Temporal structures of stimulated Raman scattering in a capillary-filling liquid with transverse laser pumping

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Quasiperiodic modulation structures, soliton-like trains, and single pulses less than 20 ps wide were observed in the time evolution of the spectrum of stimulated Raman scattering in carbon disulfide filling a capillary with transverse laser pumping. Up to seven Stokes and three anti-Stokes SRS components were observed. © *1995 American Institute of Physics.*

1. Stimulated light scattering has been studied extensively and, in particular, several papers on stimulated Raman scattering (SRS) in liquids in a capillary excited by radiation propagating along the axis of the capillary have been published (see, for example, Refs. 1–3 and the references cited there). In the present letter we report the results of an experimental study of the spectral and temporal characteristics of SRS in liquid carbon disulfide in a glass capillary with transverse laser pumping. Initially, the stimulated Brillouin scattering (SBS) component is excited at a scattering angle of 90° (SBS-90) and propagates along the axis of the capillary. The SBS-90 component give rise to a SBS scattering at an angle of 180° (SBS-180), followed by further SBS-180 and SRS components. In Ref. 4 large-scale oscillations with a period of $\sim 2-6$ ns and small-scale modulation with periods equal to the period of the hypersonic wave accompanying SBS-90 and SBS-180 (~ 0.2 ns and ~ 0.13 ns) and its subharmonics were observed in the time evolution of the SBS-90 and SBS-180 intensity in the absence of SRS. In our study we observed a complex temporal structure in the SRS components obtained at higher pump power. In this struture quasiperiodic soliton-like trains of pulses and single pulses were identified and new modulation structures in SBS were observed. The physical reason for this complex temporal structure is the nonlinear interaction of radiation with different modes of motion in the medium and the competition between the forms of scattered light that arise.

2. Stimulated scattering was excited by the second harmonic of a neodymium laser (530 nm) with pulse width (at half-height) $t_p \simeq 15$ ns and maximum energy ~ 200 mJ. The laser radiation was expanded to 6 cm and focused with a cylindrical lens into a capillary containing carbon disulfide. The capillary had an inner diameter of 170 μm and length $L = 18$ cm (the light transit time was $T_0 = Ln/c \approx 1$ ns and the light transit time between the canted exit windows was $T_1 \approx 1.15$ ns). After spectral decomposition with a Pellin–Broca prism the stimulated scattering component from the end of the capillary was focused with a cylindrical lens onto the slit of an Agat image-converter camera, and the spectrum was swept in time. A sweep of 10 ns/cm and sometimes 2 ns/cm was employed. We estimated that this gave a resolution of not worse than ~ 20 ps. Up to

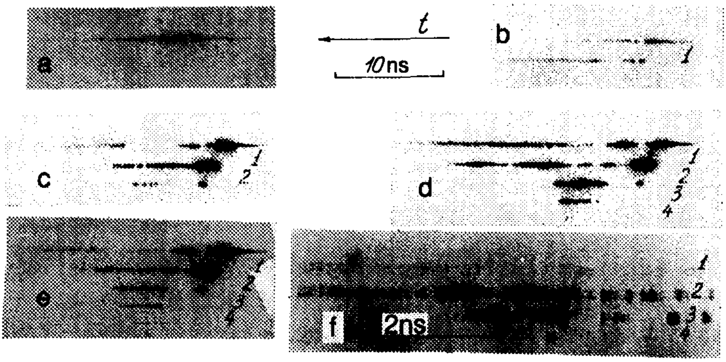

FIG. 1. Time evolution of SBS (a) and SBS and SRS (b–f) spectra. a–e — 10 ns/cm sweep, f — 2 ns/cm sweep. 1–4 — Successive SRS-1–SRS-4 components, the wavelength of the light increases from top to bottom, and the spectral shift of the SRS is equal to 656 cm^{-1}.

seven Stokes and three anti-Stokes SRS components ($\vartheta = 656\ \mathrm{cm}^{-1}$) were recorded in the static regime at maximum pump intensity.

3. The following dynamics of the development of the stimulated scattering process was observed. At low pump intensities we observed SBS-90 and SBS-180 components which were not spectrally resolved in the present experiment (Fig. 1a) (but which were resolved in Ref. 4 with an interferometer). Just as in Ref. 4 , their intensity is weakly modulated with intervals of 2–6 ns and sometimes 9–10 and 11-12 ns between the maxima. As in Ref. 4, small-scale modulation, corresponding to the periods of the hypersonic waves (0.1–0.15 ns and ~0.2 ns) and their subharmonics excited with the SRMS, as described in Sec. 1 of Ref. 4, is observed against the background of these large-scale oscillations.

As the pump power increases, the appearance of the first Stokes component (SRS-1), 6–8 ns after SBS begins, is accompanied by almost complete exhaustion of SBS in the time interval of the SRS radiation (Fig. 1b), and two or more such intervals of alternating SBS and SRS can occur. In the process, single pulses and groups of pulses with increasing time intervals 0.2–0.9 ns are often observed in the SBS and SRS radiation. Stimulated Raman scattering can start with a single isolated pulse or a group of short pulses. Longer intervals of SRS radiation appear much later, and the SRS intervals are accompanied by exhaustion of the SBS radiation to some degree during the first half of the pump pulse. At the end of the pump pulse they can be emitted simultaneously in the form of weakly modulated cw radiation. The same large-scale oscillations and small-scale modulation as in SBS radiation (in the absence of SRS) appear in both SBS and SRS.

At even higher pump power and with the appearance of a second Stokes component (SRS-2), the temporal structures of the stimulated scattering change and become ordered (Fig. 1c). The qualitative features of these structures also appear with a large number of SRS components. Small-scale modulation can often be separated during the 5 to 8-ns rising part of the SBS pulse. At the end of this part of SBS the radiation reaches a maximum intensity, SRS radiation starts, and the SBS radiation decreases sharply, often dropping below the level of detectability. Then, 0.5–0.8 ns later, a short (<0.2 ns) intense SBS pulse appears and 1–2 ns later a longer, also intense pulse appears (sometimes several pulses appear), after which SBS is not detected or weak radiation is observed during 4–6 ns, sometimes with maxima with no discernible regularity of the intervals between them, and the SBS is completed with a long (7–15 ns) pulse with decreasing intensity, against the noise background of which small-scale quasiperiodic structures often appear. SRS-1 starts with two to six intense pulses of duration ~0.4–0.5 ns separated by ~0.4–0.5 ns, so that they are often difficult to resolve in the photographs. Next, there follow with some interruptions groups of pulses whose separation in time often increases from ~0.3–0.5 ns to 0.8–1 ns (Figs. 1c and 1d). SRS-2 often begins (Figs. 1c–1e) with a single intense pulse (but not always), which is followed 2–3 ns later by groups of pulses separated in time by 0.4–1 ns. The intervals between these pulses often increase systematically from 0.4–0.5 ns to 0.7–1 ns (Fig. 1c), and sometimes the pulses are almost equally spaced with intervals of 0.4–0.6 ns. Such soliton-like groups of pulses are often observed especially clearly with the excitation of several SRS components in the higher-order components. Sometimes the last SRS component is observed in the form of one (Fig. 1d) or several (Fig. 1e) short (less than 20 ps) pulses.

An anti-Stokes component with temporal structure similar to that of Stokes SRS was observed with pump intensities at which three Stokes components appeared.

The temporal dynamics of the SRS with a sweep of 10 ns/cm and resolution ~50–100 ps was described above. Photographs of the SRS spectra with a sweep of 2 ns/cm and resolution ~20 ps (Fig. 1f) were also obtained. Only part of the Stokes stimulated-scattering pulse (it is not clear which part), practically not visible in SRS-1 and SBS, was registered but a fine temporal structure was observed in SRS-2–SRS-4, which makes it possible to determine more accurately the structures described above.

Large groups of short (duration less 20 ps) pulses with large-scale (0.4–1 ns) intensity modulation with alternating intervals of ~85 ps and ~100 ps and smaller groups or pairs of pulses with intervals of 40–70 ps are observed in SRS-2 (Fig. 1f). Identical groups are observed in SRS-3, but there are fewer pulses and intervals of 0.45–0.7 ns are clearly observed. If pulses with the intervals indicated above are grouped according to the intensity maxima of the large-scale modulation into groups taking into account the intense single pulses, then a sequence of groups is obtained for Fig. 1f with intervals increasing from ~0.4 up to 1–1.2 ns for SRS-2 and from 0.45 up to 0.85 ns for SRS-3. In a different realization such groups are approximately equally spaced with an interval of ~0.5 ns; a similar situation is observed in other realizations. In SRS-4 a pair of pulses with an interval of 60–70 ps is observed.

4. The facts that the SBS structure is transferred to SRS and therefore the periods of the hypersonic waves of 185 ps (SBS-90) and 130 ps (SBS-180) and their subharmonics can appear must be taken into account in the analysis. Waves with identical structure but shifted in phase, because of rereflection (even weak) from the windows and induced gratings and subsequent amplification, can appear. For example, the above-described alternating 85-ps and 100-ps structure (Fig. 1f) could be the sum of two modulated structures which are shifted in phase by an amount close but not equal to $\sim\pi/2$ as a result of SBS-90. Taking into account SBS-180 and SBS-90 and their subharmonics, it is thus possible to explain the time intervals of 40–70 ps and others.

Large-scale intensity oscillations with peaks separated by more than 0.4 ns are superimposed on the small-scale modulation. We assume that the oscillations with period $\sim 2-6$ ns which were observed in Ref. 4 in SBS and in the present work in SRS are due to feedback as a result of reflection from the exit windows and subsequent amplification. Since the double-passage time for light through the capillary $2T_0 \approx 2-2.3$ ns is comparable to the phonon lifetimes during SBS-90 ($T_r \approx 3.4$ ns) and SBS-180 ($T_r \approx 1.7$ ns), and since a steady state is established over the time $\tau \sim G^{1/2} T_r$ (G is the gain), the period of the oscillations must be longer than $2T_0$ (Ref. 5) and it must depend on the intensity, in qualitative agreement with experiment.

Another reason why large-scale oscillations appear are relaxation oscillations of the intensity,[5] which are produced as a result of the interaction of oppositely propagating pump beams (in this case SBS-90) and scattered light beams (SBS-180 excited by SBS-90). If the period of these oscillations is determined by the saturation time of the SBS-180 intensity, then for short T_r the maximum period of the oscillations would be $\sim T_0 = 1$ ns (half the length L) and since T_r is finite, it can be appreciably longer. At the same time, the saturation of the SBS-180 intensity can also occur over a distance much shorter than $l = T_0 c$, for example, at the boundary of the illuminated part of the capillary or even sooner, and then the period of the oscillations will be $\sim 0.6-0.7$ ns or shorter, which is close to the observed spacing of the soliton-like pulses $\sim 0.45-1$ ns. Oscillations with a period of $2T_0$ or longer and finite T_r can appear with the appearance of subsequent SRS-180 components. Here the lifetime T_2 of the optical phonons, the competition between different stimulated-scattering components, and the periodic depletion of the intensity of the lowest-order components at the expense of the higher-order components must also be taken into account.

In the process of stimulated scattering the temporal structures of the components with the lowest frequencies are transferred to higher frequencies in such a way that the most intense components in the maxima of the large-scale oscillations are accentuated by nonlinear amplification. This can result in the formation of ordered groups of pulses in the higher-order SRS components, as in Fig. 1c for SRS-2 and in Fig. 1e for SRS-4, or even single pulses, as in Fig. 1d for SRS-4. The higher temporal resolution shows that these pulses probably can have a temporal fine structure with duration less than the resolution limit ~ 20 ps.

To analyze theoretically the stable soliton-like temporal structures described above, we must analyze several coupled nonlinear equations (for SBS-90, SBS-180, and SRS), taking into account the self-modulation and cross-modulation. The interaction of the pump radiation and SRS-1 in the region of normal dispersion does not give stable structures (see Ref. 6 and references cited there). In Ref. 7 it was shown for a specific case by means of modeling that when several successive SRS components appear, it is possible, with allowance for the finiteness of T_2, for groups of solitons to form even in the region of normal dispersion. This is important for the interpretation of temporal structures in SRS of any form. Although the temporal structures described there are similar to a certain extent (but not completely) to the structures which we observed, they cannot be comapred because the conditions are different.

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