obtained from measurements of the linewidth or intensity as a function of θ and T very near T_c. Although carbon dioxide was in a sense a fortunate choice for this experiment since it has been studied more thoroughly than any other gas near its critical point, it will be interesting to have these results for a monatomic gas, for then there will be no possibility that the results will be influenced by the internal degrees of freedom.

Investigation of Rayleigh Wings and Brillouin-Stimulated Scattering in Liquids*

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Measurements of the spatial gain associated with stimulated Brillouin and Rayleigh-wing scattering are presented. These results are obtained by amplifying a signal in a short cell. This technique enables us to obtain reliable data even in liquids where self-focusing has a very low threshold. Comparison of measured and calculated gains in different liquids demonstrates the steady-state character of the gain at the Brillouin frequency for larger-linewidth liquids and the transient effect in liquids with sharper linewidths. We give values for the gain at the Stokes frequencies on the Rayleigh wings of nitrobenzene and toluene, and compare them with the calculated values using simple models for the scattering cross section.

I. INTRODUCTION

THE scattering of light by density and entropy fluctuations,[1-3] random motion of anisotropic molecules,[4-8] and other molecular degrees of freedom[9,10] in matter has been known for a long time. These scattering effects can give a great deal of information about the matter itself and have been the subject of investigation over a period of many years. To each of these scatterings the general quantum formulation of the interaction between light and matter associates a stimulated emission. When scattering occurs with frequency change, the stimulated effect gives rise to amplification or absorption of light at the scattering frequencies with a variety of spatial and time behaviors depending upon the ratio of different time constants. When several intense waves are present, interactions are more complex.[11-14] All these effects have only recently been carefully analyzed theoretically, because of the interest generated by the advent of powerful Q-switched lasers.[15-19]

Because of the relatively large linewidth of Raman lines compared with laser linewidths, the stimulated

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[15] R. W. Hellwarth, Phys. Rev. **130**, 1850 (1963).
[16] R. Y. Chiao, E. Garmire, and C. H. Townes, in *Proceedings of the International School of Physics "Enrico Fermi"* (Academic Press Inc., New York, 1963).
[17] D. I. Mash, V. V. Morozov, V. S. Starunov, and I. L. Fabelinskii, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu **2**, 41 (1965) [English transl.: Soviet Phys.—JETP Letters **2**, 25 (1965)].
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Raman effect has been the first to be shown to induce a purely spatial gain or absorption of exponential character at the scattered frequencies in liquids.[20] The correct dependence of this effect on several different pertinent parameters has been investigated experimentally although some results are not yet clearly interpreted.[21,22] Stimulated Brillouin and Rayleigh-wing scattering, on the contrary, have drawn less experimental interest.

For Brillouin scattering, only the frequency relation has been well established.[23-27] Demonstration of the relative spatial and temporal dependence of the growth of the Brillouin wave has been the subject of some controversy[28-30] although some convincing results on the transient effect have been obtained in gases at different pressures.[31]

The purely spatial character of gain or absorption on the wings of the Rayleigh line seems evident from the comparison of linewidths, but this effect has been demonstrated only qualitatively by large amplification in high-intensity filaments and by the creation of many sidebands of multifrequency exciting light.[17,32-34]

The relaxation time of entropy fluctuations is relatively long compared with laser pulse duration, and often very long compared with the coherent time of the laser light. The associated stimulated emission is therefore difficult to study experimentally, and only recently has an associated process in light-absorbing media been proposed and observed.[35]

In the present work, we first relate the expected gains due to stimulated emission at Stokes frequencies to spontaneous emission parameters and discuss the different experimental possibilities. In Sec. III, we describe our experimental setup and give the characteristics of

our Q-switched laser which emits a very monochromatic light. Section IV and V are devoted to our experimental results in liquids. We show that stimulated Brillouin scattering exhibits a purely spatial gain in most of the liquids studied. A considerable simplification of the analysis is then possible and one can relate precisely the observed amplification to spontaneous emission parameters. We demonstrate the existence of a spatial gain on the Stokes wings of the Rayleigh line in nitrobenzene and toluene and we compare this gain with known data obtained in spontaneous emission. In the last section, we point out some experimental difficulties arising from competition between mechanisms with different time constants.

II. THEORY

To take into account the stochastic nature of the different broadening mechanisms, a complete analysis of stimulated emission in a two-photon process would imply a description in terms of the density matrix.[18,36] However, this method is not necessary to obtain the simple results which we want to demonstrate experimentally and which demand only a knowledge of the electromagnetic fields. Our present work is limited to the interaction of electromagnetic waves at two frequencies with liquids which are isotropic and where the intermolecular distance is much shorter than a wavelength. Furthermore, only the laser field is supposed to be very intense, the other field being associated with a weak test light used to measure amplification at different frequencies. In this case, the formulation can be relatively simple and can include Rayleigh, Brillouin, and Raman scattering in the same formalism.[37]

The correspondence between gain or absorption associated with stimulated emission and spontaneous emission at the same frequencies may be obtained by quantizing the electromagnetic field and using second quantization to describe the elementary excitation of the material system.[38-42] The spontaneous emission in turn, can be characterized either by a scattering cross section and a linewidth which can be measured experimentally, or by an appropriate model for the scattering of the light by the medium.

In a cavity of volume V the electromagnetic fields, at frequencies ν_L for the laser and ν_s for the probe, and the field associated with the excitation of interest are represented in term of normal modes with occupation numbers N_L, N_s, and N_p, respectively. Creation or ab-

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[21] See, for example, *Physics of Quantum Electronics* (McGraw-Hill Book Co., New York, 1965), Part 2.

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[33] C. W. Cho, N. D. Foltz, D. H. Rank, and T. A. Wiggins, Phys. Rev. Letters **18**, 107 (1967).

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[41] J. A. Fleck, University of California Internal Report, 1964 (unpublished).

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sorption rates for these bosons are proportional to $(1+N)$ and N. Interaction of the electromagnetic field with matter is then described by a net rate of creation of photons at scattered frequency:

$$dN_s/dt = K[N_L(1+N_s)(1+N_p) - N_s N_p(1+N_L)], \quad (1)$$

where K is characteristic of the medium.

Energy and momentum conservation imply

$$\nu_L - \nu_s = \nu_p,$$
$$k_L - k_s = k_p.$$

In the case where $N_s \ll 1$, the spontaneous emission is simply given by

$$dN_s/dt = KN_L(1+N_p). \quad (2)$$

K may either be related to measured spontaneous intensities on both polarizations, and relation (1) will then enable us to compute the effective gain associated with stimulated emission at the same frequency or be deduced from a model for scattering of light by the medium. In this case, gain measurements that are discussed in the experimental part of this paper will be a test of the model and can easily give a very high spectral resolution.

If the exciting light is monochromatic of spectral distribution $\delta(\nu_L)$ the scattered light has a normalized spectral distribution $f(\nu_s) \otimes \delta(\nu_s) = f(\nu_s)$, which in the case of a Lorentzian shape is simply given by

$$f(\nu_s) = \frac{2}{\pi \Delta\nu_s[1 + (2(\nu_s - \nu_{0s})/\Delta\nu_s)^2]},$$

where $\Delta\nu_s$ is the width at half-intensity.

When the spectral distribution of the source is $g(\nu_L)$ having width $\Delta\nu_L$, the scattered light has the distribution $g(\nu) \otimes f(\nu)$ whose width $\Delta\nu$ depends on the shapes of $g(\nu)$ and $f(\nu)$. For example, with Gaussian distributions,

$$\Delta\nu = (\Delta\nu_s^2 + \Delta\nu_L^2)^{1/2}.$$

In what follows we shall use

$$\Delta\nu = \Delta\nu_L \otimes \Delta\nu_s.$$

For one polarization the relation between K and the scattering cross section per unit volume $(d\sigma/d\Omega)_v$ for the same polarization is simply given by

$$\frac{4\pi\nu_s^2 n^3}{C^3}\left(\frac{dN_s}{dt}\right)_{\nu_s} = 4\pi N_L\left(\frac{d\sigma}{d\Omega}\right)_{v}\frac{\nu_L}{\nu_s}f(\nu)\otimes g(\nu),$$

$$K = \frac{C^3}{n^3}\left(\frac{d\sigma}{d\Omega}\right)_{v}\frac{\nu_L}{\nu_s^3}\frac{f(\nu)\otimes g(\nu)}{(1+N_p)}.$$

At thermodynamic equilibrium

$$(1+N_p)^{-1} = (1 - e^{-h\nu_p/kT}).$$

When $N_s \gg 1$, but $N_s \ll N_L$ stimulated scattering is

preponderant and Eq. (1) takes, if $N_p \ll N_s$, a simple form,

$$dN_s/dt = KN_s N_L.$$

The supposition that $N_p \ll N_s$ is valid in the case of stimulated Brillouin scattering because there are more modes in the acoustic field than in the electromagnetic field[42a] and in the case of Raman and Rayleigh wings scattering because we suppose that our probe linewidth is smaller than $\Delta\nu$, implying a longer lifetime for the photons N_s than for the excitations N_p.

For plane waves, we can then introduce a gain per unit of length by writing

$$G(\nu_s) = dN_s/N_s = (nK/c)N_L,$$

$$G(\nu_s) = \frac{C^2}{n^2}\frac{\nu_L}{\nu_s^3}[f(\nu)\otimes g(\nu)]\left(\frac{d\sigma}{d\Omega}\right)_v N_L(1 - e^{-h\nu/kT}),$$

where N_L is now the flux of laser photons per unit area.

A similar calculation using $N_L \ll N_s$ and writing dN_L/dt would have led to an anti-Stokes absorption: $A_{as} = -(\nu_{as}\nu_L^2/\nu_s^3)G(\nu)$ larger than the Stokes gain.

If $h\nu_p/kT \ll 1$, as in a Brillouin or Rayleigh transition, $1 - e^{-h\nu_p/kT}$ is well approximated by $h\nu_p/kT$ and $G(\nu_s)$ becomes

$$G(\nu_s) = \frac{C^2}{n^2}\frac{\nu_L}{\nu_s^3}[f(\nu)\otimes g(\nu)]\left(\frac{d\sigma}{d\Omega}\right)_v\frac{h\nu_p}{kT}N_L.$$

Since $(\nu_L - \nu_s)$ is now small compared with ν_L, the attenuation at the anti-Stokes frequencies is nearly equal to the gain computed at the Stokes frequencies. These different effects are represented in Fig. 1 for $G(\nu_L) = \delta(\nu_L)$.

In the case of Raman scattering, we have

$$(1 - e^{-h\nu_p/kT}) = \frac{Ni - (gi/gj)Nj}{Ni},$$

where Ni and Ni are the numbers of molecules per cubic centimeter in the ground state and first excited state with degeneracy factors gi and gj, respectively. In most of the vibrational Raman transitions at room temperature, $h\nu_p \gg kT$, and Ni is not very different from the total number of molecules per cubic centimeter, N_0. Writing $(d\sigma/d\Omega) = (1/N_0)(d\sigma/d\Omega)_v$, we have

$$G(\nu_s) = \frac{2}{\pi\Delta\nu_s}\frac{C^2}{n^2}\frac{\nu_L}{\nu_s^3}\frac{1}{1 + 2[(\nu_s - \nu_{0s})/\Delta\nu_s]^2}\left(\frac{d\sigma}{d\Omega}\right)N_0 N_L, \quad (3)$$

which is the steady-state gain quoted elsewhere.[15]

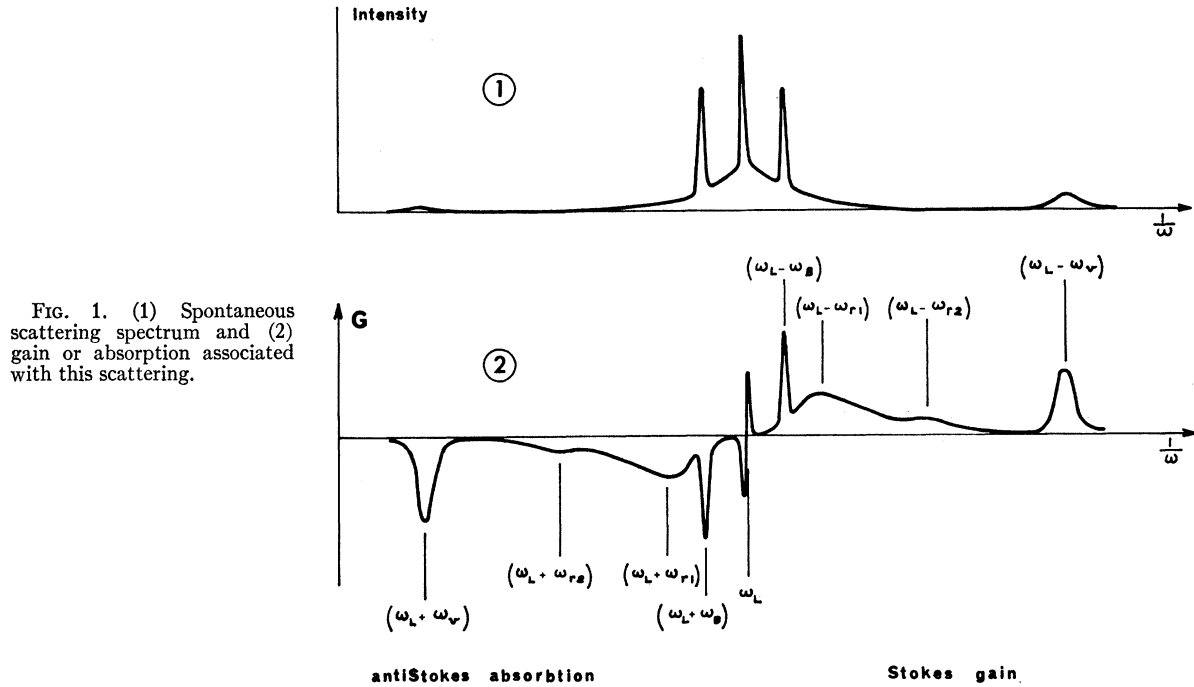Anisotropy fluctuations are usually related to two main mechanisms[43-45]: the Brownian motion of mole-

[42a] See G. Mayer, in "Enrico Fermi" Summer School, Varenna Lectures, 1967 (unpublished).
[43] I. L. Fabelinskii, Fiz. Akad. Nauk SSSR **9**, (1957).
[44] M. A. Leontovich, J. Phys. Radium **4**, 499 (1941).
[45] V. S. Starunov, Opt. i Spektroskopiya **18**, 300 (1965) [English transl.: Opt. Spectry. (USSR) **18**, 165 (1965)].

FIG. 1. (1) Spontaneous scattering spectrum and (2) gain or absorption associated with this scattering.

cules and their vibration in the field of neighboring molecules. The scattering cross section associated with Brownian motion is

$$\left(\frac{d\sigma}{d\Omega}\right)_{||} = \frac{16}{45}\frac{\omega_s^4}{C^4}(\alpha_{||}-\alpha_\perp)^2\frac{\tau}{1+\omega_p^2\tau^2}[\tfrac{1}{3}(n^2+2)]^4 N_0$$

and

$$\left(\frac{d\sigma}{d\Omega}\right)_\perp = \frac{3}{4}\left(\frac{d\sigma}{d\Omega}\right)_{||}.$$

$\Delta\nu_L$ is always very small compared with the width of $f(\nu)$ and G is given by

$$G_{||} = 64\pi^2\omega_s\frac{(n^2+2)^4}{81}\frac{(\alpha_{||}-\alpha_\perp)^2}{45kT}\frac{N_0}{n^2C^2}\frac{hN_L\omega_p\tau}{1+\omega_p^2\tau^2},$$

$$G_\perp = \frac{3}{4}G_{||}. \quad (4)$$

In the case of Brillouin scattering by longitudinal waves the backward scattering cross section is

$$\frac{d\sigma}{d\Omega} = \frac{kT\omega_s^4\gamma^2}{32\pi^2\rho_0c^4v_p^2} \quad (5)$$

for parallel polarization, and

$$d\sigma/d\Omega = 0$$

for crossed polarizations.

If the Brillouin line has a Lorentzian shape the gain

at the center of the line is, for monochromatic excitation,

$$g(\nu_L) = \delta(\nu_L),$$

$$G_{\max} = \frac{2\pi}{n^3C^3}\frac{\nu_s}{\Delta\nu_s}\frac{\gamma^2 v_p h\nu_s\nu_L}{\kappa}, \quad (6)$$

a value already obtained by Tang.[46]

If the laser has a non-negligible linewidth, then G is smaller and of the order of

$$G = \frac{\Delta\nu_s}{\Delta\nu_s\otimes\Delta\nu_L}G_{\max}, \quad (7)$$

and corresponds to the spatial part of the gain in the transient behavior discussed by Kroll.[47]

For light with polarization perpendicular to the laser light polarization the gain must be 0.

In liquids $\Delta\nu_s$ is of the order of 200 Mc/sec; the steady-state gain given by Eq. (6) will therefore be observed only when the exciting and measuring lights have a smaller linewidth.

Recently, Starunov, Tiganov, and Fabelinskii,[48] and Stoïcheff[49] have independently demonstrated the existence of a doublet near the central Rayleigh line for perpendicular polarization. This type of scattering has not been included in the present experimental work.

[46] C. L. Tang, J. Appl. Phys. 37, 2945 (1966).
[47] N. Kroll, J. Appl. Phys. 36, 34 (1965).
[48] V. S. Starunov, E. V. Tiganov, and I. L. Fabelinskii, Zh. Eksperim. i Teor. Fiz. Pis'ma Redaktsiyu 5, 317 (1967) [English transl.: Soviet Phys.—JETP Letters 5, 260 (1967)].
[49] B. P. Stoïcheff, in Symposium on Nonlinear Optics, Erevan, USSR, 1967 (unpublished).

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TABLE I. Orders of magnitude of steady-state Stokes gain, relaxation time, and frequency shift associated with the different stimulated scattering processes in liquids.

Stimulated process	Time constant (sec)	Stokes gain (cm MW^{-1})	$\nu_L - \nu_s$ (cm^{-1})
Raman	10^{-11}	10^{-3}	200–4000
Brillouin (longitudinal waves)	10^{-9}	10^{-2}	0.05–2
Rayleigh wings: Brownian	10^{-12}	10^{-3}	0–50
Oscillation	10^{-12}	10^{-4}	30–150
Rayleigh central	10^{-7}	10^{-4}	10^{-3}

Orders of magnitude for the steady-state gains per unit of length are given in Table I. It is clear that simple experimental evidence will be obtained only with excitation from a Q-switched laser. Under these circumstances, the time of excitation will not be longer than a few times 10^{-8} sec and the stimulated emission in the central Rayleigh component will present a transient effect which will drastically reduce the spatial part of the gain.

III. EXPERIMENTAL PROCEDURE

Initial experiments to demonstrate gain in Brillouin and Rayleigh-wing stimulated emission have used either large amplification of noise[28–30] in long cells or threshold measurement for oscillation in a cavity.[29] This type of experiment requires very large gains, which enhance beam inhomogeneities and limit the choice of liquids to those that are nonfocusing. In the present work, we measure a net gain on a signal produced in a separate generator. Since, in this case, the amplification may be small, the laser intensity can also be kept at a low enough level so that there is no self-trapping of the beam in the amplifying medium, even with liquids such as carbon disulfide. The frequencies of the incident laser light and of the Stokes light undergoing amplification are very close together, and no spectroscopic separation is possible; we have therefore used the backward amplifier scheme sketched in Fig. 2 in which it is possible to have a good spatial separation of the two beams.

In our experiment, photoelectric cells measuring I_{B1} and I_{B2}, the intensities of Stokes light, subtend a small solid angle and receive about 10^{-5} of the laser light. A diaphragm selects the central part of the beam which is roughly homogeneous over a 4-mm diam. A_1 and A_2 are two attenuators which are important for several reasons in this type of measurement.

(1) We focus the laser beam in cell C_2. Therefore, threshold for backward-stimulated Brillouin scattering is very low. Without attenuators the backward wave
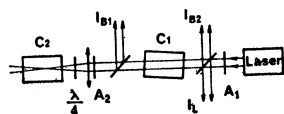
would be nearly as intense as the incident wave. No gain would appear in cell 1 because of the saturation effect, and the Brillouin light would be amplified in the laser and would create other components in C_2. The total attenuation is always kept sufficiently large, and only a small part of the outgoing beam is due to amplification of Brillouin light. The focal distance of the lens is 6 cm. The useful backward Brillouin beam generated in the cell is only 2 mm in diam because we work near threshold and use only the most intense part of the laser beam. The spread in frequency due to different angle of incidence[40] in the focal region is therefore small

$$\delta\nu = ((\nu_L - \nu_B)/7.200)$$

and does not influence our results.

(2) By varying A_1 and keeping A_2 such that the backward Brillouin intensity I_{B1} remains constant, one can measure the gain in C_1 as a function of the laser intensity I_2.

(3) By varying A_2 alone one can measure the gain in C_1 as a function of the incident Brillouin intensity. We shall see that, as expected, this gain is constant at low intensity and decreases when saturation of the laser beam occurs.

The case $\frac{1}{4}\lambda$ plate enables us to obtain a backward Brillouin light polarized at 90° to the incident light. Initial polarization of the laser has been chosen in the plane of the figure. In the case of crossed polarization signals I_{B1} and I_{B2} are larger than in the case of parallel polarizations. This is important, since otherwise a small admixture of the perpendicular polarizations would have changed completely the results in the parallel polarization case.

A careful selection of longitudinal modes has been made in our laser. The ruby rod has faces parallel to the mirrors. The high-reflectivity mirror is a thick Fabry-Perot made of two parallel glass plates. Its reflectivity is about 82%. The output mirror is a plate of glass with parallel faces.

Most of the time, this laser fires on one strong mode and a weaker one, or on only one mode. By changing slightly the mirror setting, it is possible to obtain two consecutive modes of equal intensities. Figure 3 shows two pictures of the laser spectrum obtained with a
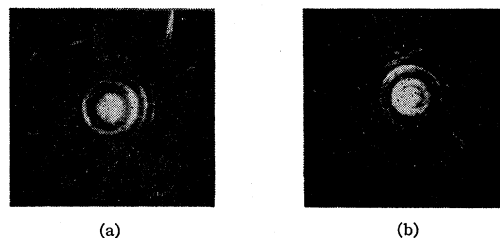

(a)   (b)

FIG. 3. Spectral structure of the laser beam observed with a 10-cm-thick Perot-Fabry interferometer: (a) monomode structure and (b) bimode structure.

FIG. 2. Experimental setup.

10-cm-long Fabry-Perot. The mode linewidth is of the order of 100 MHz to 200 MHz.

IV. GAIN MEASUREMENT IN STIMULATED BRILLOUIN SCATTERING

The spatial character of the gain in an amplification experiment will be demonstrated if the ratio of the test light intensities I_{B2}/I_{B1} is found to be independent of the incident light intensity I_{B1} and has the form

$$I_{B2}/I_{B1}=e^{G(\nu)lI_L},\qquad(8)$$

where I_L is the laser light intensity and l the length of interaction. Purely spatial gain will exist, however, only if G is equal to the value obtained for steady-state gain in Sec. II. We first demonstrate the spatial character of the gain at the Stokes Brillouin frequency in benzene and find good agreement with the computed value. Then we study other liquids and finally show that the ratio of the measured gain over the computed gain is constant for liquids with linewidths larger than 170 MHz and decreases with linewidth for liquids having smaller linewidth. These results are a good demonstration of the steady-state character of the gain in larger-linewidth liquids.

A. Spatial Gain in Benzene

Cells C_1 and C_2 are filled with benzene and in the first experiment the polarizations of laser and Brillouin light are kept parallel. For a given laser intensity the incident Brillouin intensity in C_1 is varied. Figure 4 shows the measured amplification as a function of
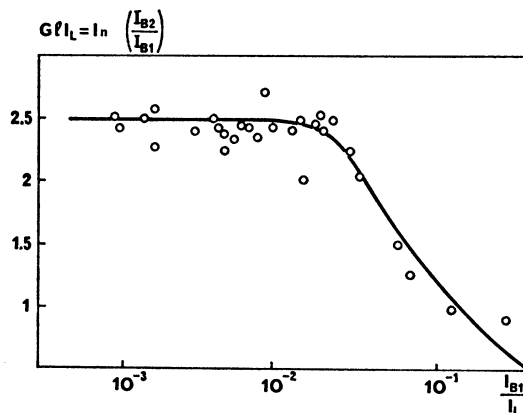
FIG. 4. Measured Brillouin Stokes gain GlI_L in benzene versus the ratio I_{B1}/I_L.

I_{B1}/I_L. At low intensity, the amplification is constant and equal to, $12.5=e^{2.5}$ When the incident Brillouin intensity reaches 4% of the laser intensity, saturation occurs because at the output of the cell C_1, Brillouin and laser intensities are of the same order.[50] For larger Brillouin intensities, there is no amplification in C_1 but we have optimum conditions for generating a strong acoustic wave.[51] Then the following experiments are all performed in the region

$$10^{-4}<I_{B1}/L_L<10^{-2},$$

when saturation does not occur and where noise due to stray light is still unimportant.

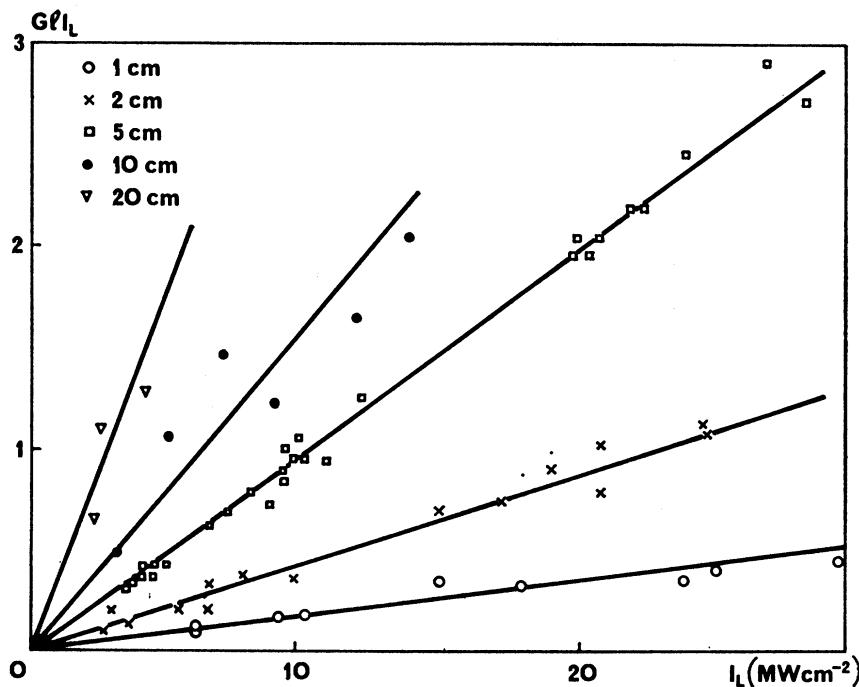Figure 5 gives the logarithm of I_{B2}/I_{B1} as a function

FIG. 5. Measured Brillouin Stokes gain in benzene versus incident laser intensity.

[50] M. Maier, W. Rother, and W. Kaiser, Appl. Phys. Letters **10**, 80 (1967).
[51] A. Kastler, Compt. Rend. **159**, 4233 (1964).

TABLE II. Computed steady-state Brillouin Stokes gain $G=2\pi(\omega_b/\Delta\omega_b)(\gamma^2\rho/\kappa)_s$ and measured gain for different liquids. The bulk-modulus κ is deduced from compressibilities given in the Handbook of the American Physical Society. The electrostrictive constant γ is calculated by means of the Lorentz-Lorenz formula. $\Delta\nu^*$ and v_p^* are the spontaneous-scattering linewidths and the sound velocity as measured by Chiao (Ref. 27).

Liquids	C_6H_6 (benzene)	$C_6H_5CH_3$ (toluene)	$C_6H_5NO_2$ (nitrobenzene)	CS_2 (Carbon disulfide)	$CO(CH_3)_2$ (acetone)	C_6H_{12} (cyclohexane)	CH_3OH (methanol)	$CH_3(CH_2)_4CH_3$ (n hexane)	CCl_4 (carbon tetrachloride)	H_2O (water)
r (6943 Å)	1.50	1.50	1.56	1.62	1.36	1.43	1.33	1.37	1.46	1.33
γ^2	3.06	3.06	4.35	6.5	1.25	1.91	0.9	1.29	2.48	0.9
$10^{-3}v_p$ (m/sec)	1.5*	1.38*	1.56*	1.25*	1.19*	1.35*	1.118*	1.113	1.05*	1.488*
$10^{-10}\kappa$	1.96	1.64	2.82	1.99	1.11	1.41	0.99	0.82	1.74	2.22
$\Delta\nu_{spont}$(MHz) measured	245*	480*		55*	175*	670*	200*		430*	220*
10^3G(cm MW^{-1}) computed	18.5	10	0.143/$\Delta\nu$(cm^{-1})	120	18	5.7	12.4	0.134/$\Delta\nu$(cm^{-1})	7.1	6.4
10^3G(cm MW^{-1}) measured	18	13	4.5	45	10.8	6.8	13.2	19	8	4.8
$\Delta\nu_{spont}$(MHz) from gain			900					212		

of laser intensity for different lengths of interaction. For each length, the exponential character of the amplification is well demonstrated. Figure 6, where gain is plotted versus length, shows the exponential dependence of amplification on length of interaction. From these results, we can compute a gain per unit of length using relation (8):

$$G=18\times10^{-3} \text{ cm MW}^{-1}.$$

This value is in good agreement with the steady-state gain that one can compute using known data listed in Table II and Eq. (6). However, this measurement includes a determination of laser intensity by calorimetric means. Because of the poor precision of this type of measurement and because of inhomogeneities of the beam, a factor of 2 or even 3 is not unlikely in the experimental result, and agreement between experimental and theoretical values is not yet sufficient to demonstrate the purely spatial character of the gain.

When the quarter-wave plate $\frac{1}{4}\lambda$ is rotated through 45°, the laser and Brillouin light have perpendicular polarizations. The gain should be 0 in this case. However, a very small amplification remains that we can relate to stimulated Rayleigh-wing scattering.

B. Spatial Gain in Other Liquids

Filling C_1 and C_2 with identical liquids, it is possible to perform the same type of measurement. Figure 7 demonstrates the exponential dependence of the amplification on laser intensity in various liquids. All these experiments were performed with a length of 5 cm of liquid, except in CS₂, where the length was reduced to 2 cm to eliminate self-focusing. The gains computed from these results are listed in Table II together with the steady-state gains obtained using known data.[52]

The gain in nitrobenzene has been obtained in the experiments described in Sec. III on Rayleigh-wing amplification.
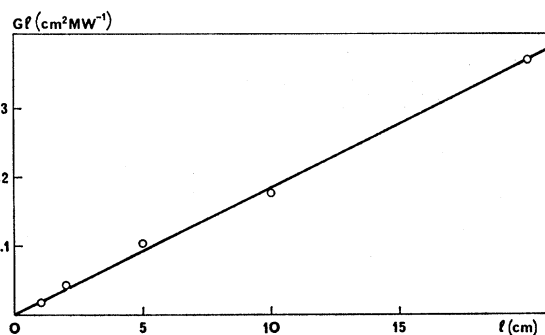
FIG. 6. Measured Brillouin Stokes gain in benzene versus cell length.

[52] American Institute of Physics Handbook (McGraw-Hill Book Co., New York, 1963); Handbook of Chemistry and Physics (Chemical Rubber Publishing Co., Cleveland, Ohio, 1966).

C. Spatial versus Temporal Growth of Brillouin Waves

In most of the liquids studied the spontaneous Brillouin linewidth is known[53] and one can therefore compare the measured and computed gains. It is interesting to examine the ratio of these two gains versus Brillouin linewidth. This ratio is plotted in Fig. 8. The lower point corresponds to carbon disulfide where the measured linewidth is only 55 MHz. In this case the spatial part of the gain is smaller than the steady-state gain. In acetone, the measured linewidth is 175 MHz and the gain is slightly lower than the computed gain. For liquids with larger linewidths, the ratio is constant showing that in these liquids the picture of steady-state amplification is valid. It is also possible from Fig. 8 to deduce the approximation linewidths of our laser and of the Brillouin light used as a test light. They are of the order of 100 MHz. This result shows also
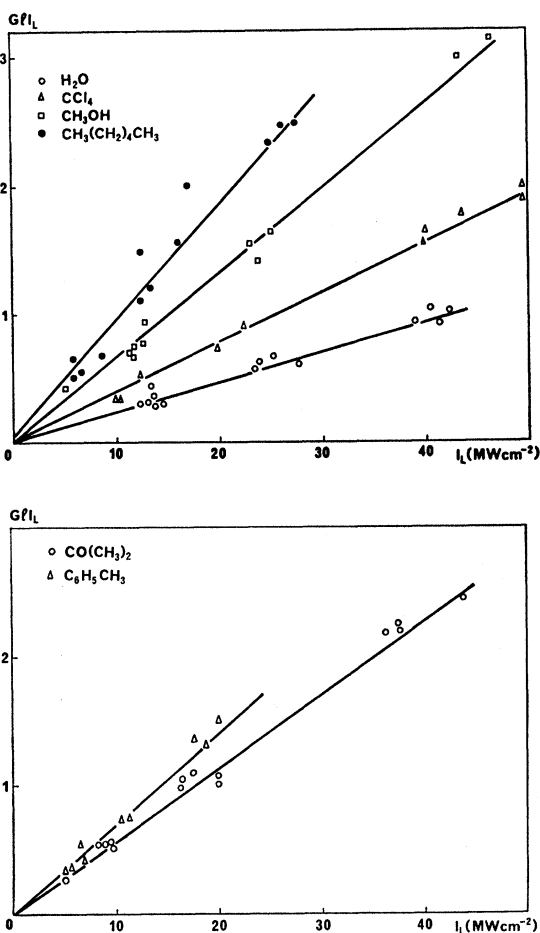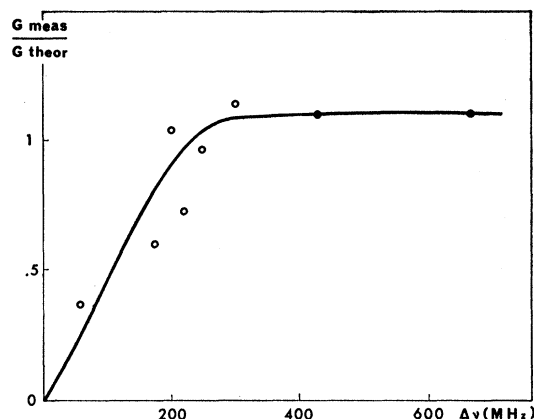
Fig. 8. Ratio of measured to theoretical gain as a function of spontaneous Brillouin linewidth.

that stimulated Brillouin light generated in cell C_2 under high excitation has a much smaller linewidth than the spontaneous Brillouin light and that it is emitted at the central frequency of the Brillouin Stokes line. This was expected, since in this case total gain if of the order of e^{25}, giving a gain narrowing factor of 4.

Another point of interest is that this curve demonstrated definitively the purely spatial character of the gain in benzene. The ratio $G_{\text{measured}}/G_{\text{computed}} = 1.15$ shows simply that our calorimetric estimate of the laser power was optimistic.

A last point is that when the Brillouin linewidth is not well known as in nitrobenzene and in hexane, a gain measurement can lead to a linewidth estimate. If the linewidth so computed is larger than 200 MHz, it corresponds to a steady-state gain and is a good approximation to the spontaneous linewidth.

Results for nitrobenzene and n-hexane are also given in Table II. This type of experiment may then be used to obtain values of spontaneous Brillouin linewidths.

V. GAIN MEASUREMENT IN STIMULATED RAYLEIGH-WING SCATTERING

By changing the liquid in cell C_2 one may explore the amplification spectrum of the liquid in cell C_1. Because of the fact that we use Brillouin shifts in our present experimental setup, the range is limited (0.05 cm^{-1} in gas to 2 cm^{-1} in sapphire). We have studied this amplification on the Stokes Rayleigh wing of nitrobenzene and toluene.

A. Nitrobenzene

This first liquid was chosen because it has a high viscosity which implies a maximum of gain in the spectral range at our disposal, the strong anisotropy of its molecule leads to a large gain, and the Rayleigh scattering has been well studied in spontaneous emis-
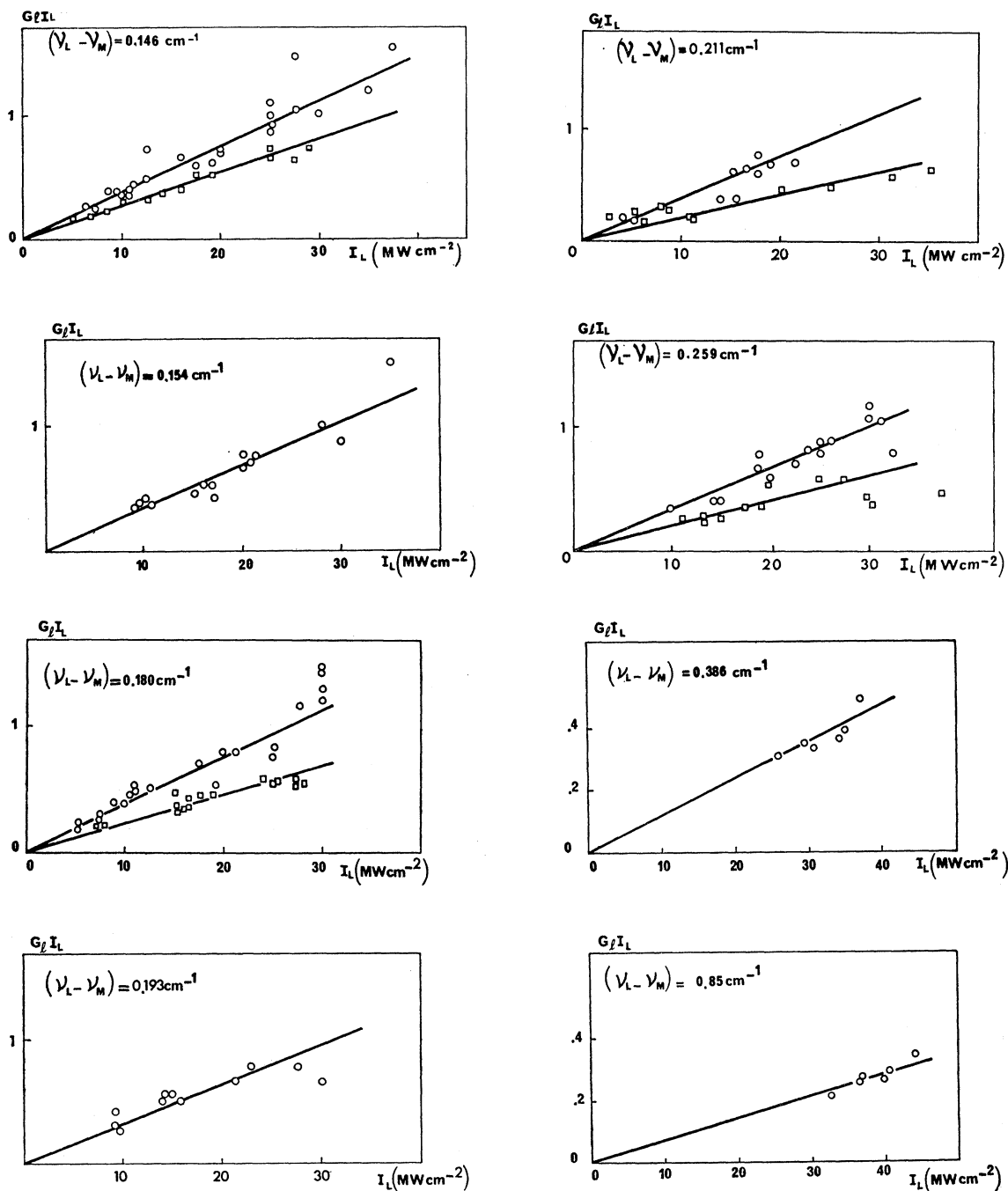
Fig. 7. Measured Brillouin Stokes gain versus laser intensity for different liquids in a 5-cm-long cell.

[53] R. Y. Chiao and P. A. Fleury, *Physics of Quantum Electronics* (McGraw-Hill Book Co.. New York, 1965), p. 241.

FIG. 9. Measured gain versus laser intensity in nitrobenzene at different Stokes frequencies of I_M for parallel (O) and perpendicular (□) polarizations of I_L and I_M.

sion.[54,55] We can therefore compare the measured gain with the gain deduced from Eq. (4) and spontaneous-emission data:

$$G(\omega) = \frac{G\omega\tau}{1+\omega^2\tau^2} \times 10^{-3} \text{ cm MW}^{-1},$$

where the relaxation time is of the order of 4×10^{-11} sec at room temperature.[54] This expression corresponds only to Brownian motion of the molecules and would con-

[54] V. S. Starunov, E. V. Tiganov, and I. L. Fabelinskii, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktskiyu **4**, 262 (1966) [English transl.: Soviet Phys.—JETP Letters **4**, 176 (1966)].

[55] A. Szöke, E. Courtens, and Ben-Deuven, Chem. Phys. Letters **1**, 87 (1967).

tain additional terms of others mechanisms were taken into account.

Logarithms of the measured gains are plotted versus laser intensity at different frequencies in Figs. 9 and 10. These frequencies were obtained by filling C_2 with different liquids or using different solids. At each frequency, measurements were performed with parallel and perpendicular polarizations. The results for $\nu_L - \nu_M$ (in cm^{-1}) are

carbon tetrachloride[6]:	0.146,
acetone[6]:	0.154,
cyclohexane[6]:	0.180,
toluene[6]:	0.193,
benzene[6]:	0.211,
nitrobenzene[6]:	0.228,
anilin[6]:	0.259,
glycerin[6]:	0.386,
lucite:	0.420,
quartz[22]:	0.85.

For parallel polarizations, and when C_2 also contains nitrobenzene (Fig 10), the measured gain is a superposition of two gains, one associated with molecular orientation, the other with stimulated Brillouin scattering. This second gain is negligible for crossed polarizations. A measure of amplification only at the Brillouin frequency would have given too large a Brillouin gain. All these results are summarized in Fig. 11, where gain is plotted versus frequency. Several comments must be made about this figure:

(1) One can deduce the part of the amplification which is due to stimulated Brillouin scattering. This is the gain listed for nitrobenzene in Table II. The correction at the Brillouin frequency for gain associated with molecular orientation is negligible for all other liquids studied in Table II but toluene. The Brillouin gain in toluene, listed in Table II. is given after a correction made from the results of Sec. V C.

(2) A value of τ may be chosen in relation (4) to give a correct fit to the data. From data obtained with
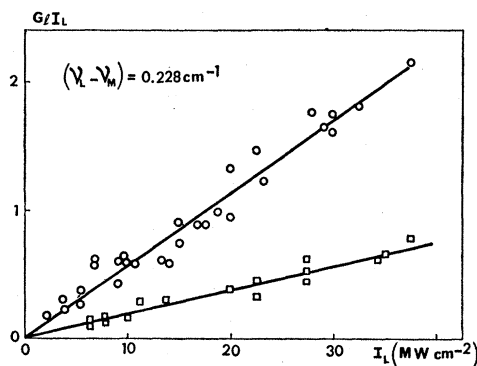

FIG. 10. Measured gain versus laser intensity in nitrobenzene, for parallel (O) and perpendicular (□) polarizations of the beams and at the Brillouin frequency of this liquid.
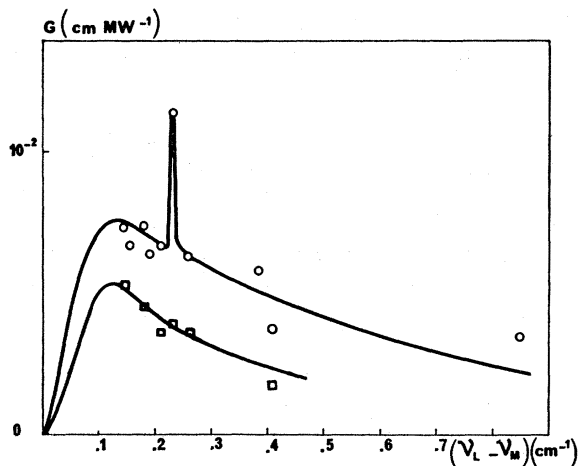

FIG. 11. Measured gain, versus frequency of I_M in nitrobenzene for parallel (O) and perpendicular (□) polarization of I_L and I_M.

parallel polarization, a relaxation time is found:

$$\tau = 4.4 \times 10^{-11} \text{ sec},$$

in agreement with Starunov's observations.[54]

(3) Simple theory predicted a $\frac{4}{3}$ ratio between spontaneous intensities and gains for parallel and crossed polarization. This ratio seems to hold in Starunov spontaneous data.[54] For small frequency shifts $(\nu_L - \nu_M) < 0.2$ cm, this ratio is also found in our data. In the region between 0.2 and 0.4 cm^{-1}, our measured ratio is somewhat small. Points at higher frequencies are obtained with little precision and the $\frac{3}{4}$ ratio cannot be deduced with confidence from these data. In the intermediate region, however, a ratio of $\frac{1}{2}$ would better fit the data. This descrepancy has been observed in several experimental runs over a period of six weeks and is difficult to explain only by experimental uncertainty.

B. Multimoding Effects

Present results have been obtained with a frequency monomode laser. From the Brillouin gain a Brillouin linewidth of 800 MHz at a frequency 0.228 cm^{-1} from the laser line is computed (Table II). It is therefore normal to measure only the orientation gain at 0.259 cm^{-1} from the laser frequency which is 930 MHz on the side of the Brillouin line. But the gain at 0.211 cm^{-1} from the laser frequency, 510 MHz on the side of the Brillouin line, is apparently too low. Several explanations are possible.

(a) Data on the parameters of nitrobenzene are not correct or our estimate of the Brillouin linewidth from gain measurement is too large. A 600-MHz linewidth would explain this low gain at 0.211 cm^{-1} from the laser frequency.

(b) There is a dip in the Rayleigh gain around 0.211 cm^{-1} that would be imaged on the gain measured with crossed polarization.

This effect in turn would lead us to increase the part of the gain due to stimulated Brillouin scattering in our estimate and to decrease accordingly the linewidth. This interpretation would explain why our estimated linewidth is too large, and why the ratio $\frac{3}{4}$ apparently does not hold in this spectral region; but it would not explain the shape of the Rayleigh-wing spectrum that we obtain in this case.

Figure 12 enables us to compare the previous results in the region of the Stokes Brillouin frequencies with those that are obtained using a laser operating in two modes whose frequencies are 450 MHz apart. In this case, we have the following:

(i) Light at two frequencies is also created in cell C_2 even if one mode is weaker than the other. In fact, we can consider that as soon as one strong laser wave and its associated Brillouin wave are present in a steady state, cell C_2 contains an index grating able to reflect any weak light of a close enough frequency. This grating has a reflection coefficient equal to the ratio of intensities of the laser and Brillouin waves.

(ii) The spectrum of the amplifying liquid in cell C_1 is now the superposition of two shifted spectra of the type in Fig. 1, the shift being equal to the frequency difference between the two laser modes.

(iii) By changing the liquid in C_2, we scan the amplification spectrum of the liquid in cell C_1 with two components and the resulting Brillouin sp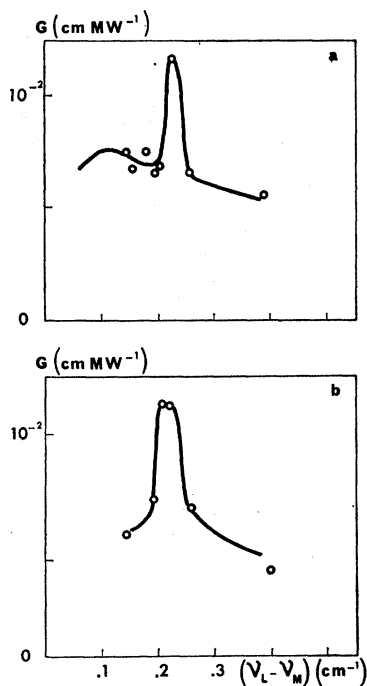ectrum is broader. In the present case the measured Brillouin linewidth is about 1300 MHz. Cells C_1 and C_2 were close enough together so that the gain in the central region of the Brillouin line is about the same in both cases.

C. Toluene

Results in toluene have been obtained with a multimode laser (Fig. 13); the Brillouin line is therefore large but the multimode structure does not change the amplification on the wings of the Rayleigh line. The relaxation time for the orientation of molecules is smaller than in nitrobenzene. The spectrum is therefore wider and since we have not performed any measurement beyond 0.8 cm^{-1} we have only an indication of a maximum in this spectral region that would give us $\tau = 7 \times 10^{-12}$ sec in agreement with other data.[56]
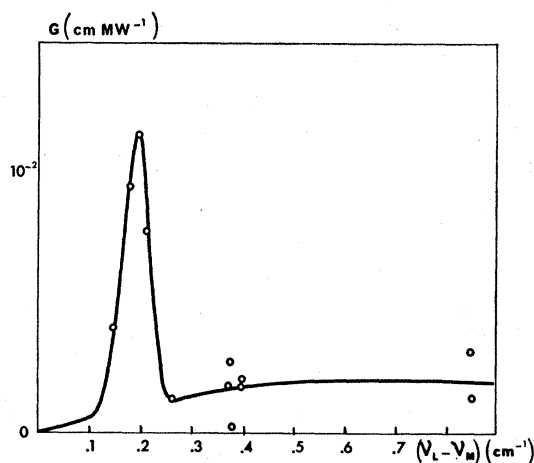
Fig. 13. Measured gain versus the frequency of the light I_M in toluene for multimode excitation.

VI. COMPETITION EFFECTS BETWEEN DIFFERENT PROCESSES

One conclusion of this work is that in most experiments where a liquid is illuminated by a powerful Q-switched laser, at least three processes exhibit spatial gain with a steady-state character but different time constant, the gain at the Brillouin frequency being by far the larger one in all cases. Another important point is that a frequency-independent change of index of refraction is associated with the electrostrictive and orientation effect and can lead to self-focusing of the laser beam. However, in the case of electrostriction (Brillouin scattering) the time constant of this process corresponds to the propagation of a mechanical stress across the beam with the speed of sound and is therefore long even with a focused beam. This process may have some effects, however, in small-scale filaments of the type

Fig. 12. Measured gain versus the frequency of the light I_M in the neighborhood of the Brillouin frequency in nitrobenzene, for (a) monomode and (b) multimode laser emission.

[56] V. S. Starunov, Opt. i Spektroskopiya 19, 300 (1965) [English transl.: Opt. Spectry. (USSR) 18, 165 (1965)].

described by Brewer and Townes.[57] Apart from this case, the only important variation of index of refraction is then related to orientation effects with a short time constant ($\sim 10^{-11}$ sec). All the above processes, amplification from noise at different frequencies and self-trapping will therefore compete in the liquid under strong laser excitation, and the competition will be completely dependent on the characteristics of the exciting light. One can consider several typical cases.

A. Very Monochromatic Laser in Nonfocusing Liquid

Under parallel-beam or focused-beam excitation, the steady-state Brillouin scattering is always reached, and when the power of the laser is increased the liquid behaves as a good reflector. This corresponds to a gain at the Brillouin frequency of the order of e^{25}, and therefore a much smaller gain at a Raman frequency. In this case, an observation of spontaneous Raman light amplified in the liquid will be completely disturbed and subsequent phenomena such as reflection of the Brillouin light into the laser may further complicate the picture. Figure 14 illustrates this situation. The laser beam is focused into a mixture of 20% CS_2 and 80% acetone. The beam is entirely reflected and does not go beyond the focal region.[58]

B. Broad-Band Laser in Nonfocusing Liquids

In this case, amplification at Raman frequencies may become larger than at the Brillouin frequency and a study of stimulated Raman emission by measurement of amplification from noise may be possible.

C. Study of Focusing Liquids

In a given geometry, there is an intensity threshold for each liquid. Above this threshold a part of the laser beam is trapped in a very short time (10^{-11} to 10^{-12} sec). Dissipation of energy in filaments and their subsequent
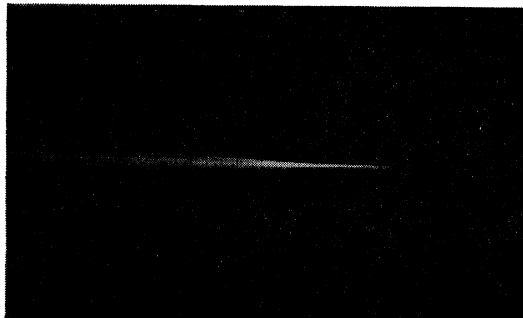

Fig. 14. Propagation of the focused laser beam in an 80% acetone, 20% carbon disulfide mixture. The laser light here is completely reflected by stimulated Brillouin scattering.

disparition is related to orientation effects and stimulated Raman scattering in the early part of their life and then to stimulated Brillouin scattering which gives a growing acoustic wave inside the filament with a time constant of the order of 10^{-9} sec.

Since gain at the Brillouin frequency is much larger than at the Rayleigh wings or Raman frequencies the predominance of the first two processes will last only a few times 10^{-10} sec.

In strongly focusing liquids such as carbone disulfide and nitrobenzene, gains at Raman and Brillouin frequencies are small just below threshold and stimulated emission from noise will be observable only in filaments above threshold. In less-focusing liquids such as acetic acid and cyclohexane the threshold for self-focusing is higher and below-threshold competition between stimulated Brillouin and Raman amplification from noise will have the same dependence on laser linewidth as in nonfocusing liquids.[59]

This discussion shows only, in most experiments, a measure of gain on a signal is better suited than a measure of amplification from noise to determine the gain due to stimulated emission at different frequencies.[60]

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[57] R. G. Brewer and C. H. Townes, Phys. Rev. Letters **18**, 196 (1967).

[58] G. Bret and M. Denariez, J. Chem. Phys. **64**, 222 (1967).

[59] G. Bret and M. Denarier, Appl. Phys. Letters **8**, 151 (1966).

[60] This was an early remark made by N. Bloembergen when abnormal gains were observed at Raman frequencies in focusing liquids.

FIG. 14. Propagation of the focused laser beam in an 80% acetone, 20% carbon disulfide mixture. The laser light here is completely reflected by stimulated Brillouin scattering.

FIG. 3. Spectral structure of the laser beam observed with a 10-cm-thick Perot-Fabry interferometer: (a) monomode structure and (b) bimode structure.