# INVESTIGATION OF THE LINE WIDTH OF STIMULATED RAMAN SCATTERING

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An important parameter of both spontaneous Raman scattering (SpRS) and stimulated Raman scattering (SRS) is the radiation width. The width of the SpRS lines has been well investigated, but data on the SRS line widths are very limited [1 - 4].

We have measured the line widths of the first Stokes component of the SRS of a number of substances. The substances were investigated in both the liquid and the solid state. In the latter case, both single crystals and powders were used.

The SRS spectra were excited by a ruby laser with a passive Q-switch (solution of vanadium phthalocyanine in toluene). The radiation power in the pulse was $\sim$ 10 - 15 MW and the pulse duration $\sim$ 20 - 30 nsec. No selection of the angle modes was made. The laser radiation was focused on the sample with a lens of $f$ = 12 cm. The exciting radiation and the SRS radiation passing through the sample were gathered with a lens of $f$ = 25 cm and directed to a Fabry-Perot interferometer. The interference pattern was photographed with a UF-89 camera ($f$ = 0.8 m). If the sample was a single crystal or a liquid, a ground-glass plate was placed past the sample. If the sample was a powder, no such plate was used. The investigated radiation was separated with interference filters mounted either in front of the lens of the UF-89 camera, or at a distance 7 - 10 cm in front of the photographic plate. The spectra were registered on infrachromatic plates of high sensitivity, prepared at the State Chemical and Photographic Design Institute and furnished kindly by A. S. Kheinman. We developed a method of superimposing density markers by using the SRS of the investigated samples. This method makes it possible to take into account the dependence of the properties of the photographic material on the wavelength of the incident radiation and on the duration of the exposure.

In the investigation of liquid or powdered substances, we used glass cells whose windows were not specially checked for plane-parallelism. The cell length was 5 cm for liquids and 4 mm for powders. The powder grain dimension was 0.2 - 0.3 mm. Polycrystalline benzene was obtained by rapidly freezing liquid benzene in a quartz cell 2 mm thick. The single-crystal samples were either plates or parallelepipeds. The stilbene crystal was rather non-uniform, its rear face being skewed by 3 - 5°; in the case of the calcite crystal, the laser beam was incident not perpendicularly but at an angle 3 - 5°.

The measurement results are shown in Table 1. Examples of the spectra are shown in the figure. The table lists for comparison the SpSR line widths of the same substances. The SpSR spectra were investigated with a DFS-12 spectrophotometer with excitation by the 4880 and 5145 Å lines of an argon laser.

Interference patterns of stilbene-powder SRS radiation at $\nu = 997$ cm$^{-1}$. The SRS lines are on the right and the exciting laser radiation on the left ($\lambda = 6943$ A). The distance between rings is 1.7 cm$^{-1}$.

The measurement of the SRS line widths in acetone and benzene was hindered by the influence of the stimulated Mandel'shtam-Brillouin scattering (SMBS). The interference pattern of the Stokes SRS radiation of benzene and acetone shows, against a background of 0.6 cm$^{-1}$ width, a structure whose components were apparently excited by the individual SMBS components. This makes it possible to propose that the true width of the SRS line in benzene is smaller than the distance between the SMBS components, i.e., smaller than about 0.2 cm$^{-1}$. The data for benzene and acetone must be regarded only as roughly approximate. We note that a fine structure of the SRS line of benzene, with individual component widths 0.05 cm$^{-1}$, was likewise observed in [4].

Table 1

| Substance | Aggregate State | $T$, °K | $V$, cm$^{-1}$ | $\delta\nu_{SpRS}$, cm$^{-1}$ | $\delta\nu_{SRS}$, cm$^{-1}$ |
|---|---|---|---|---|---|
| Stilbene | crystal $\ell = 1,5$ cm | 293 | 1593 | 4,8 | 0,6 |
|  |  |  | 997 | 2.5 - 3 | 0.14 |
|  | powder | 250 | 1593 | 4,6 | 0.37 |
| Calcite | crystal $\ell = 3$ cm | 293 | 1086 | 1.1 | 0.22 |
| Potassium nitrate | powder | 250 | 1051 | — | 0.05 |
| Benzene | polycrystal | 100 | 992 | — | 0.1 |
| Benzene | liquid | 293 | 992 | 2 | < 0.2 |
| Acetone | liquid | 293 | 2930 | 17 | < 0.2 |

It is seen from Table 1 that the SRS line width is smaller by a factor 10 - 20 than the SpRS line width. According to the theory [5, 6], the line width is

$$\delta\nu_{SRS} = \delta\nu_{SpRS}\sqrt{\frac{\ln 2}{K\ell}} \,,$$

where K is the gain at the SRS frequency and $\ell$ the sample length. For most substances $k\ell \sim$ 20 - 40. Thus, the SRS should become narrower by a factor 5 - 7. A much larger line narrowing was observed in the experiment.

We have investigated the temperature dependence of the SRS line widths of stilbene

($\nu$ = 1593 cm$^{-1}$) and potassium nitrate ($\nu$ = 1051 cm$^{-1}$) (see Table 2). The investigations were made with powders. The SRS line became narrower with decreasing temperature, and the temperature

Table 2

| Substance | $T, °K$ | $\delta\nu_{SpRS}$, cm$^{-1}$ | $\delta\nu_{SRS}$, cm$^{-1}$ |
|---|---|---|---|
| Stilbene | 250 | 4.6 | 0.37 |
| | 100 | 3.7 | 0.25 |
| Potassium nitrate | 250 | — | 0.05 |
| | 100 | — | 0.03 |

dependence of $\delta\nu_{SRS}$ follows that of $\delta\nu_{SpRS}$ within the limits of experimental accuracy ($\sim$ 20%).

The results obtained by us for calcite crystals coincide with the estimates of [6]. The already mentioned appreciable decrease of the SRS line width for other substances is apparently not connected with self-focusing, for it is observed in certain substances in which there is no self-focusing (acetone). The most natural explanation is that the appreciable line narrowing is due to multiple plasage of the SRS radiation through the cell, owing to relfection from the end faces, scattering by inhomogeneities, etc., but in our experiments we took precautions to eliminate these effects. Another possible explanation is given in [5].

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