Simple technique for scattering experiments of submillimeter droplets

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A simple technique is presented of allowing the generation of submillimeter-sized single droplets for scattering experiments. Multiorder Raman spectra (including Stokes, anti-Stokes, and combination emission) from a liquid CCl_4 spherical droplet formed by a hollow glass fiber is also obtained by this technique, with a frequency-doubled Nd:YAG pulsed pumping (532 nm). © *1995 American Institute of Physics.*

Spherical resonators have been widely used because of the very high quality factor (Q value) of electromagnetic whispering-gallery-mode (WGM)[1] or morphology-dependent resonances (MDR) in the dielectric sphere with micrometer-sized droplets[2,3] or solid particles.[4–6]

In this note, we report a simple technique for scattering experiments of submillimeter-sized single droplets, whereby the multiorder Raman spectra (MRS) (including Stokes and anti-Stokes emission) from a liquid CCl_4 droplet formed by a hollow glass fiber are observed.

The liquid CCl_4 spheres are formed, by a surface tension of the material itself, around a hollow glass fiber (micropipet) having ~1 cm length and 10 μm outer diameter that was pulled in our laboratory. The micropipet was prepared by pulling with heating a glass tube having ~1 mm outer diameter (o.d.) and 0.5 mm inner diameter (i.d.). The CCl_4 spheres are suspended around the micropipet by pushing out the CCl_4 liquid, using a connected syringe.

Figure 1(a) shows a microphotograph of a single CCl_4 droplet (~400 μm diameter) suspended at a nib (which is an exit of the pushed CCl_4 liquid) of the micropipet called the "straight-shank micropipet"). Faster pulling of the glass tube made the pipet taper in diameter along its axis and resulted in creeping of the liquid droplet toward a larger diameter along the outer wall of the pipet [Fig. 1(b)].

The arrangement for the MRS experiments is similar to those described previously;[6] however, the present experiment substitutes a frequency-doubled Nd:YAG pulsed-laser [Nd:YAG (*SH*); 532 nm] pumping with ~3 mJ pulse energy and 12 ns pulse duration for a UV N_2 laser (337 nm) to excite spherical CCl_4 droplets.

Figure 2 shows single-shot MRS spectrum obtained from a single CCl_4 droplet with 415 μm diameter, including Stokes (with the −), anti-Stokes (with the + signs), and combination emission, corresponding to the various modes of CCl_4.

The well-known Raman shifts of the three strong CCl_4 peaks are ν_1=459 cm^{-1}, ν_2=218 cm^{-1}, and ν_4=314 cm^{-1}. In the spectral results exhibited in Fig. 2, the *SH* corresponds to the elastic spectrum of pumping Nd:YAG *SH* light (532 nm), and the number n (with the vertical line across the top and bottom in the figure) corresponds to the nth order of the CCl_4 ν_1 mode, in which the numerals with the negative (−) or positive (+) signs denote the Stokes (−) or anti-Stokes (+) lines, respectively. Moreover, the respective nth-order modes consist of three modes, a, b, and c, which correspond to the ν_1, ν_2, and ν_4 modes of CCl_4, respectively.

Figure 2 also shows that additional combination modes of the three strong CCl_4 modes (ν_1, ν_2, ν_4) occur, which are listed in Table I.

The observation of multiorder Raman spectra (MRS) including Stokes, anti-Stokes,[7] and multiorder combination and sum frequencies[8] in the CCl_4 droplet seems to be analo-

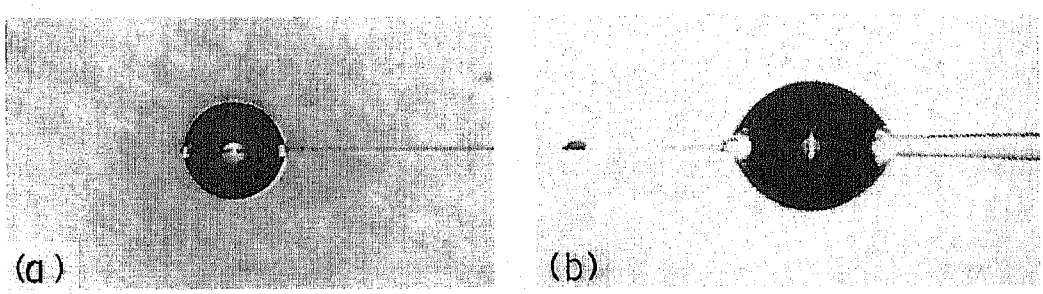

FIG. 1. Micrograph of a CCl_4 droplet (~400 μm diameter) suspended at a nib of the (a) straight-shank micropipet and (b) tapered pipet.

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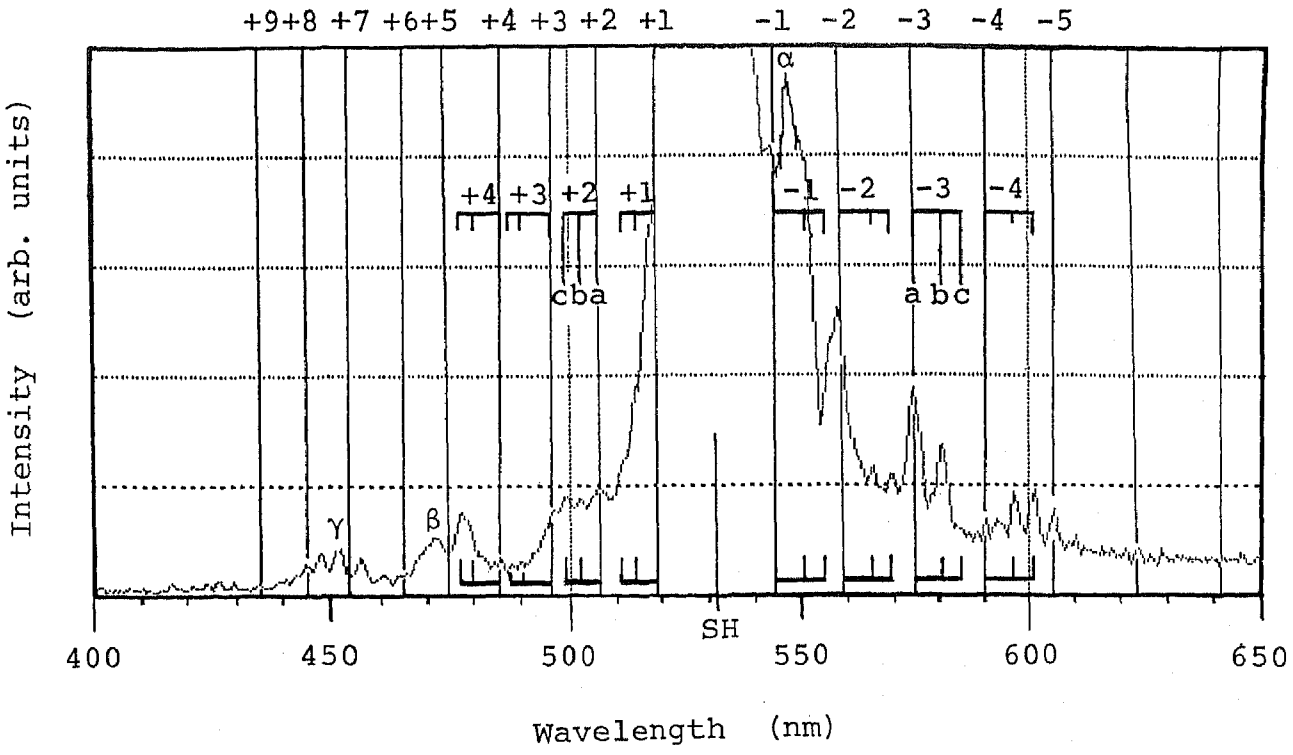

FIG. 2. Single-shot MRS spectrum obtained from a single CCl_4 droplet with 415 μm diameter. *SH*: the elastic spectrum of pumping Nd:YAG *SH* light (532 nm), and the number n (with the vertical line across the top and bottom in the figure) corresponds to the nth order of the CCl_4 ν_1 mode, in which the numerals with the negative (−) or positive (+) signs denote the Stokes (−) or anti-Stokes (+) lines, respectively. Moreover, the respective nth-order modes consist of three modes, *a*, *b*, and *c*, which correspond to the ν_1, ν_2, and ν_4 modes of CCl_2, respectively. Combination modes α, β, and γ correspond to $\nu_{SH}-(\nu_2+\nu_4)$, $\nu_{SH}+4\nu_1+(\nu_2+\nu_4)$, and $\nu_{SH}+6\nu_1+(\nu_2+\nu_4)$, respectively, where ν_{SH} denotes the *SH* frequency (532 nm).

TABLE I. Line assignments of the observed combination spectra.

Symbols[a]	Assignments[b]
Stokes region	
α	$\nu_{SH}-(\nu_2+\nu_4)$
Anti-Stokes region	
β	$\nu_{SH}+4\nu_1+(\nu_2+\nu_4)$
γ	$\nu_{SH}+6\nu_1+(\nu_2+\nu_4)$
(right-hand side of γ)	$\nu_{SH}+6\nu_1+\nu_4$
(left-hand side of γ)	$\nu_{SH}+7\nu_1+\nu_4$

[a]Symbols are attached to the spectra in Fig. 2.
[b]ν_{SH}, ν_1, ν_2, and ν_4 correspond to the *SH* (532 nm) and the three strong CCl_4 Raman modes, respectively.

gous to the previous effects based on MDRs; however, the present observation is due to a simple technique.

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