Stimulated Raman scattering of a high-\(Q\) liquid-hydrogen droplet in the ultraviolet region

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We show that a liquid-hydrogen droplet can achieve high-\(Q\) values that exceed \(10^8\) for whispering-gallery modes in the ultraviolet. We show also that pumping high-\(Q\) liquid-hydrogen droplets with ultraviolet laser radiation generates many vibrational and rotational Raman sidebands that cover a broad spectral range from the ultraviolet to the near infrared. © 2002 Optical Society of America

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Droplets (microspheres) act as high-\(Q\) optical resonators through whispering-gallery modes (WGMs). Although many optical processes have been reported that use various kinds of droplet, reports have addressed chiefly visible and near-IR operating wavelengths.1,2 This is so essentially because the conventional media for which the droplets are found have nonnegligible Rayleigh scattering and photoabsorption losses in the UV. These losses may inhibit the possibility of achieving high-\(Q\) values. In this Letter we show that, by using liquid hydrogen as the droplet medium, one can achieve a high-\(Q\) value that exceeds \(10^8\) for WGMs, even in the UV. We show also that a high-\(Q\) liquid-hydrogen droplet (LHD) can generate stimulated Raman scattering (SRS) sidebands that cover a broad spectral range from the UV to the near IR.

Liquid hydrogen is the simplest molecular liquid and consists of \(H_2\) molecules. A unique property of liquid hydrogen is that the \(H_2\) molecules have well-defined vibrational and rotational quantum states, as for free molecules in the gas phase, and their characteristic frequencies are almost equal to the gas-phase values.3 Moreover, the widths for the Raman transitions are narrower than those for conventional liquids; e.g., the vibrational Raman width is narrower than \(0.1\) \(\text{cm}^{-1}\) HWHM and the rotational Raman width is \(1.4\) \(\text{cm}^{-1}\) HWHM.4

Figure 1(a) illustrates the interaction scheme for SRS in liquid hydrogen. SRS occurs for a pure vibrational transition of \(Q_1(0)\) \((v = 1-0, J = 0-0)\) and for a pure rotational transition of \(S_0(0)\) \((v = 0-0, J = 2-0)\) for the \(H_2\) molecule. The Raman shifts for the \(Q_1(0)\) and \(S_0(0)\) transitions are 4151.4 and 353.3 \(\text{cm}^{-1}\), respectively.5 The pump radiation is marked \(p\), and the first Stokes radiation for the \(Q_1(0)\) and the \(S_0(0)\) transitions is marked \(v_{-1}\) and \(r_{-1}\), respectively. Note that the electronic excited states of liquid hydrogen are located more than \(90,000\) \(\text{cm}^{-1}\) above the ground state, as in the gas phase. This situation results in good transparency for liquid hydrogen up to the vacuum-UV region and in the smallest refractive index among molecular liquids.

The method of producing a LHD is essentially the same as that described previously.5 The droplet was produced in an optical cell under cryogenic conditions. Liquid hydrogen [nearly pure (\(\geq 99.9\%\)) parahydrogen] was fed through a fused-quartz capillary with an outer diameter of \(20\) \(\mu\text{m}\) such that the droplet appeared at the edge of the capillary. A photograph of a typical droplet is displayed in Fig. 1(b). The droplet shows a spheroidal shape with an equator diameter of \(400\) \(\mu\text{m}\).

In the research reported here we carried out experiments with LHDs of \(400-500-\mu\text{m}\) diameters.

The SRS experiments were carried out with two single-longitudinal-mode pulsed lasers in the UV; one at 266 nm and the other at 202 nm. The 266-nm laser was a frequency-quadrupled \(Q\)-switched YAG laser (Hoya-Continuum Model Powerlite 9010) that was injection seeded by a cw single-longitudinal-mode YAG laser. The 202-nm laser radiation was generated by sum-frequency mixing of the 266-nm laser radiation with single-longitudinal-mode optical parametric oscillator laser radiation at 830 nm (Hoya-Continuum 0146-9592/02/060421-03$15.00/0 © 2002 Optical Society of America

Fig. 1. (a) Energy diagram for the two SRS processes: a pure vibrational transition \(Q_1(0)\) \((v = 1-0, j = 0-0)\) and a pure rotational transition \(S_0(0)\) \((v = 0-0, j = 2-0)\) for the \(H_2\) molecule. The pump field is denoted \(p\) and the first Stokes fields for the \(Q_1(0)\) and \(S_0(0)\) transitions are denoted by \(v_{-1}\) and \(r_{-1}\), respectively. Raman shifts for the \(Q_1(0)\) and \(S_0(0)\) transitions are 4151.4 and 353.3 \(\text{cm}^{-1}\), respectively. (b) Typical photograph of a LHD hung at the edge of a quartz capillary with an outer diameter of \(20\) \(\mu\text{m}\).
Model Mirage 500) by use of a β-barium borate crystal. The pulse durations were 6 and 4 ns for the 266- and 202-nm lasers, respectively. The laser beam was vertically polarized and was focused upon the edge of the droplet’s equator with a spot diameter of 100 μm. We adjusted the focal point such that we could restrict the emission spot sharply to the equator by monitoring the emission pattern. The temporal and spectral behaviors of the SRS emissions were measured perpendicularly to the laser beam by two grating spectrometers. One spectrometer was used as a monochromator with a photomultiplier for temporal measurements; and the other, as a polychromator with a CCD detector (Andor Model DU420-BU) for spectral measurements. We also measured the emissions in the vacuum-UV by using a vacuum spectrometer with a windowless CCD detector (Roper Scientific Model SX-TE/400PB). All measurements were carried out on single-shot basis.

Figure 2 shows the temporal behavior of the $v_{-1}$ SRS sideband at 299 nm pumped by the 266-nm laser. The lighter curve denotes the experimental observation. The temporal behavior was observed just above the SRS threshold, so only the $v_{-1}$ component was generated. The pump energy was 15 μJ/pulse, corresponding to a focused intensity of 11 MW/cm². It can readily be seen that the SRS radiation in the droplet decays on a very long time scale compared with the pump-pulse width of 6 ns. The slow decay was not sensitive to the pump intensity, provided that the higher SRS sidebands were not generated. When the higher sidebands were generated, the decay profiles showed the various kinds of dynamic behavior reported in Ref. 5.

The observed slow decay means that the SRS fields generated in the droplet are resonant to WGMs and decay through the cavity lifetimes of the WGMs. We observed many spikes in the temporal profile, which were not reproducible shot by shot. The spikes could be due to beating between the SRS fields that are resonant with the various WGMs. We fitted the observed temporal profile by assuming that the two WGM resonances had the same cavity lifetime. The darker curve denotes the temporal profile that we calculated by setting the cavity lifetime and the mode separation to 660 ns and 18 MHz, respectively. The calculated curve reasonably reproduces the main features of the spikes observed in the experimental curve, revealing that the spikes are due to mode beating between the WGMs. From the fitted cavity lifetime of 660 ns we obtained a cavity $Q$ value of $4.2 \times 10^9$ for the LHD by using the relation $Q = \omega \tau$, where $\omega$ and $\tau$ are the angular frequency of the Stokes field and the cavity lifetime, respectively.

Although a higher $Q$ value of $8 \times 10^9$ was reported for the WGM of a fused-silica sphere in the visible, the present $Q$ value is the highest obtained so far in the UV, to the best of our knowledge. In Fig. 3 we plot $Q$ values versus wavelength for a LHD together with those for a silica sphere. The solid curve denotes the upper limit for the LHD $Q$ value, which we calculated by using the relation $Q^{-1} = \alpha \lambda/2 \pi n^2$, where $\alpha$ and $n$ denote the internal loss and the refractive index, respectively, of the droplet substrate at wavelength $\lambda$. We have assumed that the internal loss of LHD is limited by the Rayleigh-scattering loss. Photoabsorption loss was not included in the calculation because no photoabsorption data for liquid hydrogen were available. We estimated the refractive index of liquid hydrogen by using the empirical formula found in Ref. 10; the value at a wavelength of 300 nm was 1.126. The measured $Q$ values for LHD are marked by filled circles in Fig. 3. The value at 683 nm was taken from Ref. 5. The calculated curve shows the high $Q$ values, exceeding $10^{10}$, can be realized for LHDs even in the UV near 300 nm. The observation at 299 nm clearly demonstrates the superiority of LHD in the UV.
The dotted and dashed curves in Fig. 3 denote the upper limit for the silica spheres calculated by the Rayleigh-scattering loss and by the Rayleigh-scattering and the photoabsorption losses, respectively. Measured $Q$ values for the silica spheres are plotted by filled squares. Note that the Rayleigh limit of a silica sphere is more than 1 order of magnitude less than that of a LHD because of the inherently larger refractive index of silica. The difference becomes more prominent in the shorter-wavelength region (<400 nm), where photoabsorption becomes the main mechanism of loss for silica spheres.

It should be mentioned that the present $Q$ value at 299 nm is close to the value at 683 nm. This result may imply a limitation of the present decay-time measurement, the single-shot measurement just above the SRS threshold. Because the photon number in the WGM is limited at the threshold, when the cavity decay time has become quite long the emitted photon distribution for a single decay event should become sparse, as a result, the signal-to-noise ratio for the decay measurement would become worse, inhibiting quantitative measurements of the decay time. The measurements have led us to believe that the limit of the measurable longest decay time is in the range 500 ns–1 $\mu$s, corresponding to $Q$ values of the order of 10$^9$. We speculate that some WGM with higher $Q$ values might have been excited but that they could not be detected owing to measurement limitations.

Next we turn to SRS of the high-$Q$ LHD under strong pumping conditions. Figure 4 exhibits a typical spectrum pumped by 202-nm laser radiation. The spectrum was measured by two CCD polychromators at a pumping intensity of 120 $\mu$J/pulse (300 MW/cm$^2$). Many emission lines, covering the whole UV–near-IR range from 186 to 900 nm and corresponding to a frequency spread of 40,000 cm$^{-1}$, are clearly observable. All the emission lines were assigned as SRS sidebands that arise from both the vibrational $Q_1(0)$ and the rotational $S_0(0)$ sequences. Assignments are given as $v_q$ and $r_q$, where the integer $q$ ($q'$) denotes the order number of the vibrational (rotational) SRS sideband. The vibrational $Q_1(0)$ sequence was observed up to ninth order, $v_9$, located near 800 nm. Observation of the higher Stokes sidebands might be limited by the CCD sensitivity. For the $S_0(0)$ rotational sequence, sidebands were observed for each vibrational sequence, and the number of rotational sidebands increased for higher-order vibrational sequences. Rotational Stokes sidebands were observed up to eleventh order, $r_{11}$, from the sixth to the eight vibrational sequences. For the $S_0(0)$ anti-Stokes components, the sidebands were observed up to fifth order for the ninth vibrational sequence, but the intensities were much weaker than those for the Stokes sidebands. For the $Q_1(0)$ anti-Stokes sideband we observed only the first anti-Stokes component at 186 nm with much weaker intensity than for the Stokes sidebands.

It should be noted that such very wide SRS sequences that cover the whole UV–near-IR region have not to our knowledge been achieved in any droplet. Although we have not obtained a quantitative understanding of the detailed SRS characteristics in a high-$Q$ LHD, the features observed could be explained qualitatively through cascaded SRS gain and non-phase-matched four-wave mixing, as was done previously.

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