noting that many properties of the system are insensitive to the statistics of the particles. We then wrote down a set of basis functions for the fermion problem as products of a determinant of plane waves and an eigenstate of the Bose problem. The off diagonal parts of the Hamiltonian were interpreted as a coupling of the fermions to the elementary excitations of the Bose system. It was then shown that this fermion-phonon interaction dresses a moving fermion with its backflow and that two-fermion scattering takes place via the interaction of the backflows. Finally we discussed the expansion parameter for strong interactions and outlined some simplifications of the theory which it allows.

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Stimulated Thermal Rayleigh Scattering in Liquids*

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Additional measurements of the critical absorption coefficient and the anti-Stokes shift of light backscattered from absorbing solutions illuminated by a giant-pulse ruby laser are presented. Frequency shifts of less than the predicted value of one-half the incident linewidth are thought to be due to saturation effects and failure to remove completely the incident light from the interferogram of the backscattered light. The low values of the critical absorption coefficient observed in several liquids are discussed and measurements of the nonlinear attenuation of some liquids are presented.

INTRODUCTION

The stimulated thermal Rayleigh scattering (STRS) was first observed by Rank, Cho, Foltz, and Wiggins[1] in liquids, and in gases by Wiggins, Cho, Dietz, and Foltz,[2] following its theoretical prediction made by Herman and Gray.[3] It is well known[4] that the light scattered from the density fluctuations associated with localized thermal fluctuations in a fluid medium gives rise to the spontaneous Rayleigh peak whose frequency is unshifted from the incident light frequency. When molecules which absorb at the incident light frequency are introduced into the medium, the thermal fluctuations can be greatly enhanced by those molecules which undergo rapid thermalization. It has been shown[3] that in an intense laser light field, the nonlinear gain for the thermal Rayleigh scattering in the medium may reach a value sufficiently high to be competitive with

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other nonlinear gains, e.g., the gain for stimulated Brillouin scattering.

Unlike the spontaneous thermal Rayleigh scattering which produces light usually indistinguishable from the original incident light, STRS exhibits a striking feature which can be used for its identification. The first results reported[1,2] showed that the STRS light was gain-narrowed and anti-Stokes shifted, the shift $\Delta\omega$ being approximately equal to the predicted value[3] given by

$$\Delta\omega = \omega_{SR} - \omega_L = \tfrac{1}{2}(\Gamma_L + \Gamma_R). \qquad (1)$$

ω_{SR} and ω_L are the angular frequencies of the stimulated thermal Rayleigh and laser lights, respectively, and Γ_L and Γ_R are the half-intensity linewidths of the laser and spontaneous thermal Rayleigh, respectively. Values of Γ_R in liquids are very small in comparison with Γ_L, and consequently can be neglected. The observed frequency shift in STRS results from the convolution of the gain function with an assumed Lorentzian laser profile.

To observe a particular process when two or more competing nonlinear processes exist in a medium, it is necessary that the corresponding nonlinear gain be larger than or equal to other existing gains. A comparison of the gains per unit length for STRS and the stimulated Brillouin scattering led to the following condition for the absorption coefficient of the medium, α, with which STRS may be observable:[3]

$$\alpha \geqslant \alpha_{cr} = \left(\frac{n^2 - 1}{2}\right)\left(\frac{n^2 + 2}{3}\right)\frac{c_p(\Delta\omega)_B}{\beta n v^2 c}\frac{\Gamma_L + \Gamma_R}{\Gamma_L + \Gamma_B}, \qquad (2)$$

where α_{cr} is the critical absorption coefficient. The nomenclature used in the above equation is consistent with those in Ref. 1.

Although the previous results[1,2] showed qualitative agreement with the theory, the frequency shift and the values of α_{cr} were both observed to be smaller than predicted. To account for these differences, and to extend the work to other liquids and different absorbers, additional work was carried out. We report here measurements on seven different liquids in various combinations with four absorbers, giving the values of α_{cr} and frequency shifts observed with some explanations of the causes of the discrepancy between the theoretical and experimental results.

EXPERIMENTAL

The spectral profile of backscattered light from a liquid cell containing various dye solutions in the neighborhood of the ruby-laser frequency was studied in an arrangement similar to that described previously.[1,2] A Korad ruby-laser system producing a single-mode giant pulse of power up to 50 MW and pulse duration of about 14 nsec served as a laser-light source. The spectral linewidth of the laser light was observed to be in the range of 0.017 –0.026 cm^{-1}. The laser beam, having a divergence of 6 mrad, was focused into a 10-cm liquid cell by means of a 10-cm focal-length lens. The backscattered light was then photographed through a Fabry-Perot interferometer with various spacers up to 101 mm. The backscattered light was observed simultaneously with the incident laser light, but they were photographed on separate quadrants of the interferogram by means of a combination of a quarter-wave plate and analyzers as used in previous works.[1,2,5] This method of concurrent observation of backscattered and incident laser light made it possible to determine accurately a small frequency shift of the scattered light when used in conjunction with an interferometer having a large spacer. Care was taken to eliminate any possible distortion in interferogram introduced by insertion of the analyzers near the focal plane of the camera which might prevent accurate determination of the frequency shifts. Comparison of line profiles was also possible from the interferograms thus obtained.

The STRS was studied in the following solutions of absorbing substances: iodine solutions in carbon tetrachloride, carbon disulfide, methyl alcohol, toluene, benzene, xylene, and ethylene glycol; $CoCl_2$, $CuCl_2$, and $NiNO_3$ solutions in methyl alcohol; and some organic dyes in various solvents. All the solutions were kept at room temperature throughout the investigation.

It was necessary to investigate the absorption of the laser beam in the dye solutions as a function of incident laser power. The absorption, or more appropriately attenuation, was determined directly from the intensities of incident and transmitted lights at the laser frequency. Two beam splitters, one placed in front of the liquid cell and one behind the cell, served to sample the incident beam and the transmitted beam, respectively, and to direct them onto two MgO blocks. The reflectivity of each beam splitter at the actual angle of operation was found to be approximately 10%. Light reflected from each MgO block was monitored by a fast-response ITT photodiode connected to a Tektronix-519 oscilloscope. The relative intensity of each beam was calculated from the temporal profile of the pulse recorded in the oscilloscope trace. Calibration of the intensity of the transmitted light in relation to the incident intensity was carried out by repeating the measurements at any given incident laser power with the liquid cell removed from the optical path, correction being made for the transmission of the cell windows.

In addition, the transmission of the various liquids was measured at low intensity using a white-light source, a 6943 Å filter, and a photomultiplier.

RESULTS AND DISCUSSION

Stimulated thermal Rayleigh scattered light was observed in most of the solutions studied and showed the same experimental properties as those discussed in the previous report.[1] The general character of STRS is clearly illustrated in an interferogram reproduced in Fig. 1. The interferogram was obtained for a solution of iodine in ethylene glycol having an absorption coefficient of 0.045 cm^{-1} measured with a light source of conventional intensity at 6943 Å. The incident laser power was 12 MW. In
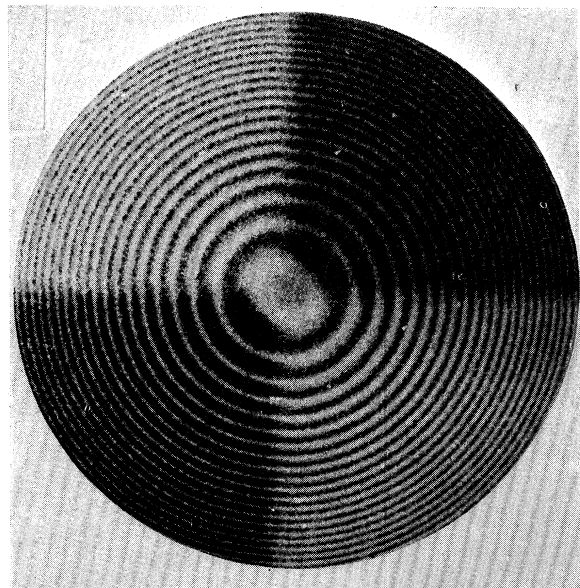
FIG. 1.   Interferogram with a 101-mm spacer showing the incident and back-scattered light from ethylene glycol with iodine added to produce an absorption coefficient of 0.045 cm^{-1}. The darker quadrants are due to the incident laser light. The sharp rings in the lighter quadrants are due to the anti-Stokes shifted STRS.

TABLE I. Observed and calculated values of the critical absorption coefficient in several liquids with various added absorbers. The measured values of the anti-Stokes shift are also given.

| Liquids | α_{cr} (cm^{-1}) | | | | Measured anti-Stokes shift (cm^{-1}) |
	Calculated	Observed	Gires[6]	Dye	
CH_3OH	0.31	0.32		$CoCl_2$	0.0071
CH_3OH	0.31	0.30		$CuCl_2$	0.0021
CH_3OH	0.31	0.32		$NiNO_3$	0.0046
CH_3OH	0.31	0.19		I_2	0.0071
CCl_4	0.25	0.11	0.08	I_2	0.0055
CS_2	0.62	0.24		I_2	0.008
C_6H_6	0.65	<0.018	0.2	I_2	0.0064
$C_6H_5CH_3$	0.46	0.008		I_2	0.0069
$C_6H_4(CH_3)_2$	0.58	0.006		I_2	0.0072
$(CH_2OH)_2$	0.52	0.039		I_2	0.013

Fig. 1, the lighter sectors are the portion of the interferogram produced by the backscattered light while the darker sectors correspond to that of the incident laser light. A gain-narrowed and anti-Stokes shifted STRS can easily be identified in the backscattered sectors, the measured frequency shift being 0.013 cm^{-1}, which is equal to one-half of the linewidth of the incident light. As is evident in Fig. 1, stimulated thermal Rayleigh scattered light is linearly polarized with no observable depolarization, its direction of polarization being unchanged from that of the laser light.

Onset of STRS in an absorbing liquid was observed to depend on the measured absorption coefficient α and the incident laser power in the following way: First, α should be made to exceed a certain critical value, α_{cr} before STRS can be produced. When α is smaller than α_{cr}, stimulated Brillouin scattered light usually dominates the backscattered beam and STRS is absent. Second, the laser power incident on the liquid has to be larger than a threshold power before STRS can be detected.

A summary of the pertinent experimental results obtained in the present investigation is given in Table I. STRS was observed in all the solutions listed in the table. The calculated values of α_{cr} are evaluated by employing Eq. (2) and by using known molecular constants of the various liquids. Organic dye solutions, such as Sudan Schwarz B and Oracet Blue in various solvents, failed to produce backscattered light which showed any measurable frequency shift from the original laser frequency. As can be seen in Table I, the observed values of α_{cr} for the solutions of inorganic dyes other than iodine agree well with the calculated values, while the iodine solutions give rise to con-

sistently lower values of α_{cr} than the calculated values. The observed anti-Stokes frequency shifts of STRS for most of the solutions, given in Table I, are somewhat smaller than the theoretically expected value of one-half the incident linewidth. This disagreement may arise from the saturation in the STRS process permitting an asymmetric gain as discussed previously.[2] It may, however, be a result of superposition of the light due to STRS with a strong backreflected light with no frequency shift.

The values of α_{cr} for some iodine solutions have recently been measured by Gires[6] using a direct method of determination of nonlinear optical gain in the liquids. His observed values are also given in Table I for the purpose of comparison.

As is seen in Table I, the agreement between the observed and calculated values of α_{cr} for certain solutions does not appear to be good. It is noted that the absorption coefficient α as defined is the value at the laser frequency measured at the actual light intensity used. A number of investigators have indeed reported[7] the observation of intensity-dependent nonlinear absorption increases in certain solutions. It seems thus imperative that the relationship between α and incident laser power density should be determined before the agreement can be quantitatively examined. Attempts were made with several solutions and their solvent liquids to find this relationship by means of measuring transmission coefficients for the giant-pulse laser light through a liquid cell containing the sample liquids. Results obtained with a solution of iodine in ethylene glycol and those with pure ethylene glycol are given in Fig. 2, in which the attenuation coefficient $(1/L)\log_{10} \times (I_0/I)$ is plotted against laser power density in the liquid. It is interesting to note that the data for both pure ethylene glycol and the solution of iodine in glycol show a straight-line relationship between the attenuation coefficient and laser power density, the slopes of the lines being approximately equal. In the experiments, care was taken to maintain a nearly uniform power density for the whole length of the liquid cell by the use of a 60-cm focal-length lens, and the actual size of the
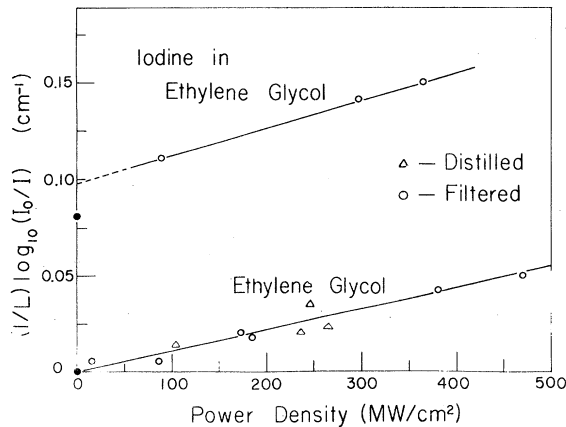
FIG. 2.   Attenuation of light in ethylene glycol and ethylene glycol with iodine for various input power densities. The solid circles indicate the attenuations measured by a conventional light source.

beam at various positions in the liquid was determined to be constant within $\pm 10\%$. With pure ethylene glycol which was passed through "Millipore" filters of $0.1- \mu$ pore size prior to the experiment, no stimulated effect of any kind was detected in either forward or backward directions. When the same liquid was purified further by means of repeated distillation, weak stimulated

Brillouin scattering was observed in the backward direction. As can be seen in the figure, the measured attenuation coefficient for the two cases lie on the same line. The results presented in the figure thus lead to the following conclusions: (1) The transmission of the laser beam through ethylene glycol is not strongly affected either by the purity of the liquid or by stimulated Brillouin scattering in the liquid, and (2) the nonlinear absorption in the solution of iodine in ethylene glycol is apparently attributable to the nonlinear behavior of the solvent itself.

Similar experiments were performed in pure CS_2 and CCl_4, and no attenuation was observed that could not be accounted for by stimulated Brillouin or Raman scattering. It is noted that these liquids gave a much more satisfactory agreement with the theoretical values of α_{cr}.

The small experimental values α_{cr} in ethylene glycol, benzene, etc., may be due to nonlinear absorption which does not thermalize rapidly enough. In addition, the stimulated Brillouin scattering in the solutions may have lower gain than expected because of the formation of complexes, thus resulting in a smaller value of α_{cr}.

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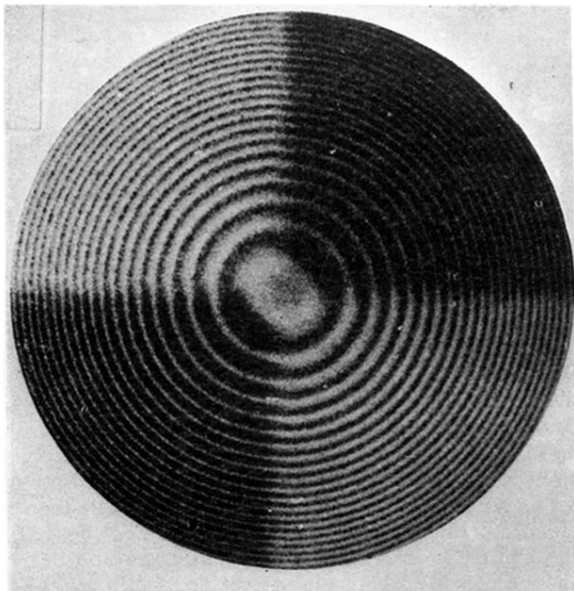
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