Levitated droplet dye laser

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Abstract: We present the first observation, to our knowledge, of lasing from a levitated, dye droplet. The levitated droplets are created by computer controlled pico-liter dispensing into one of the nodes of a standing ultrasonic wave (100 kHz), where the droplet is trapped. The free hanging droplet forms a high quality optical resonator. Our 750 nL lasing droplets consist of Rhodamine 6G dissolved in ethylene glycol, at a concentration of 0.02 M. The droplets are optically pumped at 532 nm light from a pulsed, frequency doubled Nd:YAG laser, and the dye laser emission is analyzed by a fixed grating spectrometer. With this setup we have achieved reproducible lasing spectra in the visible wavelength range from 610 nm to 650 nm. The levitated droplet technique has previously successfully been applied for a variety of bio-analytical applications at single cell level. In combination with the lasing droplets, the capability of this high precision setup has potential applications within highly sensitive intra-cavity absorbance detection.

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1. Introduction

Optical micro cavities in general are receiving considerable interest due to their size and geometry dependent resonant frequency spectrum and the variety of applications [1]. Micro-droplets constitute an interesting medium for micro-cavity optics in terms of e.g. lasing emission properties [2] and manifestation of chaotic wave dynamics [3, 4]. Combined with ultrasonic levitation techniques [5] micro-droplets may also provide an interesting environment for analytical chemistry [6] including intra-cavity surface-enhanced Raman spectroscopy [7, 8].

Lasing in freely falling droplets was first reported by Qian *et al.* [2] which stimulated a significant interest in the optical properties of droplets, see e.g. Datsyuk [9] and references therein. Parallel to this there has been a significant attention to ultrasonic levitation [5] from the chemical community [6] allowing for studies of e.g. protein crystallization [10]. Combining ultrasonic levitation with the optical properties of droplets holds great promise for highly sensitive intra-cavity absorbance detection system with prospects for single-molecule detection [8]. However, such applications rely heavily on both reproducible loading of droplets and subsequent reproducible generation of laser emission spectra by external pumping.

In this paper we present the first observation, to our knowledge, of reproducible lasing from levitated 750 nl dye droplets. Droplets are optically pumped at 532 nm by a pulsed, frequency doubled Nd:YAG laser and emission is analyzed by fixed grating spectrometry. The levitated droplet constitutes a highly sensitive intra-cavity absorbance detection system with prospects for single-molecule detection [8] and the possibility for computer-generated on-demand droplets holds great promise for applications in high-throughput chemical analysis.
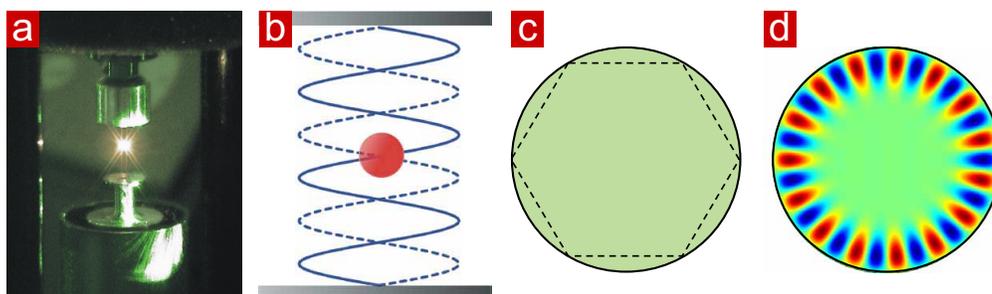

Fig. 1. a) Photograph of a lasing levitated micro-droplet. b) Schematics of ultrasonic field with the micro-droplet being trapped at a node in the ultrasonic field. c) Schematics of whispering-gallery modes in a (2D) spherical cavity. d) Numerical example of a whispering-gallery mode in a (2D) spherical cavity.
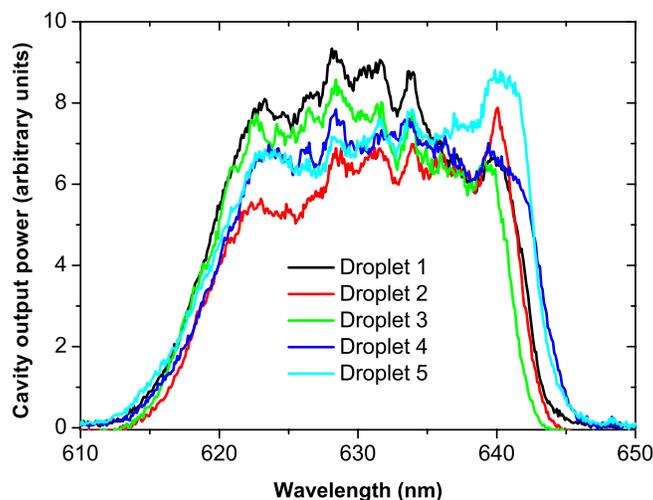
Fig. 2. Reproducible lasing spectra from dye doped micro-droplets. Each spectrum is obtained in a fixed setup by a well-controlled loading of an EG droplet with a Rh6G dye which is subsequently pumped above threshold, see Fig. 3(b). The spectra are averaged over three pump pulses.

2. Experimental setup and results

Ultrasonic levitation is a technique that facilitates the performance of a variety of investigations on small volumes of samples, i.e. liquid droplets and particles. It suspends the object levitated in the nodal point of an ultrasonic standing wave, see Fig. 1(b). The technique was introduced in the 1930's and does not rely on any specific properties of the sample except size and mass. The method has been used extensively in bio-analytical and analytical chemistry applications, see e.g. [6, 11] and references therein.

The ultrasonic levitator [12] consists of an ultrasonic transducer and a solid reflector supporting standing waves, see Fig. 1(a) and (b). In this work the levitator is operated at a frequency of $\Omega_{vib}/2\pi \sim 100\,$kHz corresponding to a wavelength of $\Lambda \sim 6\,$mm. The levitator can hold particles of radius $a < \Lambda$ and droplets with $a \sim \Lambda/6$ require a minimum of ultrasonic power. Large droplets may be deformed by the body forces (gravity and ultrasonic pressure gradients), which in practice limits the droplet size to $a < \xi$ where the capillary length ξ is in the millimeter range for a water droplet. Furthermore, the droplet shape may also be spherically deformed by applying a large ultrasonic pressure amplitude.

In our experiments we used Rhodamine 6G (Rh6G) laser dye dissolved in ethylene-glycole (EG). The liquid sample was placed in a nodal point of the levitator by means of a computer controlled piezo-electric micro dispenser [13]. A droplet with a total volume of $V = (4\pi/3)a^3 \sim 750\,$nl was formed by repeated addition of pL drops.

The levitated dye droplet was optically pumped by a pulsed, frequency-doubled Nd:YAG laser ($\lambda = 532\,$nm) with a pulse length of 5 ns and a repetition rate of 10 Hz. The light emitted from the micro-droplet was collected by an optical fiber placed at an angle of approximately 50 degrees relative to the pump laser beam. The emission was analyzed in a fixed-grating spectrometer with a resolution of 0.15 nm.

Evaporation and dye bleaching could hinder the applicability of the dye droplet as a lasing device. In order to minimize these effects, we used a measurement scheme, where nominally

identical droplets are loaded consecutively, and each droplet is only pumped with 100 pulses (corresponding to a duration of 10 s) from the Nd:YAG laser, before it is replaced by the next droplet. We have not systematically investigated the performance of the dye droplet lasers for more than 100 pulses.

In Fig. 2 we show 5 emission spectra obtained from normally identical $V = 750$ nl EG droplets, with a Rh6G concentration of 2×10^{-2} mol/L. The laser was pumped above the threshold. The observed variations in output power are attributed to fluctuations in the pump power.

In a second measurement series we demonstrate the lasing action of the dye droplet where consecutively loaded droplets are pumped at different average pump power. In Fig. 3(a) we
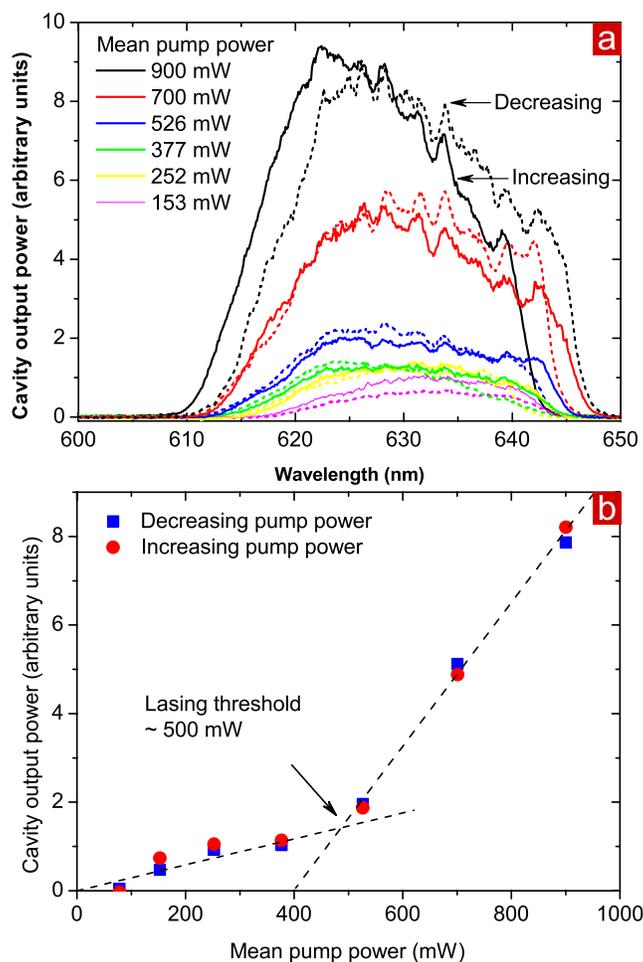

Fig. 3. a) Cavity output power for increasing and decreasing average pump power. Each spectrum is obtained in a fixed setup by pumping an EG droplet with a Rh6G dye. The pump power is first increased from zero up to level around 1000 mW (dashed curves) and subsequently again lowered (solid curves). The spectra are averaged over three pump pulses. b) Cavity output power versus mean pump power. The dashed lines are guides to the eyes indicating a lasing threshold of around 500 mW in the mean pump power.

show the measured spectra and in Fig. 3(b) we show the dye droplet output power versus pump power. In the measurement sequence the pump power was first increased from 150 mW to 900 mW and subsequently decreased again. The reproducibility of the obtained spectra and the lasing threshold is seen from panels a and b, respectively.

3. Discussion

In the following we briefly address the optical and mechanical modes, to assess the influence on the optical performance of levitated droplet dye lasers and their applications for intra-cavity sensing.

3.1. Optical modes

For a spherical resonator the whispering-gallery modes (WGMs) are characterized by their radial quantum number p as well as by their angular momentum quantum number ℓ and the azimuthal quantum number m which can have $(2\ell+1)$ values, i.e. $\omega_{p\ell}$ has a $(2\ell+1)$ degeneracy [8]. For the lowest-order radial modes ($p=1$), see Fig. 1(d), the resonances are to a good approximation given by $n\chi \sim \ell$ where $\chi = ka$ is the so-called dimensionless size parameter, n is the refractive index, a is the radius of the droplet, and $k = \omega/c = 2\pi/\lambda$ is the free-space wave vector. The modes are thus equally spaced in frequency and the corresponding spacing in wavelength is [8]

$$\Delta\lambda = \left|\frac{\partial\lambda}{\partial\chi}\right|\Delta\chi \simeq \frac{\lambda^2}{2\pi a}\frac{\tan^{-1}(n^2-1)^{1/2}}{(n^2-1)^{1/2}},\tag{1}$$

where the last fraction indeed has a $1/n$ asymptotic dependence for a large refractive index, $n \gg 1$. For the EG droplets in the experiments the corresponding mode spacing is of the order $\Delta\lambda \sim 0.1$ nm which is not resolved in our present experiments. However, $V = 100$ pL droplets ($a \sim 30\,\mu$m) have been achieved in different experiments by operating the levitator at 100 kHz. This would increase the mode spacing to $\Delta\lambda \sim 1$ nm.

3.2. Droplet shape and mechanical modes

The shape of the droplet may be understood from Gibbs free-energy arguments where one naturally defines the characteristic capillary length $\xi = \sqrt{\gamma/\rho g}$ with γ being the surface tension, ρ is the liquid mass density, and g is magnitude of the gravitational field [14]. A droplet with a characteristic size $a \ll \xi$ will have a shape strongly determined by the surface tension and a spherical shape will then obviously minimize the Gibbs free energy. For a free water droplet in air $\xi \sim 2.7$ mm. In our experiments, where $a \sim 500\,\mu$m, the spherical shape is not perturbed significantly by body forces.

We emphasize that in principle the levitated droplet is a complicated dynamical system, see e.g. [15]. However, as analyzed already by Rayleigh in 1879 [14] the vibrational spectrum of a liquid droplet originates from two classes of modes; surface-tension driven surface modes and compression-driven compressional modes. Since the liquid can be considered incompressible the latter class of modes is in the high-frequency regime while the low-frequency response is due to surface-shape oscillations conserving the volume. The surface vibrational modes are similar to the optical WGMs and are characterized by their angular momentum quantum number ℓ_{vib}. For low amplitude oscillations Rayleigh found that [14]

$$\omega_{\text{vib}} = \sqrt{\frac{\gamma\ell_{\text{vib}}(\ell_{\text{vib}}-1)(\ell_{\text{vib}}+2)}{\rho a^3}}, \ell_{\text{vib}} = 2,3,4,\ldots\tag{2}$$

A droplet of a given radius a can thus not be vibrationally excited by the ultrasonic pressure field at frequencies $\Omega_{\text{vib}} < \sqrt{8\gamma/\rho a^3}$. For a driving frequency of 100 kHz this implies that water droplets of radius below $10\,\mu$m are not vibrationally excited.

3.3. Prospects for intra-cavity sensing

The prospects for intra-cavity sensing in liquid dye droplets correlate strongly with the optical cavity Q factor. The WGMs each have a resonant frequency ω with a width $\delta\omega = 1/\tau$ where τ is the lifetime of a photon in the mode. The corresponding quality factor $Q = \omega/\delta\omega$ of WGMs is determined by several factors including intrinsic radiative losses originating from the finite curvature of the droplet surface Q_{rad}, absorption due to the cavity medium Q_{abs}, and broadening of resonances due to vibrational interaction with the ultrasonic field Q_{vib}, i.e.

$$Q^{-1} = Q_{\text{rad}}^{-1} + Q_{\text{abs}}^{-1} + Q_{\text{vib}}^{-1}. \tag{3}$$

For the radiative loss Q_{rad} increases close to exponentially with the size parameter χ and for a refractive index $n \sim 1.45$ we have $Q_{\text{rad}} \sim 10^5$ and $Q_{\text{rad}} \sim 10^{12}$ for χ equal to 50 and 100, respectively [16]. In the case of bulk absorption we have [17]

$$Q_{\text{abs}} = \frac{2\pi n}{\alpha\lambda} = \frac{\chi}{\alpha a} \tag{4}$$

where α is the absorption coefficient of the cavity medium.

The broadening of the WGMs by interaction with the vibrational modes is complicated, but we may immediately disregard the high-frequency compressional modes leaving only the low-frequency surface-tension driven modes for concern. Even though the realistically attainable droplet ($a > 40\,\mu$m) will be vibrationally excited by the ultrasonic pressure field, the influence of the disturbance could be suppressed by short pump pulse operation. Finally, static, deformations of the cavity may give rise to partial chaotic ray dynamics with a universal, frequency independent, broadening of the WGM resonances which will decrease Q_{rad} [3, 4]. A similar vibration-induced decrease of Q_{rad} is expected.

4. Conclusion

In conclusion we have demonstrated a reproducible laser action from an ultrasonically levitated laser dye droplet, when nominally identical 750 nL droplets are consecutively loaded and optically pumped by a pulsed frequency-doubled Nd:YAG laser. The present droplets show reproducible multi-mode lasing. This system is considered a potential candidate for intra-cavity sensing, and the limitations induced by ultrasonic field were discussed.