COMPARISON BETWEEN THE TEMPORAL CHARACTERISTICS OF PICOSECOND SRS FROM THE CELL AND SRO FROM THE DROPLET

Shixiong QIAN, Shu YUAN, Yufen LI
Physics Department, Fudan University, Shanghai, P.R. China

Hezhou WANG, Xuguang HUANG and Zhenxin YU
Ultrafast Laser Spectroscopy Laboratory, Zongsan University, Guan Zhou, P.R. China

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We have studied the temporal behavior of picosecond stimulated Raman scattering (SRS) from a liquid cell and stimulated Raman oscillation (SRO) from liquid droplets. Big differences have been found between the multiorder stimulated Raman emissions from the cell and the droplets. There is nearly no delay of SRS emission from the cell, but a long delay and slow decay of the Stokes SRO from the droplets. Basing on the basic properties of the droplet cavity, the experimental results were well explained.

1. Introduction

Recently the nonlinear Raman interaction inside micrometer liquid droplets has been studied, which include coherent Raman mixing (CRM) [1], stimulated Raman oscillation (SRO) [2–4] and coherent anti-Stokes Raman scattering (CARS). The unique properties of these processes in the droplets are the high efficiency, high-order of the Stokes SRO emission. From carbon tetrachloride droplets pumped by a 532 nm laser beam, Stokes emission with an order as high as 17 have been observed, which is much higher than that of SRS from the usual liquid cell [5]; a similar high-order Stokes SRO was obtained from other liquid droplets. With picosecond laser pumping, ps SRO was observed, but the threshold of the ps SRO from the droplet was much higher than that of ns SRO.

From the ns SRO and ps SRO experiment, the micrometer droplet demonstrates a behavior as a spherical resonant cavity which can maintain the oscillation inside this small volume [6,7]. Basically, there must be a large difference between SRS from a liquid cell and SRO from liquid droplets. In the cell, there is a travelling wave interaction with matter [8], but in the droplet, there is a standing wave interaction. It is interesting to investigate the dynamical behaviors of the SRS emission from the cell and SRO emission from the droplet. Zhang et al. have studied the time-resolved spectra of SRO from ethanol droplets and got some information about the photon time inside the droplet [9]. Pinnick et al. [10,11] and Hsieh et al. [12] used nanosecond laser pulses to study the SRO process in the droplet and have found that there was an obvious ns order delay between SRO emission and the pump pulse. In this paper, we report the research result of time-resolved high-order picosecond stimulated Raman emission from the cell and the droplets and have found that there was a ps order delay between SRO and the pump.

2. Experimental set-up

We used a SHG beam of a mode-locked YAG laser as the pump beam for our experimental research. The SHG pulse had a pulse duration of about 30 ps, pulse energy of 2 mJ. The beam was focused onto the droplets which were generated by a home-made device. The diameter of the droplets could be adjusted by the modulated frequency of the applied voltage on the piezoelectric ceramic and was in the range of 60–100 μm. The emission from the droplets was col-

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lected by two lenses and was sent to a monochromator which was followed by a streak camera. With this set-up, we could get the time-resolved spectra. The streak camera had several choices of the temporal resolution; in our experiment the time resolution was set to be 5 ps. The experimental set-up is given in fig. 1. With each laser pulse we could get the time-resolved spectrum of the emission from the sample. The streak camera had two windows. In the experiment, we used an optical fiber to transfer a part of the SHG beam into one of the windows as a reference and used the other window to detect a given order Stokes emission. The length of the optical fiber was about 60 cm, the location of the input side of this fiber was carefully adjusted to get the near equal length of the optical path with that of the SRO emission from the droplet or SRS from the cell. With this arrangement, we could easily compare the emitted Stokes emissions with the reference SHG signal to get the temporal information of the different Stokes emissions.

3. Results and discussion

In the experiment, we first tested the time-resolved pump signal from the optical fiber and that from the monochromator simultaneously to check whether the signal from the fiber could be used as a time reference in the experiment. We have found that for 532 nm pump, the wave profile from the fiber was nearly the same as that from the monochromator and the time relation between these two signals kept the same for different pulses. The time-resolved spectra are given in fig. 2. In some cases, when the pump pulse duration was changed by the variation of the mode-locked dye concentration, the pulse signal from the fiber followed this variation. This result confirmed that we could use the signal from the fiber as the reference and the fiber would not introduce any extra time delay to our experimental results.

From the experiment, we found that for SRS emissions from the benzene cell of 11 cm long, the first order Stokes emission occurred almost at the same time with the pump. There were other high-order Stokes and anti-Stokes emissions from the cell. The time delays among 3s (third Stokes), 2s, 1s, 1a (first

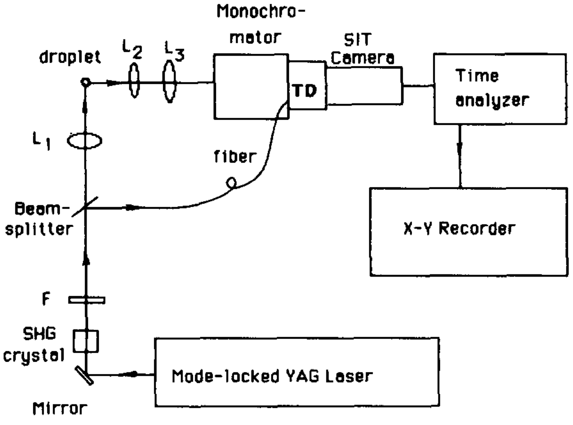

Fig. 1. Experimental set-up for transient SRS and SRO research.

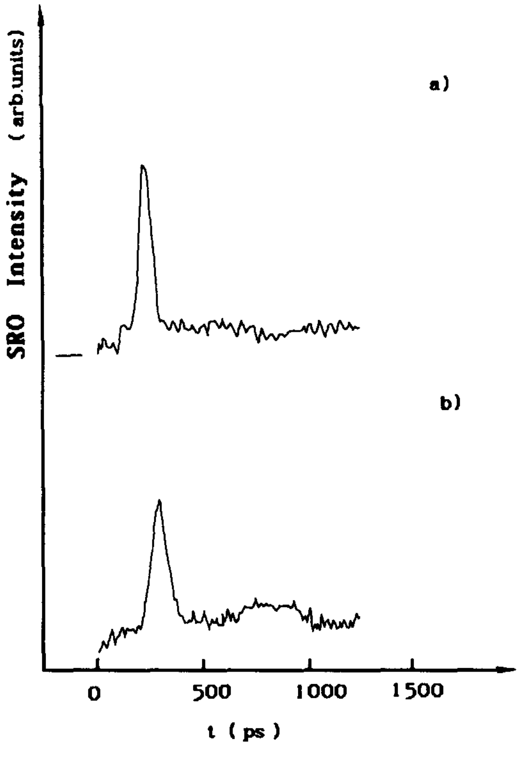

Fig. 2. Wave profile for the SHG signals (a) from the monochromator and (b) from the optical fiber for the same 30 ps pump pulse. The relative shift in the time scale between the two signals is due to the small difference between the optical length in the monochromator and fiber path. The result showed that the signal from the fiber reproduced the incident pulse profile and could be used as a time reference for the time-resolved experiment.

anti-Stokes), 2a and 3a SRS emissions were less than 20 ps which was less than the pulse duration of the pump. This result means that there is nearly no delay among SRS emissions within the experimental accuracy.

For the droplet, however, the sequential SRO emissions extended widely on the time scale. From ethanol droplets, the detected 1s emission was obviously delayed from the pump. The average delay time was less than 40 ps which was about the order of the pulse duration of the pump.

From benzene droplets, we have observed SRO emissions until 4th order with this time-resolved equipment and got the temporal feature of these emissions. In fig. 3, the time-resolved spectra of the SRO emissions from the benzene droplets are given. The average delay between the pump and first SRO emission is about 25 ps, the delay of the second SRO emission is 60 ps, the third SRO 95 ps, and the fourth SRO delay 120 ps. So, comparing to the SRS emission from the cell, there is a clear long delay of the SRO emissions from the droplets.

The experimental result of the temporal resolution of SRO emission from the droplets showed some main features. First, there is a longer delay of SRO emission relative to the pump than that of the SRS from the cell. Second, the higher-order SRO emissions are more delayed than the 1s SRO emission. Third, there is some scatter of the delay time for a given SRO order. Basing on the basic properties of the droplet, we can explain these results.

According to the calculation, the high feedback which comes from internal reflection at the interface of the liquid–air boundary of the ideal droplet can make the Q value of the cavity modes cover a big range, from low values less than one to a very high value larger than 10^{10} [13]. Just based on this high feedback, i.e., a small leakage out of the boundary, when the gain which can be got from the material inside the droplets by intense pumping is large enough to overcome this leakage, the oscillation would build up. For the stimulated Raman process, the incident pump field fist excites the 1s signal; the intense spot of the pump beam in the forward direction inside the droplet focused by the forward convex surface of the droplet can cause a large Raman gain for the first Stokes emission. No sooner the first Stokes oscillation build up, than the second Stokes can gradually build up by the pumping of the intense first Stokes field, and so on. So we could observe high-order Stokes emissions from the droplets.

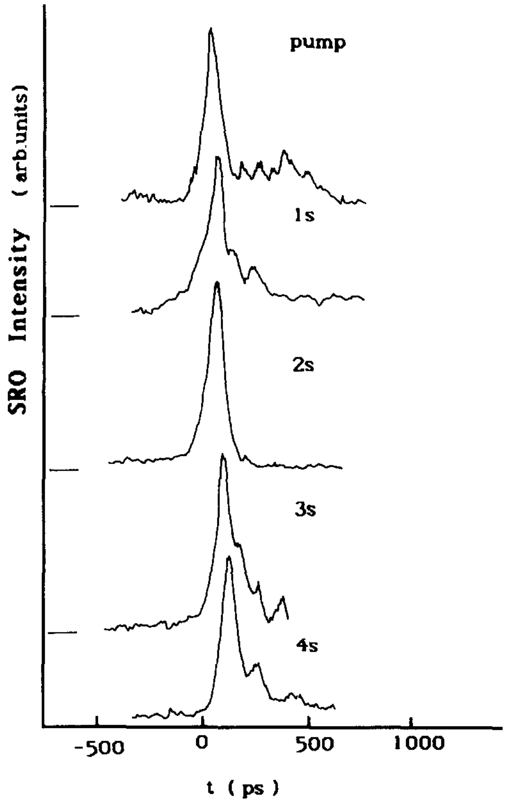

Fig. 3. The time-resolved spectra of the Stokes SRO from C_6H_6 droplets. The diameter of the droplets was about 100 μm. The delay time of four orders Stokes SRO were calibrated relative to the incident pump by the use of the reference fiber. It can be seen that there is a long delay among the SRO emissions from the droplet. Notice that because the higher-order SRO emission is weaker than that of lower-order SRO, the intensity scales in the figure for different order SRO are not the same.

In the cell, the travelling incident field pumps the first Stokes field and then other Stokes fields, because after the pump leaves the cell the Stokes emission cannot get any gain and stops immediately. Though there may be a delay among the pump and SRS emissions, the delay should be very short.

The situation for the droplets is quite different from the cell. Of course, the establishment of the Stokes oscillation inside the droplet needs time for getting enough growth, so in the time scale the emission of the Stokes SRO usually occurs after the peak of the pump. The SRO emission from the droplet is a result of the oscillation inside this spherical cavity. According to the calculation for a picosecond pump [5], if the pump power density is 35 GW/cm^2, the building-up of the first SRO inside the droplet with diameter of 60 μm needs about 30 ps. In this calculation we assumed that the pump pulse had a rectangular shape and did not take account the real pulse profile of the pump, but the requirement of the high pump power density and the threshold for SRO is basically right. So this result means that for ps pump beam with a power density of 30 GW/cm^2, only at the second half of the ps pump pulse the oscillation inside the droplets can be established. This explains why there was obviously a delay of 1s SRO emission relative to the pump. Of course, the variation of the pump density would affect the delay time.

Because the higher-order SRO was pumped by the lower-order SRO oscillation, it should be much more delayed relative to the pump than the first SRO in the droplet. The ns experiment has proved this [10–12]. Compared to the picosecond pump field for the 1s SRO, the 1s SRO field within the droplet served as the pump for 2s oscillation. At the first half duration of the pump, the 1s field was still very weak so there was very small gain for the 2s field and really no growth of the 2s field could be expected. Only when the 1s field became very intense and turned to oscillate, there would be the possibility for the building of the 2s field inside the droplet. For the incident pump, the field at the focusing spot was very intense but this intense spot only occupied a small part of the circumference of the droplet. On the contrary, though the intensity of the built 1s field inside the droplet was weaker than that of the pump field, the peculiar distribution of the Stokes field along the surface of the droplet could compensate for the weakness of the field, and after integral of the gain in the roundtrip the total gain of the 2s field would be comparable with the gain of the first Stokes [14]. In addition to this, because the 1s SRO was a standing wave oscillation inside the droplet with low loss, it could maintain longer time than the pump field. So, the second Stokes could grow within the duration time of the 1s oscillation in the cavity which was longer than and delayed from that of the pump. The actual delay of the 2s emission relative to the 1s emission was determined by the 2s SRO building process inside the droplet. Due to the same reason, the higher-order SRO had a longer delay than that of lower-order SRO. That was in agreement with the experimental results.

Because the size of the droplets in the experiment could not be controlled exactly, we really did not know whether the emissions from the droplets were corresponding to the same cavity mode of the spherical cavity. As there were a great number of modes and there was a very big difference in the Q values for different cavity modes, so for modes with higher Q value the building of the oscillation would be easier than for modes with smaller Q value. From previous experiments, we knew that a small variation of the radius of a droplet would cause a change of the size parameter $x=2\pi a/\lambda_0$ at a given wavelength λ_0. Then it would shift the position of the modes on the wavelength scale. This means that new modes would shift to the position with large Raman gain on the Raman profile and replace the original oscillating mode. In this way, the gain of the oscillating mode would change with the size of the droplet. Surely, there may be a large change of the Q value of the oscillating modes, but according to the calculation of the gain for different modes and the real uniformity of the droplets of about 10^{-4} in our experiment, we concluded that the Q value of the really oscillating modes was about 10 to at most 10^6. The establishment of the oscillation of the modes with too large Q value required very uniform shape of the droplet and these modes could not oscillate inside the real liquid droplets which were generated in our experiment.

From the experiment, we found that in some cases there was a big broadening of the SRO in the time scale, i.e. a long duration of SRO emission. Such broadening has been observed for Stokes oscillations of different order and a typical spectrum for 2s SRO from a benzene droplet is given in fig. 4. Because this broadening was at the rear side of the SRO emission, this was really the long lifetime of the oscillation inside the cavity comparing to the pump. From the viewpoint of the resonant cavity, the lifetime of the oscillation depends on the Q value of that mode. The larger Q there is, the slower decay there would be. The longest decay time of SRO in the experiment was about 500 ps. This long decay time reflected the fact

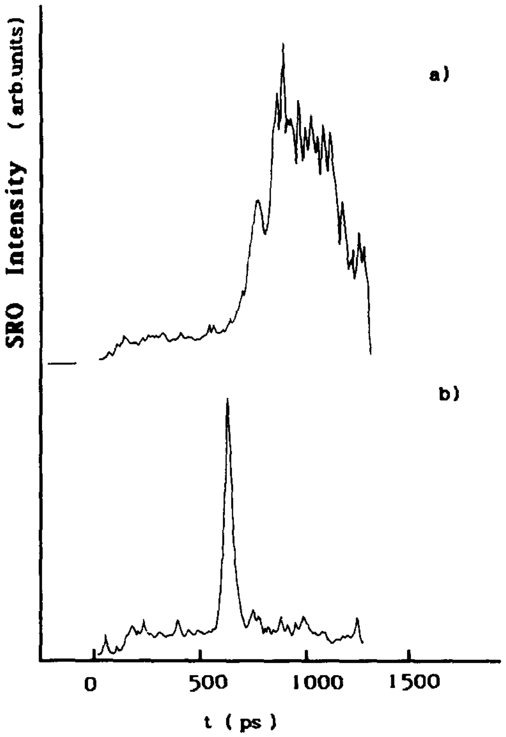

Fig. 4. (a) The broadening of 2s SRO signal from the C_6H_6 droplets. For comparison, the SHG pump signal from the fiber for the same incident ps pulse is given in (b). This slow decay of 2s SRO emission reflects the long photon lifetime of the oscillation inside the droplet for a special mode with large Q value.

that due to the high Q value of the cavity mode, the SRO oscillation could maintain many round trip in the droplet cavity even after the elapse of the pump. In turn, the high-order SRO pumped by this broadened SRO would be delayed even longer. The effects of both the fluctuation of the incident power density and the variation of the oscillation duration of the SRO could explain why the delay time of the SRO emission did not keep the same value in the experiment. The same reason is for the delay of the higher-order SRO. The average delay, however, clearly showed the time relation among the SRO emissions of different order.

4. Conclusion

The experimental result of the temporal behavior of the ps SRO emissions from the droplet has clearly shown the fact that inside the liquid droplet, the building of the different order SRO starts from the excitation of the first-order SRO and then evolves to high-order SRO. The building up process needs the time which is much longer than that in the cell. Building at the different modes, the SRO emissions showed a different decay behavior which revealed the variation of the Q value of the resonant modes of the cavity. The research of the transient spectra provides a new way to investigate the dynamic process inside the spherical cavity. Combining with other techniques, we can investigate different processes of interaction of laser radiation with liquid and solid particles.

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