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TIME AND FREQUENCY DEPENDENCE OF STIMULATED THERMAL RAYLEIGH SCATTERING

W. Rother, D. Pohl,† and W. Kaiser
Physik-Department der Technischen Hochschule München, München, Germany
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Theoretical investigations of the transient light amplification of stimulated thermal Rayleigh scattering predict a gain proportional to t^2 and, most interesting, a gain at $\omega = \omega_L - \omega_S = 0$ (ω_L and ω_S are the laser and signal frequency, respectively). Experimental investigations using a light amplifier with high-frequency resolution ($\omega/\omega_L \simeq 10^{-8}$) confirm our calculations.

It has been shown by Herman and Gray[1] that in absorbing media light amplification is expected in the neighborhood of the Brillouin frequency and close to the frequency of the incident laser wave. This theory is only concerned with the steady-state situation. The first process, stimulated thermal Brillouin scattering, was investigated in an amplifier-oscillator system with photocells of high time resolution (0.3 nsec). Excellent agreement was found between theory and experiment.[2] The second phenomenon, stimulated thermal Rayleigh scattering (STRS), was observed by several authors[3-6] with time-integrating devices (Fabry-Perot interferometer and photographic plates or time-integrating photodetectors). Discrepancies were noticed[5] when the experimental frequency shifts and the critical absorption coefficients were compared with values predicted from the steady-state theory.

In this Letter we wish to show theoretically and experimentally that STRS is a highly transient phenomenon with a time constant longer than the intense part of the laser pulse. The time and frequency dependence of the amplification was calculated taking the phase of the amplified signal into account. A transient amplification is predicted even for the case where the frequencies of the pump and signal waves are equal. Our experimental observations with high time and frequency resolution support our theoretical investigations.

Our calculations start with the following three equations[1]: (1) the linearized hydrodynamic equation extended to include electrostriction and absorptive heating,[7] (2) the equation of the heat flow, and (3) the nonlinear wave equation for the electromagnetic field. Assuming slow changes of the amplitude of the density and temperature fluctuations, formal solutions of the first two equations were obtained by Fourier transformation. For STRS the signal frequency ω_S is close to the incident laser frequency ω_L, i.e., $\omega = \omega_L - \omega_S \simeq 0$. The corresponding amplitude of the density fluctuation has the form

$$\rho(\omega,t) = (-\gamma^{a} k/16\pi v)\int_{-\infty}^{t} dt'\, E_L(z,t') E_S^{*}(z,t') \exp[(\tfrac{1}{2}\Gamma_R + i\omega)(t'-t)], \quad (1)$$

where $E_L(z,t)$ denotes the amplitude of the electric field of the intense laser wave traveling in the $+z$ direction and $E_S(z,t)$ the weak field of the signal propagating in the opposite direction. The thermo-

optical coupling constant $\gamma^a = \alpha\beta v c^2/\omega_L c_p$ contains the light absorption coefficient at ω_L, α; the cubic thermal expansion coefficient β, the velocity of sound v, and the specific heat c_p. The spontaneous Rayleigh linewidth of the medium under investigation (full width at half-maximum intensity) is represented by Γ_R. Equation (1) was derived under the assumption of small gain.

Introducing $E_S = \hat{E}_S \exp(i\psi_S)$, we obtain from the nonlinear wave equation for the real amplitude $\hat{E}_S$ and for the phase ψ_S the following integrodifferential equations:

$$(\alpha - 2\partial/\partial z)\hat{E}_S(z,t) = -\Gamma_R \hat{g}_R \int_{-\infty}^{t} dt' [I_L(z,t')I_L(z,t)]^{\frac{1}{2}} \hat{E}_S(z,t') \exp[\tfrac{1}{2}\Gamma_R(t'-t)] \times \sin[\psi_S(z,t') - \psi_S(z,t) - \omega(t'-t)], \quad (2)$$

$$2\hat{E}_S(z,t)[\partial\psi_S(z,t)/\partial z] = -\Gamma_R \hat{g}_R \int_{-\infty}^{t} dt' [I_L(z,t')I_L(z,t)]^{\frac{1}{2}} \hat{E}_S(z,t') \exp[\tfrac{1}{2}\Gamma_R(t'-t)] \times \cos[\psi_S(z,t') - \psi_S(z,t) - \omega(t'-t)], \quad (3)$$

with $\hat{g}_R = \gamma^e \gamma^a \omega_L^2/2nc^3 v\rho_0\Gamma_R$ the maximum steady-state gain factor for STRS (effective at $\omega = -\frac{1}{2}\Gamma_R$, anti-Stokes side) and the photoelastic coupling constant $\gamma^e = \rho_0 \partial\epsilon/\partial\rho \simeq \frac{1}{3}(n^2-1)(n^2+2)$. In our small signal theory, where $E_S \ll E_L$, the laser field E_L is considered to be constant.

Equations (2) and (3) allow the calculation of the amplified signal when the time and space dependence of the incident signal and the laser pulse are known. In this short note we present theoretical results for simple initial conditions, where the signal and laser wave are approximated by step functions. In this case analytical solutions are readily obtained which show clearly the main feature of the physical process.

For times $t < 1/\omega$ one finds to a first approximation that

$$\psi_S(z,t) = -\hat{g}_R I_L z[1 - \exp(-\tfrac{1}{2}\Gamma_R t)] + \psi_S^0, \quad (4)$$

i.e., the phase ψ_S changes with time approaching a constant value with a time constant $2\Gamma_R^{-1}$. For many liquids $2\Gamma_R^{-1}$ is approximately 20 nsec which is somewhat longer than the duration of the intense part of the laser light; in other words, a steady-state situation is never reached with our short laser pulses. The time dependence of ψ_S causes a transient contribution to the amplitude $\hat{E}_S$, which predicts a gain even at $\omega = 0$, where the steady-state gain is known to be zero.[1] The constant initial phase ψ_S^0 introduced in Eq. (4) is irrelevant since Eqs. (2) and (3) contain phase differences only.

Expanding the signal field in powers of z and t, one obtains for $\frac{1}{2}\hat{g}_R\Gamma_R I_L z t < 1$ and for a Gaussian intensity profile with a spot size w an outgoing signal

$$P_a = P_i \exp(-\alpha l)[1 + t^2(\tfrac{1}{2}C^2 - \omega C)], \quad (5)$$

where P_i is the incident signal and

$$C = \hat{g}_R \Gamma_R P_L l/2w^2\pi. \quad (5a)$$

It is seen from Eq. (5)[8] that the gain $G \equiv P_a \exp(\alpha l)/P_i - 1$ has the following properties: (1) G rises proportional to t^2; (2) G depends linearly upon ω with a contribution at $\omega = 0$; for $\omega > \frac{1}{2}C$ (Stokes side) an attenuation of the incident signal is predicted. (3) For $\omega = 0$, G is proportional to the square of the absorption coefficient α, since $\hat{g}_R$ is proportional to α. (4) For $\omega = 0$, G is proportional to l^2, where l is the length of the "amplifier." We have made a detailed study of the small-signal gain of STRS and were able to confirm all the properties of G outlined above. Only the first two points will be discussed in detail in this paper.

Our experimental system is depicted schematically in Fig. 1(a). A single-mode ruby laser produced pulses with a peak power of approximately 0.2 MW. By the use of a beam splitter S a weak incident signal P_i and an intense pump pulse P_L were obtained. Both pulses interact collinearly in a small cell[9] of $l = 3$ mm, which is closely located to the focus of two lenses L of $f = 30$ cm. Recently it has been shown[10,11] that the frequency of a ruby laser changes continuously during the emission of a giant pulse. For our laser system a frequency drift of approximately $d\nu/dt = 10$ MHz/nsec has been determined for the intense center of the light pulse.[11] This frequency drift was used in our experiments to establish small frequency differences between the two interacting light waves. Moving the prism Pr, differences in optical path of the pulses P_i and P_L were produced and corresponding frequency dif-

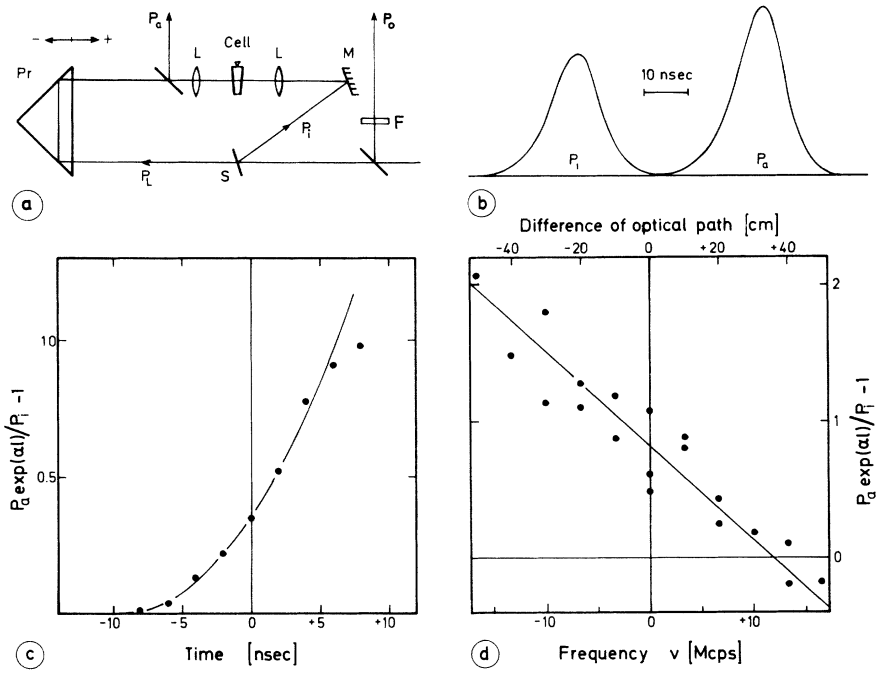

FIG. 1. (a) Schematic of the experimental system. (b) Oscilloscope trace depicting the incoming signal P_i and the amplified signal P_a. (c) Amplification versus time (at $\nu=\omega/2\pi=0$) for a solution of iodine in CCl_4 ($\alpha=0.07$ cm^{-1}). (d) Amplification at the pulse maximum versus frequency difference.

ferences between the interacting pulses of up to $\nu=\pm 15$ MHz were achieved with a resolution ν/ν_L better than 10^{-8}. The incoming light pulse P_0, proportional to P_i and P_L, and the amplified pulse P_a were measured with the same phototube using appropriate delays [see Fig. 1(b)]. The filter F is chosen to make $P_0=P_i$. The overall time constant of the photodetection system is approximately 0.3 nsec. Experiments were performed with CCl_4 ($\Gamma_R/2\pi \simeq 18$ MHz) which was slightly colored by addition of small amounts of iodine to give $\alpha=0.007$ cm^{-1} at the ruby frequency. Measurements made with methanol colored by $CoCl_2$ gave results identical to those reported here.

By evaluating oscilloscope traces such as that shown in Fig. 1(b), the time dependence of $G=P_a\exp(\alpha l)/P_i-1$ was determined. Figure 1(c) presents experimental data for the case of $\omega=0$. The maximum of the laser pulse is taken arbitrarily as the zero point of the time scale. It is interesting to see that the gain increases monotonically during the whole laser pulse; there is no indication of a steady-state situation. For a quantitative comparison with the theory outlined above, a parabola of the form $G=\frac{1}{2}(t+t_0)^2C^2$ was drawn through our experimental data. The parameters obtained in this way are $t_0=-9.5$ nsec and $C=9\times10^{-2}$ $nsec^{-1}$. The determination of t_0 connects our real pulse with the time zero point of the step function used in Eq. (5). It will be shown below that the value of t_0 obtained here is consistent with other independent experiments, e.g., where the frequency dependence of G was investigated [Fig. 1(d)]. From Eq. (5a) we calculate $C=11\times10^{-2}$ $nsec^{-1}$ using the following numbers: $n=1.46$, $v=0.95\times10^5$ cm/sec, $\rho_0=1.6$ g/cm^2, $\beta=1.22\times10^{-3}$ $°K^{-1}$, $c_p=0.84\times10^7$ erg/g °K, $\alpha=0.07$ cm^{-1}, $l=3$ mm, $P_L=0.2$ MW, and $w=1.7\times10^{-2}$ cm. The good agreement between the value of C obtained from Fig. 1(c) and the value calculated from Eq. (5a) is better than expected considering the large number of parameters entering the calculation and the assumptions made in theory. In Fig. 1(d) the frequency dependence of G_m is shown, where G_m represents the gain at the maximum of the laser pulse [$t=0$ in Fig. 1(c)]. The upper scale in Fig. 1(d) indicates the difference in optical path between the two light pulses P_i and P_L. Figure 1(d) shows clearly the expected linear dependence of G upon ω. In addition, the positive gain for $\omega=0$ and the attenuation at the Stokes side for larger ω should be emphasized. When the solid line in Fig. 1(d) is com-

pared with Eq. (5), $G = t_0^2(\frac{1}{2}C^2 - \omega C)$, and we obtain the following parameters: $t_0 \simeq 9$ nsec and $C = 14 \times 10^{-2}$ nsec^{-1}. The value of t_0 agrees well with the number estimated from the time dependence of G [Fig. 1(c)]. The accuracy of C is limited here due to the uncertainty in the absolute frequency scale.

We have measured for $\omega = 0$ the dependence of G upon absorption coefficient α between $\alpha = 0.03$ and 0.16 cm^{-1}. Good agreement was found with the α^2 dependence predicted from Eq. (5). The parameters t_0 and C obtained from this investigation agree well with the data discussed above.

In conclusion it should be noted that the good agreement between theoretical and experimental transient gain allows a ready determination of material parameters. For instance, the value of c_p can be determined with a single laser pulse. A detailed account of this work will be published elsewhere.

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†Present address: IBM Research Laboratories, 8803 Rüschlikon, Switzerland.

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PIEZOELECTRIC EFFECTS IN LIQUID CRYSTALS*

Robert B. Meyer
Gordon McKay Laboratory, Harvard University, Cambridge, Massachusetts 02138
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A theory of the relationship between curvature strains and electric polarization in liquid crystals is developed in analogy to piezoelectric theory in ordinary crystals. The theory may explain some recently observed phenomena in nematic liquid crystals.

There is currently a great deal of interest in electric field effects in liquid crystals, especially in connection with electro-optical phenomena. Although it has been recognized that an electric field can affect the internal structure of a liquid crystal by acting on the anisotropy of the dielectric constant, many of the observed field effects cannot be explained by this interaction alone. It has been suggested occasionally that nematic liquid crystals may be ferroelectric, which would aid in explaining some of the unusual field effects. However, the generally accepted symmetry properties of nematic and cholesteric liquid crystals are incompatible with ferroelectricity.[1] In this Letter, another kind of interaction between an electric field and liquid-crystal structure is shown to be compatible with the symmetry properties generally attributed to liquid crystals, and it is shown how this interaction can explain some unusual effects observed in nematic liquid crystals.

Frank has demonstrated through symmetry arguments the intrinsic relationship between splay strains and electric polarization in liquid crystals; both splay and polarization are manifestations of polar symmetry.[1] In his curvature elastic theory of liquid-crystal structure, Frank shows that if there is an intrinsic polarity in the molecular ordering of a liquid crystal, then that liquid crystal in its lowest energy structure will tend to be uniformly splayed and ferroelectric. He notes also that since no continuous three-dimensional structure can contain uniform splay, this polar liquid crystal would have a fine domain