Raman lasing in glycerol water microdroplets on a superhydrophobic surface

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Abstract: We report on the first observation of Raman lasing near 630 nm from 532-nm-pumped, glycerol-water microdroplets on a superhydrophobic surface. Results of cavity-enhanced Raman scattering and Raman lasing experiments are described.

Summary

Potential applications in cavity quantum electrodynamics, optical communications, and chemical analysis have initiated a great deal of interest in the development of compact, microcavity-based sources of light[1-4]. When fabricated with high quality (Q) factors, these structures can give a large effective interaction length between the pump and the whispering gallery modes (WGM) and can enable low-threshold optical oscillation. Lasing from microcavities can be obtained either by introducing an active gain medium such as quantum dots [5] or by using a nonlinear gain mechanism such as the Raman effect [2]. One particular advantage of Raman lasing is that the output wavelength can be continuously adjusted over a broad range within the transparency range of the material by varying the excitation wavelength. Raman lasing has already been demonstrated in solid-state microcavities of various geometries and materials [2]. In addition to solid-state microcavities, cavity-enhanced Raman scattering (CERS) has also been demonstrated in liquid microcavities prepared in the form of microdroplet streams [6], electrodynamically levitated microdroplets [7], and optically trapped microdroplets[8]. However, to date, Raman lasing has not been reported from liquid microdroplets. Furthermore, resonant Raman emission has not been studied in stationary microdroplets on a surface. Two challenges need to be overcome in order to achieve Raman lasing from stationary microdroplets. First, a suitable hydrophobic surface is needed to minimize the geometric deformation of the microdroplets. Second, the microdroplets formed on the surface are required to have high Q factors for the WGMs so that oscillation can be achieved with reasonable pumping intensities.

In this work, we report on the first observation of Raman lasing from stationary glycerol/water microdroplets on a superhydrophobic surface[9]. The superhydrophobic surface was formed by spin coating a glass substrate with hydrophobically coated silica nanoparticles. The stationary microdroplets were excited at 532 nm. Two different regimes of operation were observed in the experiments: CERS and Raman lasing. During CERS, enhanced peaks were observed at the resonant frequencies of the WGMs. In the case of Raman lasing, the contrast ratio between the Raman peak near 630 nm and the background was larger than 30 dB. Raman lasing was not sustained and oscillation occurred in the form of temporally separated bursts. Observations suggest that the ‘on-off’ behavior is possibly due to the limited rate of heat removal. Increasing the rate of convective cooling via nitrogen purging was found to reduce the inter-burst separation from 2.3 to 0.4 seconds.

In the experiments, either an atomizer, or an ultrasonic nebulizer was used to spray glycerol/water microdroplets onto the superhydrophobic surface. Once on the surface, microdroplets quickly evaporated and reached their equilibrium sizes under the ambient humidity. A Q-switched, frequency-doubled Nd:YAG laser (λ=532 nm, pulse width=100 ns, pulse repetition rate=1 kHz, average power= 60 mW) was used for excitation. Excitation beam was focused to an estimated 34-μm-diameter spot by using a 0.88-NA, 60X microscope objective. To measure the Raman spectra, scattered light was passed through a long-pass filter that blocked the pump, dispersed in a monochromator having a resolution of 0.24 nm, and monitored with a CCD camera.

Cavity-enhanced Raman scattering was observed in smaller microdroplets generated using the ultrasonic nebulizer as shown in Fig. 1. Here, the spectrum of a 13-μm-diameter microdroplet is shown. The total exposure time was 45 sec. As can be seen, WGMs are clearly visible on top of the Raman emission bands of the glycerol-water solution. Despite the high excitation power, none of the WGMs dominated the spectrum and hence no clear evidence for Raman lasing was observed in this case. We believe that the absence of lasing is due to the low Q
factors of the WGMs of smaller microdroplets. Based on the spectral measurements, the Q factors were estimated to be around ~2000 in this case.

![Cavity-enhanced Raman scattering spectrum of a 13-µm-diameter glycerol-water microdroplet.](image)

Fig. 1. Cavity-enhanced Raman scattering spectrum of a 13-µm-diameter glycerol-water microdroplet.

Raman lasing was routinely observed in larger microdroplets generated with the atomizer. As an example, Fig. 2 shows the spectra obtained from a 15-µm-diameter microdroplet, showing Raman lasing. During consecutive acquisitions, intense WGM emission is momentarily observed in the high gain region of the Raman bands at 632.3 nm (See Fig. 2(a)). This is a clear indication of Raman lasing. Raman lasing is not sustained, and the intensity of the lasing WGM drops by more than 30 dB when lasing ceases (Fig. 2(b)). Despite the “on/off” behavior, the spectral location of the lasing WGM remains stable, indicating a constant droplet size within the spectral resolution of the measurement setup.

![Raman laser spectrum of a 15-µm-diameter glycerol-water microdroplet. (b) Corresponding spectrum during the non-lasing period.](image)

Fig. 2. (a) Raman laser spectrum of a 15-µm-diameter glycerol-water microdroplet. (b) Corresponding spectrum during the non-lasing period.

In order to investigate the origin of Raman lasing, the Raman spectra of glycerol-water mixtures of various compositions were measured. Figure 3 shows the acquired spectra for solutions containing 100, 75, 50, 13 vol. % water. As can be seen, the band centered around 630 nm originates from glycerol and dominates in the mixture containing 13 vol. % water. After reaching the equilibrium sizes, the investigated microdroplets have a Raman spectrum similar to that in Fig. 3(d). Hence, this indicates that Raman lasing occurs due to the presence of glycerol which has a larger Raman gain than that of water in the 620-660 nm spectral window (see Fig. 3).
The “on/off” behavior was observed in all microdroplets that exhibited Raman lasing. This phenomenon is possibly due to thermally induced density fluctuations during lasing. As a result, the circulating mode acquires a spatially distorted phase and can no longer satisfy the resonance condition. During the “off” period, excess generated heat is dissipated and Raman lasing resumes once the microdroplet cools down. To further understand the nature of the on/off behavior, we investigated the effect of nitrogen purging on the inter-burst separation. Results show that whereas the average inter-burst separation is around 2.3 seconds without purging, it is reduced to 0.4 seconds during purging. We believe that this is due to an increase in the rate of convective cooling.

In conclusion, we have observed CERS and Raman lasing from stationary, glycerol-water microdroplets situated on a superhydrophobic surface. Measurements gave clear indication of Raman lasing within the Raman band of glycerol, not previously observed in other studies with microdroplets. The contrast ratio of the Raman laser signal was larger than 30 dB. Raman lasing was not sustained and occurred in temporally separated bursts. We note that the novel configuration based on the superhydrophobic surface does not require complicated trapping methods such as electrodynamic levitation and optical trapping[7, 8]. Finally, the system described here can be excited by commercially available neodymium-based pump lasers and could potentially become a compact, cost-effective light source for short-haul communications systems.

This work was supported by the Scientific and Technological Research Council of Turkey (Grant No. TÜBİTAK-105T500). The authors thank H. Kalaycioglu and S. Doğanay for their assistance. The authors are also grateful to the Alexander von Humboldt Foundation for the donated equipment. A. Kiraz further 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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