ted which show that the damping exceeds the Feynman value above 1.4°K for the larger orifice and above 0.8°K for the smaller orifice. This increased damping is believed to be caused by turbulent normal fluid flow. There also were no undamped oscillations at any measurable level.

A third kind of flow region occurred at velocities greater than 1.5 cm/sec for the large orifice and 4.6 cm/sec for the small orifice. Qualitatively the liquid motion in this region is characterized by almost total energy loss on the first $\frac{1}{4}$ cycle of the motion. We believe that at velocities an order of magnitude greater than the critical velocity, more complex and complete superfluid turbulent flow occurs than that pictured by the Feynman model.

We have measured in detail the energy loss associated with superfluid helium flow through orifices of radii 5.6×10^{-2} and 1.7×10^{-2} cm. The results of this experiment agree quantitatively with Feynman's model. At certain temperatures a critical velocity was measured for each orifice below which no energy loss was found. The value of the critical velocity for the large orifice was approximately 25% lower than predicted by Feynman and for the small orifice about 300% higher than predicted. Above this critical velocity we found a range of velocities where the energy loss for each orifice agrees with the Feynman model.

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THEORETICAL PREDICTION OF THE STIMULATED THERMAL RAYLEIGH SCATTERING IN LIQUIDS*

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During the past few years, stimulated light scattering from liquids has been studied both theoretically and experimentally by many workers.[1] In this connection, two phenomena—the stimulated Brillouin effect[2] and stimulated Rayleigh-wing scattering[3]—have been of recent interest. In the spontaneous scattering of light from liquids, the corresponding Brillouin and Rayleigh-wing scatterings have been observed.[4] At the same time, however, there exists another important component of the spontaneously scattered light for which a corresponding nonlinear optical process has not yet been observed. This is the ordinary thermal Rayleigh scattering (which we shall simply call Rayleigh scattering, as opposed to the Rayleigh-wing scattering mentioned above) which usually constitutes the bulk of the unshifted component in the well-known Brillouin-Rayleigh triplet.[5]

The origin of spontaneous Rayleigh scattering is well understood. It is associated with nonpropagating thermal fluctuations which manifest themselves in light scattering indirectly, through their ability to influence the density secondarily. This is required because in a description in which the density and temperature are taken as independent thermodynamic variables, the dielectric constant is insensitive to changes in temperature, i.e., $T(\partial\epsilon/\partial T)_\rho \ll \rho(\partial\epsilon/\partial\rho)_T$. For this reason[6] also, the direct production of thermal fluctuations by electric fields present in intense light beams is small—much smaller than the direct production of density fluctuations through electrostriction. Accordingly, the stimulated Rayleigh effect has not yet been observed experimentally.

In the present Letter, we discuss a different kind of direct coupling between the light and thermal fields which, indeed, is strong enough to allow the existence of the stimulated Rayleigh effect in back-scattering, provided that certain easily satisfied conditions are met. The mechanism which we propose is the production of thermal energy following light absorption through electronic transitions in liquids (e.g., colored solutions) which absorb laser light. While Brueckner and Jorna[7] have indicated the possible importance of absorptive heating in causing laser-induced instabilities in fluids, they did not obtain the specific conclusions reached in the present work.

In the following, we discuss the combined influence of absorptive heating and electrostriction on the solutions of the linearized hydrodynamic equations for fluids, under the assumption that the absorbed electromagnetic energy is instantly and locally thermalized (so that heat production is proportional to the local value of the square of the optical electric field). The possible effects of fluorescence and of long-lived excited states are ignored in the present treatment. The equation which we obtain for the nonlinear susceptibility shows the effects of electrostriction and absorptive heating to be linearly independent to a high degree of approximation. Hence, the ordinary stimulated Brillouin gain, as a function of frequency, remains unchanged. The contribution associated with the heating shows three maxima, one each lying in the lower frequency wing of both the Stokes and anti-Stokes Brillouin lines, while the third, and strongest, lies in the anti-Stokes wing of the thermal Rayleigh peak. Because the resonances in the gain function are narrow, convolution with the laser profile is necessary in order to relate the present results to laboratory situations. The results obtained after convolution suggest that (1) for laser light absorption coefficients greater than a critical value [see Eq. (17) and Table I], the stimulated Rayleigh line exhibits a lower threshold than the stimulated Brillouin line, (2) the stimulated Rayleigh gain is at least twice that of the Brillouin doublet arising from absorptive heating, and (3) the stimulated Rayleigh line shows an anti-Stokes shift approximately equal to the half-width of the laser intensity distribution.

The linearized hydrodynamic equations have been given by Hunt.[8] Mountain[9] has recently employed them in his study of spontaneous light scattering from liquids. After modification to include the effects of electrostriction and absorptive heating, these equations are

$$\frac{\partial^2}{\partial t^2}\rho-\frac{c_0^2}{\gamma}\nabla^2\rho-\frac{\eta}{\rho_0}\frac{\partial}{\partial t}\nabla^2\rho-\frac{c_0^2\beta\rho_0}{\gamma}\nabla^2T=-\frac{1}{4\pi}\left(\frac{n^2-1}{2}\right)\left(\frac{n^2+2}{3}\right)\nabla^2E^2, \tag{1}$$

$$\rho_0c_v\frac{\partial}{\partial t}T-\lambda\nabla^2T-\frac{c_v(\gamma-1)}{\beta}\frac{\partial}{\partial t}\rho=\frac{1}{4\pi}nc\alpha E^2, \tag{2}$$

Table I. Material parameters, α_{cr}, and gain ratios for representative liquids.

Liquid	n	γc_v (10^7 erg/g °K)	β (10^3 °K^{-1})	c_0 (10^5 cm/sec)	Γ_B[a] (Mc/sec)	Γ_R[b] (Mc/sec)	α_cr[c] (cm^{-1})	$(G_\mathrm{R})_\mathrm{max}/(G_\mathrm{BD})_\mathrm{max}$[c]
Carbon tetrachloride	1.46	0.84	1.18	0.95	630	18	0.25	3.60
Acetone	1.36	2.1	1.32	1.19	270	21	0.34	2.65
Methyl alcohol	1.33	2.5	1.18	1.21	300	12	0.31	2.76
Carbon disulfide	1.63	0.95	1.14	1.17	65	37	0.62	2.07
Benzene	1.50	1.7	1.18	1.17	350	27	0.65	2.83
Ethyl ether	1.35	2.3	1.51	1.03	290	16	0.36	2.71
Water	1.33	4.2	0.20	1.46	380	26	3.10	2.91

[a] Fleury and Chiao, Ref. 5.
[b] Calculated from $2\lambda k^2/\rho_0\gamma c_v$
[c] Assuming $\Gamma_L = 0.025$ cm^{-1}

where the notation is standard, ρ and T representing the density and temperature fluctuations, ρ_0 being the unperturbed density, α the light absorption coefficient for the liquid at the laser frequency, n the index of refraction, and c the speed of light. The terms on the right-hand sides of Eqs. (1) and (2) represent the electrostrictive and absorptive couplings with the electromagnetic field. [To be exact, only the low ($\ll$optical) frequency components of E^2 should be included in Eq. (2).] The Lorenz-Lorentz law has been assumed in calculating the electrostrictive coupling constant.

The electromagnetic field is analyzed through Maxwell's equations using nonlinear polarization terms as described by Bloembergen.[1] The nonlinear polarization is

$$P_{NLS}(t) = M^{-1}\rho(t)\alpha_{\mathrm{mol}}[\tfrac{1}{3}(n^2+2)]^2E(t) \tag{3}$$

with $\rho(t)$ being determined through Eqs. (1) and (2), M being the molecular mass, α_{mol} the molecular polarizability, and $\frac{1}{9}(n^2+2)^2$ a local-field correction.

The method of solution of Eqs. (1) and (2) is the usual steady-state approach where the fields are written, for the case of back-scattering, as

$$E_L(t) = \tfrac{1}{2}\{E_L\exp[-i(k_Lz-\omega_Lt)] + E_L^*\exp[i(k_Lz-\omega_Lt)]\}, \tag{4}$$

$$E_S(t) = \tfrac{1}{2}\{E_S\exp[i(k_Sz+\omega_St)] + E_S^*\exp[-i(k_Sz+\omega_St)]\}, \tag{5}$$

E_S being the scattered field, and

$$\rho(t) = \tfrac{1}{2}[\rho e^{-i(kz-\omega t)} + \rho^* e^{i(kz-\omega t)}], \tag{6}$$

where $k = k_L + k_S$ and $\omega = \omega_L - \omega_S$. An equation similar to Eq. (6) holds for $T(t)$. Upon substitution of Eqs. (4)-(6) into Eqs. (1) and (2), neglect of source terms oscillating at frequencies other than ω, elimination of T from the resulting equations by substitution, solution of the remaining equation for ρ in terms of E_L and E_S, and substitution of this result into Eq. (3), we obtain, for the nonlinear susceptibility,

$$\chi_{NLS}(\omega_S) = \frac{\alpha_{\mathrm{mol}}}{4\pi M}\left(\frac{n^2+2}{3}\right)^2|E_L|^2\left[\frac{\beta nc\alpha\rho_0c_0{}^2k^2/\gamma + [\frac{1}{2}(n^2-1)][\frac{1}{3}(n^2+2)]k^2(-\lambda k^2+i\rho_0c_v\omega)}{[-\omega^2+(c_0{}^2k^2/\gamma)-i(\eta k^2/\rho_0)\omega](-\lambda k^2+i\rho_0c_v\omega)+i[(\gamma-1)/\gamma]\rho_0c_v\omega c_0{}^2k^2}\right]. \tag{7}$$

Equation (7) reduces to the standard result for Brillouin scattering provided that α and λ are negligible—conditions which have been satisfied in standard experiments to date. Equation (7) shows resonances in the vicinity of $\omega = 0$, $\pm\omega_{\mathrm{B}}$, where $\omega_{\mathrm{B}} = c_0k$, the Brillouin line shift. We shall now limit our attention to these resonances.

$\underline{\omega \cong 0}$.—In this region one can write for the imaginary part of the nonlinear susceptibility [noting that the second term in Eq. (7) is far off resonance]

$$\mathrm{Im}\chi_{NLS} = -\frac{\alpha_{\mathrm{mol}}}{4\pi M}\left(\frac{n^2+2}{3}\right)^2|E_L|^2\frac{\beta nc\alpha}{\gamma c_v}\frac{\omega}{\omega^2+(\frac{1}{2}\Gamma_{\mathrm{R}})^2}, \tag{8}$$

where $\Gamma_{\mathrm{R}} = 2\lambda k^2/\rho_0\gamma c_v$, the spontaneous Rayleigh linewidth. Because of the narrowness of this function, it is necessary to convolve it with the laser line for practical purposes. Assuming a Lorentzian profile for the laser line gives, for the Rayleigh gain per unit length,

$$G_{\mathrm{R}} = -\frac{\alpha_{\mathrm{mol}}}{M}\left(\frac{n^2+2}{3}\right)^2 \times k_S|E_L|^2\frac{\beta nc\alpha}{\gamma c_v}\frac{\omega}{\omega^2+(\frac{1}{2}\Gamma_L+\frac{1}{2}\Gamma_{\mathrm{R}})^2}. \tag{9}$$

This curve has a positive maximum equal to

$$(G_{\mathrm{R}})_{\mathrm{max}} = \frac{\alpha_{\mathrm{mol}}}{M}\left(\frac{n^2+2}{3}\right)^2k_S|E_L|^2\frac{\beta nc\alpha}{\gamma c_v}\frac{1}{\Gamma_L+\Gamma_{\mathrm{R}}}, \tag{10}$$

indicating growth in the backscattered wave. The line is $\overline{\text{anti-Stokes}}$ shifted by an amount $\frac{1}{2}(\Gamma_L+\Gamma_{\mathrm{R}})$, which is experimentally detectable under normal circumstances. This is in sharp contrast to the cases of stimulated Brillouin and Rayleigh-wing scattering in which laser photons are converted to lower energy photons, the excess energy appearing in the liquid excitations. In the present process, laser photons are converted into higher energy photons, the extra energy being supplied by the thermal fluctuations, which in turn, are created through prior absorption of electromagnetic energy. The anti-Stokes shift can be traced to the fact that in absorptive heating, liquid molecules tend to migrate away from high-field regions (provided $\beta > 0$) thus lowering the index of refraction—in contrast to the situation for the electrostrictive and molecular-alignment effects responsible for stimulated Brillouin and Rayleigh-wing scattering.

$\omega \simeq \pm\omega_{\mathrm{B}}$.—In these regions,

$$\operatorname{Im}\chi_{NLS} = \frac{\alpha_{\mathrm{mol}}}{4\pi M}\left(\frac{n^2+2}{3}\right)^2 |E_L|^2 \left[\frac{(\beta n c\alpha/\gamma c_v)(\omega_{\mathrm{B}}^2/\omega)(\omega^2-\omega_{\mathrm{B}}^2)+[\frac{1}{2}(n^2-1)][\frac{1}{3}(n^2+2)](\omega_{\mathrm{B}}^2/c_0^2)\Gamma_{\mathrm{B}}\omega}{(\omega^2-\omega_{\mathrm{B}}^2)^2+(\Gamma_{\mathrm{B}}\omega)^2}\right], \qquad (11)$$

where $\Gamma_{\mathrm{B}} = \eta k^2/\rho_0$ is the spontaneous Brillouin linewidth. The second term leads to the usual Brillouin Stokes component while the first, which has positive maxima in the vicinity of $\pm\omega_{\mathrm{B}}$, gives rise to the Brillouin Stokes–anti-Stokes doublet mentioned previously.

Again, the convolution of the laser line with the gain curve is necessary, and (calling the first term G_{BD} and the second term G_{B}) leads to the result

$$G_{\mathrm{BD}} \cong \frac{\alpha_{\mathrm{mol}}}{M}\left(\frac{n^2+2}{3}\right)^2 k_S |E_L|^2 \times \frac{\beta n c\alpha}{\gamma c_v}\,\frac{\omega\pm\omega_{\mathrm{B}}}{(\omega\mp\omega_{\mathrm{B}})^2+\frac{1}{4}(\Gamma_L+\Gamma_{\mathrm{B}})^2}, \qquad (12)$$

where the upper sign is taken for $\omega > 0$, and the lower sign for $\omega < 0$, and

$$G_{\mathrm{B}} \cong \frac{\alpha_{\mathrm{mol}}}{M}\left(\frac{n^2+2}{3}\right)^3 k_S |E_L|^2 \frac{1}{2c_0^2} \times \left(\frac{n^2-1}{2}\right)\omega\,\frac{\frac{1}{2}(\Gamma_L+\Gamma_{\mathrm{B}})}{(\omega-\omega_{\mathrm{B}})^2+\frac{1}{4}(\Gamma_L+\Gamma_{\mathrm{B}})^2}. \qquad (13)$$

The Brillouin doublet has its greatest positive gain at frequencies $\omega = \pm\omega_{\mathrm{B}} + \frac{1}{2}(\Gamma_L+\Gamma_{\mathrm{B}})$. The maximum gain associated with these peaks is

$$(G_{\mathrm{BD}})_{\mathrm{max}} = \frac{\alpha_{\mathrm{mol}}}{M}\left(\frac{n^2+2}{3}\right)^2 k_S |E_L|^2 \frac{\beta n c\alpha}{2\gamma c_v}\,\frac{1}{\Gamma_L+\Gamma_{\mathrm{B}}}, \qquad (14)$$

with the result that the ratio of stimulated Rayleigh to Brillouin doublet gain is

$$\frac{(G_{\mathrm{R}})_{\mathrm{max}}}{(G_{\mathrm{BD}})_{\mathrm{max}}} = 2\,\frac{\Gamma_L+\Gamma_{\mathrm{B}}}{\Gamma_L+\Gamma_{\mathrm{R}}}. \qquad (15)$$

Inasmuch as this ratio is always greater than 2, the existence of the doublet is probably difficult to establish experimentally. The ordinary Brillouin gain has as its positive maximum (after convolution)

$$(G_{\mathrm{B}})_{\mathrm{max}} = \frac{\alpha_{\mathrm{mol}}}{M}\left(\frac{n^2+2}{3}\right)^3 k_S |E_L|^2 \frac{1}{c_0^2} \times \left(\frac{n^2-1}{2}\right)\omega_{\mathrm{B}}\,\frac{1}{\Gamma_L+\Gamma_{\mathrm{B}}}. \qquad (16)$$

Now, when $(G_{\mathrm{R}})_{\mathrm{max}} > (G_{\mathrm{B}})_{\mathrm{max}}$, the Rayleigh line exhibits the lower threshold and predominates over the Brillouin Stokes line. In terms of the absorption coefficient, this condition is

$$\alpha \gtrsim \alpha_{\mathrm{cr}} = \left(\frac{n^2-1}{2}\right)\left(\frac{n^2+2}{3}\right)\frac{\gamma c_v \omega_{\mathrm{B}}}{\beta n c_0^2 c}\,\frac{\Gamma_L+\Gamma_{\mathrm{R}}}{\Gamma_L+\Gamma_{\mathrm{B}}}, \qquad (17)$$

α_{cr} being a critical value for the absorption coefficient.

Table I gives material parameters for several liquids together with estimates of α_{cr} and the gain ratio. It can be seen that the values of α_{cr} are not inconveniently high, and that the stimulated Rayleigh line may be observable in a number of liquids.

In the foregoing analysis we have neglected a variety of possible influences associated with the over-all rise in temperature through absorptive heating. These could be spurious effects, such as bubble formation, together with less drastic effects such as the alteration of liquid parameters with temperature. (For example, if the sound speed changes during the laser pulse, the effective Brillouin gain could be substantially changed.) While there is no obvious reason why these effects should not considerably alter the phenomena predicted in this paper, experiments on CCl_4-iodine solutions by Rank, Cho, Foltz, and Wiggins[10] indicate substantial agreement with the predictions of this paper.

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STIMULATED THERMAL RAYLEIGH SCATTERING*

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When plane-polarized, monochromatic light is incident on certain liquids such as nitrobenzene, the backscattered light is known to consist of several components: a sharp Rayleigh peak, Brillouin components, and a broad Rayleigh wing. The sharp Rayleigh peak and the Brillouin components are totally polarized, while the Rayleigh wing is depolarized. By the use of light from giant-pulse lasers, the gain-narrowed stimulated radiation corresponding to Brillouin scattering[1] and to the Rayleigh-wing scattering[2] have been observed in some liquids. However, no stimulated radiation corresponding to the polarized Rayleigh peak scattering has been identified experimentally. The purpose of this communication is to report the observation of a new stimulated radiation which is believed to be attributable to thermal Rayleigh scattering.

The polarized Rayleigh peak which can be observed with light of conventional intensity shows no frequency shift and has been interpreted by Landau and Placzek[3] to arise from entropy fluctuations which do not propagate in the scattering medium. Recently, Herman and Gray[4] have shown theoretically that intense laser light incident on an absorbing liquid can produce stimulated Rayleigh scattering from localized thermal fluctuations. Rapid thermalization of the molecules which absorb at the laser frequency and the subsequent generation of large density fluctuations, coherent and localized, leads to the enhancement of the thermal density fluctuations already present in the liquid. The backscattered light in the liquid can then have exponential gain as it travels through the liquid. They predicted that the stimulated thermal Rayleigh line should show a frequency shift of approximately one half of the half-intensity width of the laser light to the anti-Stokes side. Further, a liquid which readily produces this stimulated effect should have a large ratio of its coefficient of volume expansion to its specific heat, and an absorption coefficient of considerable magnitude at the laser frequency.

Unlike other stimulated effects, stimulated thermal Rayleigh light is almost indistinguishable in frequency from the incident light. It was, therefore, necessary to take elaborate precautions to eliminate possible sources of error due to the reflections of laser light from the various optical surfaces. The light from