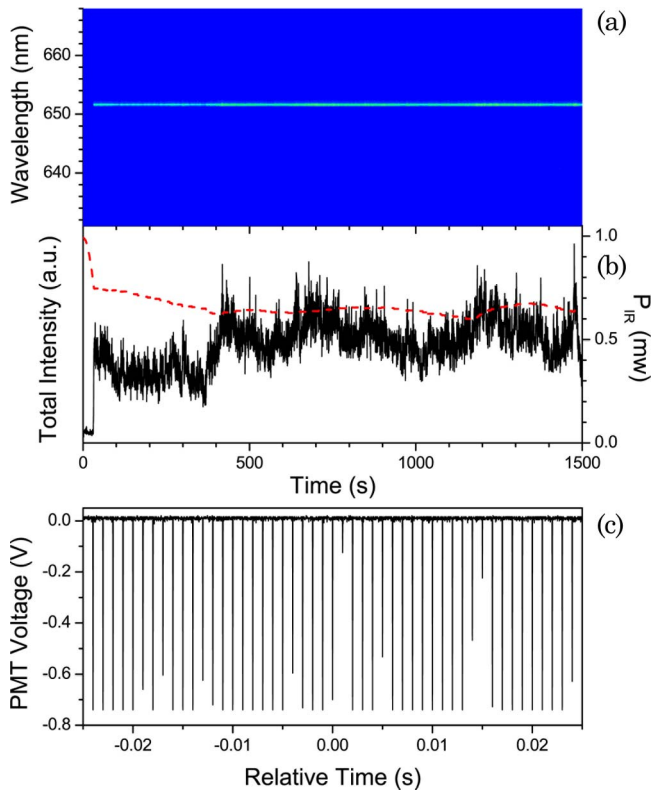
Fig. 3.   (Color online) (a) Consecutive spectra showing prolonged Raman lasing from a 14 μm diameter droplet. (b) Maximum intensity of each spectrum in (a) (black solid) and the power of the IR laser (red dashed) during the lasing period. (c) Oscilloscope trace showing pulse train of Raman lasing signal from a 19 μm diameter droplet.

lasing intensity also exhibits fast fluctuations that we attribute mainly to the nonnegligible heat release during Raman conversion.

The oscilloscope trace recorded from a 19 μm diameter droplet in the presence of Raman lasing shows laser pulses separated by 1 ms in accordance with the pump laser repetition rate [Fig. 3(c)]. Because of the narrow spectral filtering used, the background intensity due to spontaneous Raman scattering was negligible at this scale in the absence of Raman lasing. In this figure, variations in pulse amplitudes also indicate fast fluctuations in Raman lasing intensity.

In conclusion, using a self-stabilization mechanism we have been able to sustain Raman lasing from microdroplets standing on a superhydrophobic surface for as long

as 25 min. Our technique puts no fundamental limitations on the lasing duration. To our knowledge, this is the first observation of such prolonged periods of Raman lasing from individual microdroplets. This demonstration can inspire novel microdroplet-based cost-effective light sources for short-haul communication devices. It may also permit other nonlinear optics applications using microdroplets such as parametric conversion or frequency comb generation [5–7].

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