STIMULATED OPTICAL FREQUENCY MIXING IN LIQUIDS AND SOLIDS*

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The well-known phenomenon of the stimulated Raman effect is a remarkable example of nonlinear optical behavior. In this phenomenon a large percentage of very high-intensity light may be transformed into molecular sidebands (Stokes and anti-Stokes components and their true harmonics) in the process of being transmitted through the materials.

A second nonlinear phenomenon known as the stimulated Brillouin effect was first observed by Chiao, Townes, and Stoicheff[1] in solids, and by Garmire and Townes[2] and Brewer and Rieckhoff[3] in liquids. There is already a rather extensive literature on this subject. This phenomenon has been observed in gases[4] as well as in liquids and solids. Certain remarks concerning the observations made on the stimulated Brillouin effect are in order to clarify the experiments which we will describe. We have been able to observe as much as 80 % of the incident polarized light to be back-scattered from liquids and 45 % from high-pressure gases. The back-scattered Brillouin components are polarized. To the best of our knowledge no anti-Stokes components in liquids have been observed with certainty.

We have passed a single-moded ruby laser beam through liquids and solids. Power levels up to 70 MW with beam divergence of 8 mrad and a linewidth of about 0.03 cm^{-1} was used. A lens of 15-cm focal length was used to focus the laser beam in the materials.

The light emerging from the material in the direction of propagation was analyzed by means of a high-resolution grating spectrograph (resolving power 400 000, dispersion 3 cm^{-1} per cm on the photographic plate). A reproduction of the spectrum in the neighborhood of the laser frequency for two liquids is shown in Fig. 1. The spectrum labeled *A* is due to CS_2 and the one labeled *B* is due to aniline. The top section indicates the method of identifying which component is the unmodified frequency. It is an exposure made in two parts, the top half with no sample in the beam, the lower half with a CS_2 cell. With close control of the ruby temperature its frequency was constant within 0.03 cm^{-1}. In the figure the unmodified frequencies are placed in a vertical line.

The frequency separations of the components shown in Fig. 1 are identical to those observable in 180° back-scattering making use of the stimulated Brillouin effect. Our measurements give 0.197_6 cm^{-1} for the separation of the components of the spectrum of CS_2 shown in Fig. 1. The measurements were made at 26°C. In addition to the two materials which are the subject of Fig. 1, two optical glasses and a large number of liquids have been investigated in the same manner as described above and similar results have been obtained in all cases.

From the measured separation of the spectral components the velocity of sound can be calculated. Good agreement between our measurements and those of Chiao and Fleury[5] was obtained. Because of the dispersion of sound velocity at high frequency, it is necessary to compare our measurements with others made using 180° scattering.

All of the liquids and solids which we have observed show one or more "anti-Stokes" (violet-shifted) components. In the case of CS_2 we have observed 30 Stokes and 15 anti-Stokes components. Anti-Stokes as well as Stokes shifted components can occur even when the back-scattered light cannot be amplified and returned to the sample.[6] This amplification was occurring when the spectra shown in Fig. 1 were obtained.

The media investigated to all intents and purposes are completely transparent to the laser wavelength. However, the amazing situation

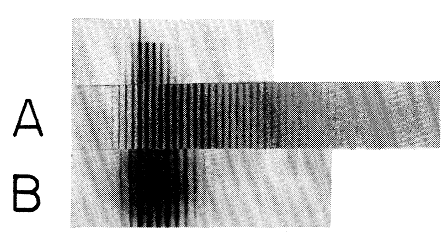

FIG. 1. Stimulated Stokes and anti-Stokes components observed in the neighborhood of the laser frequency with a high-resolution spectrograph. (A) CS_2 and (B) aniline. The top section indicates how the laser frequency was identified; the upper half is the laser alone, the lower half the laser through CS_2. The separation of the components is 0.198 cm^{-1} in CS_2 and 0.259 cm^{-1} in aniline.

occurs that absolutely none of the laser light is transmitted without being modified. The laser frequency indeed appears in the spectrum but it, as well as the modified components, is completely depolarized while the incident light and back-scattered light are completely polarized.

Although no quantitative statements can be made at this time with regard to threshold power, it is clear that there must be a threshold for the occurrence of this phenomenon since no ordinary light sources of the conventional type are capable of producing this type of behavior. The effects were observed in CS_2 using power levels between 3 and 70 MW with a lens of 15-cm focal length to focus the light.

It seems to be moderately certain that the phenomenon which we have observed is the result of some type of mixing of optical frequencies in the media which involves the acoustic frequency as well as the optical frequencies. The laser pulse excites the Brillouin effect and the created red-shifted frequency mixes with the laser frequency thus creating new frequencies which, in turn, are capable of further mixing producing the proliferation of new frequencies. We are in the process of doing further work along these lines and a full report of our experiment will appear elsewhere.

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GOVERNING INFLUENCE OF ATOMIC DEGENERACY ON MODE INTERACTIONS IN A GAS LASER

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A theoretical treatment of the behavior of a generalized gas laser has recently been completed.[1] The results of this treatment indicate that the behavior is strongly dependent on the degenerate nature of the atomic energy levels participating in a given laser transition. In the present note we report the results of an experimental study of laser mode coupling which has served to verify some of the predictions of the theory and to establish its realm of validity. This study was principally concerned with the interaction of two modes whose frequency separation is small compared to the decay constants of the participating atomic levels. In this limit the theoretical expressions simplify sufficiently to allow easy comparison with experiment. Furthermore, the theory is most appropriately tested in this limit since it is here that the key assumption used in its development is open to question.

For the special case of zero frequency splitting and equal cavity losses, the theoretical condition for the simultaneous oscillation of two modes becomes

$$\theta_{ij}/\beta_i = \theta_{ji}/\beta_j \leq 1, \quad i,j = 1,2 \text{ or } 2,1, \tag{1}$$

where the interaction parameters θ_{ij} and β_i can be written in the simplified forms

$$\theta_{ij} = Q(\nu)\sum_m \{\mathcal{P}_{m,m}{}^2[\mathcal{P}_{m+1,m}{}^2 + \mathcal{P}_{m-1,m}{}^2 + \mathcal{P}_{m,m+1}{}^2 + \mathcal{P}_{m,m-1}{}^2][1 + (\langle \vec{e}_i \cdot \vec{e}_j \rangle_{av})^2]$$
$$+ [\mathcal{P}_{m,m}\mathcal{P}_{m,m-1}\mathcal{P}_{m-1,m-1}\mathcal{P}_{m-1,m} - \mathcal{P}_{m,m}\mathcal{P}_{m,m+1}\mathcal{P}_{m+1,m+1}\mathcal{P}_{m+1,m}](\langle \vec{e}_i \cdot \vec{e}_j{}^r \rangle_{av})^2\} \tag{2}$$

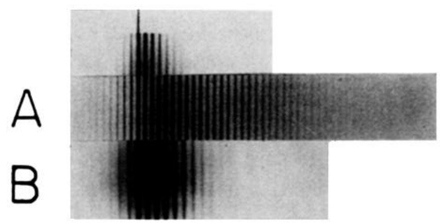

FIG. 1. Stimulated Stokes and anti-Stokes components observed in the neighborhood of the laser frequency with a high-resolution spectrograph. (A) CS_2 and (B) aniline. The top section indicates how the laser frequency was identified; the upper half is the laser alone, the lower half the laser through CS_2. The separation of the components is 0.198 cm^{-1} in CS_2 and 0.259 cm^{-1} in aniline.