Stimulated Raman scattering of surface electromagnetic waves

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Stimulated Raman scattering of surface electromagnetic waves is investigated experimentally and theoretically. It is found that, in contrast to other nonlinear interactions involving surface electromagnetic waves, allowance must be made in the study of stimulated Raman scattering for pump depletion and for size effects. The validity of the theoretical results is confirmed by their good qualitative agreement with the experimental data.

1. INTRODUCTION

The advent of simple and effective methods of exciting surface electromagnetic waves (SEW) in the optical band has initiated extensive investigations of various optical effects involving these waves.[1] These investigations have shown that SEW can exert a strong and often decisive influence on many phenomena accompanying interaction of radiation with surfaces of condensed media, primarily metals. Investigations of nonlinear interactions in which SEW participate are therefore most timely. Detailed studies have been made thus far mainly of zero-threshold nonlinear optical effects such as second-harmonic generation,[1-3] nondegenerate[1,4] and degenerate[5-7] four-wave mixing of surface electromagnetic waves, two-photon luminescence,[8] and others. The study of threshold nonlinear optic effects involving SEW, foremost among which is stimulated scattering (SS), is only in its formative stage, since their observation requires that the initial energy density (or intensity) of the source radiation exceed a certain threshold but must not damage the metal surface. Even the first studies, however, of stimulated temperature scattering[9] and stimulated Raman scattering (SRS)[10] have shown that these effects have a number of anomalies that greatly distinguish stimulated scattering from other nonlinear interactions involving SEW. This was the main motive for further research. The importance of this research was demonstrated in a recent rather detailed study[11] of stimulated temperature scattering (STS) into surface electromagnetic waves. A connection was established there between STS and the formation of surface periodic structures (SPS), a phenomenon attracting much attention recently (see the review by Akhmanov *et al.*[12]). It was found, in particular, that in many cases stimulated temperature scattering is the initial stage of formation of surface periodic structures. Allowance for the influence exerted on STS by surface electromagnetic seed waves and by size effects has contributed to the detection and interpretation of new types of surface periodic structures.

The present paper is devoted to a detailed theoretical and experimental investigation of SRS of SEW, first reported in Ref. 10. These investigations have shown in particular that, just as in the case of STS, it is wrong to neglect size effects in SRS. It has also been shown that pump depletion must be taken into account in steady-state stimulated scattering. We emphasize that although these effects were previously disregarded, they may have to be taken into account in many studies of other nonlinear interactions involving SEW.

2. LINEAR EXCITATION OF SEW AT LASER FREQUENCIES

Just as in most experiments on nonlinear interactions involving SEW, a surface electromagnetic wave at the laser frequency ω_L was excited in Ref. 10 by the frustrated total internal reflection (FTIR) method. Following Kretschmann's linear FTIR procedure,[1] we consider a metallic film of thickness l (medium 2 of Fig. 1) sandwiched between two dielectrics. We denote the dielectric constants of the media by ε_j (Fig. 1), where $\mathrm{Re}\,\varepsilon_1,\varepsilon_3>0$; $\mathrm{Im}\,\varepsilon_1,\varepsilon_3=0$; $\mathrm{Re}\,\varepsilon_2(\omega)<0$; $\mathrm{Im}\,\varepsilon_2(\omega)\ll|\mathrm{Re}\,\varepsilon_2(\omega)|$. Let a p-polarized electromagnetic wave propagating from the third medium strike the upper (see Fig. 1) boundary $z=l$ of the film at an angle θ_L. The magnetic field of the wave is given by

$$|\mathbf{k}_L|=k_L=(\varepsilon_3)^{1/2}\frac{\omega_L}{c}\sin\theta_L;$$

where

$$k_{\perp L}=\left(\frac{\omega_L^2}{c^2}\varepsilon_3-k_L^2\right)^{1/2},$$

and $\boldsymbol{\rho}$ is the coordinate in the plane of the surface. It is customary in FTIR analysis of SEW excitation to replace the real laser beam, which has a finite transverse size, with a plane unbounded electromagnetic wave. The amplitude of the SEW excited on the lower boundary $z=0$ of the film, with a field distribution in the lower medium

$$\mathscr{H}_L\exp(-i\omega_Lt+i\mathbf{k}_L\boldsymbol{\rho}+q_1z),$$

and the amplitude of the reflected wave

$$h_L\exp[-i\omega_Lt+i\mathbf{k}_L\boldsymbol{\rho}+ik_{\perp L}(z-l)]$$

are independent of position in the plane of the surface. Solution of the Maxwell equations in all media, with allowance for the boundary conditions at $z=l$ and $z=0$, yields the film's amplitude reflection and transmission coefficients $r_L=h_L/H_L$ and $\tau_L=\mathscr{H}_L/H_L$:

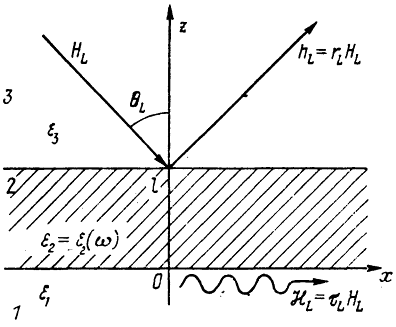

FIG. 1. Schematic of the linear excitation of an SEW by means of FTIR (1 and 3 are dielectrics; 2 is a metallic film).

$$r_L(\omega_L, k_L) = \frac{(1+\alpha_L)(1+\beta_L)\exp(q_{2L}l)+(1-\alpha_L)(1-\beta_L)\exp(-q_{2L}l)}{(1+\alpha_L)(1-\beta_L)\exp(q_{2L}l)+(1-\alpha_L)(1+\beta_L)\exp(-q_{2L}l)}, \tag{1}$$

$$\tau_L(\omega_L, k_L) = \frac{4}{(1+\alpha_L)(1-\beta_L)\exp(q_{2L}l)+(1-\alpha_L)(1+\beta_L)\exp(-q_{2L}l)}, \tag{2}$$

where

$$\alpha_L = \frac{q_{1L}\varepsilon_{2L}}{q_{2L}\varepsilon_1}, \quad \beta_L = \frac{q_{2L}\varepsilon_3}{ik_{\perp L}\varepsilon_{2L}}, \quad q_{jL} = \left(k_L^2 - \frac{\omega_L^2}{c^2}\varepsilon_{jL}\right)^{1/2},$$
$$j=2,3, \qquad \varepsilon_{2L}=\varepsilon_2(\omega_L), \quad \varepsilon_{3L}=\varepsilon_3.$$

Being simple and convenient, this approach actually does not require introduction of a surface electromagnetic wave, and is frequently used to describe nonlinear optical effects. The signal amplitudes in all media are also assumed to be independent of the coordinate in the plane of the surface. In the case of stimulated scattering, however, the effect is strongly influenced by the finite transverse size of the laser beam.[11] Size effects, which we define as effects due to the finite beam dimensions, in particular cause the amplitudes of the initial and signal waves to depend on ρ in all media. We shall show below that size effects can be neglected for linear SEW excitation in the visible and near-IR bands. We obtain then a number of expressions which we shall use in the theoretical description of SRS.

Let the x axis be directed along the vector $\mathbf{k}_L$ and let the laser beam illuminate a limited film area $0 < x < \mathscr{D}$. The amplitudes of the reflected and surface waves as functions of the coordinate x were determined earlier[13] by expanding the radiation incident on the film in plane waves. Here we obtain these dependences by using the approximation (method) of slowly varying amplitudes. An additional advantage is that this approximation is used for theoretical investigations of stimulated scattering of both bulk[14,15] and surface[11] waves. We assume that the field amplitudes in all three media are slowly varying (with scales $1/k_L, 1/k_{\perp L}, 1/q_{jL}$) functions of the coordinates x and z. Solving the Maxwell equation in all the media with allowance for the boundary conditions at $z = 0$ and $z = l$, and neglecting the second derivatives of the slowly varying amplitudes, we obtain for the SEW amplitude $\mathscr{H}_L(x)$ at $z = 0$:

$$\frac{1}{k_L''}\frac{d\mathscr{H}_L(x)}{dx} + \mathscr{H}_L(x) = \tau_L H_L, \tag{3}$$

where

$$\frac{1}{k_L''} = \frac{i}{\tau_L}\frac{\partial \tau_L}{\partial k_L} + ik_L\left(\frac{1}{k_{\perp L}^2} - \frac{1}{q_{2L}^2}\right). \tag{4}$$

Using the natural assumption that the SEW amplitude is zero at $x = 0$, we obtain the $\mathscr{H}_L(x)$ dependence, given the intensity profile in the laser beam. For a rectangular intensity profile, particularly within the area $0 < x < \mathscr{D}$ illuminated by the beam this dependence is

$$\mathscr{H}_L(x) = \tau_L H_L(1-\exp(-k_L''x)). \tag{5}$$

Since the amplitude of the bulk wave is zero outside the laser-spot boundary, the SEW for $x > \mathscr{D}$ is free and attenuates exponentially. The distance at which the intensity of the free SEW, which is proportional to $|\mathscr{H}_L|^2$, decreases by a factor e is called the range of the SEW[1]:

$$L_L = 1/2\,\mathrm{Re}\,k_L''. \tag{6}$$

Note that the slowly-varying-amplitudes approximation is valid if $L_L \gg 1/k_L$. It follows thus from (5) that the SEW intensity is formed at a distance comparable with the SEW range from the illuminated-area edge. For a far-infrared SEW the value of L_L can be several centimeters, which is usually comparable with the transverse beam size $\mathscr{D}$. The size effects play then a substantial role in the FTIR analysis of the SEW excitation.[13] In the visible and near-infrared bands, which are of primary interest to us, the experimental situation corresponds as a rule to the inequality $\mathscr{D} \gg L_L$. This allows us to neglect size effects in linear excitation of SEW by bulk excitation. Note that in a theoretical description of zero-threshold nonlinear interactions the distance from the spot edge, at which the signal-wave intensity is formed, is also comparable with the SEW range.[6] Size effects can be neglected in this case too, but must be taken into account in investigations of stimulated scattering.[11]

Proceeding to find the x-dependence $h_L(x)$ of the reflected wave amplitude at $z = l$, we note the following circumstance. Nonlinear interactions involving SEW are of interest for the most part because the SEW amplitude, for the best-chosen incidence angle θ_L and film thickness l, can greatly exceed the amplitude of the bulk wave incident on the film: $|\tau_L| \gg 1$. Solution of the Maxwell equations with allowance for the last inequality yields an approximate relation between the amplitudes with a clear physical interpretation:

$$h_L(x) = (r_L - \tau_L s_L)H_L + s_L\mathscr{H}_L(x), \tag{7}$$

where

$$s_L(\omega_L, k_L) = \frac{2\beta_L}{(1+\beta_L)\exp(-q_{2L}L)-(1-\beta_L)\exp(q_{2L}l)}. \tag{8}$$

For $|\tau_L| \gg 1$ the reflected wave can thus be represented as a superposition of a "genuine" reflected wave having a reflection coefficient $(r_L - \tau_L S_L)$ and an SEW having an amplitude $s_L \mathscr{H}_L$ in the upper medium. Note that, alongside r_L and τ_L, the complex quantity s_L is an important property of the film when nonlinear optical effects involving SEW are theoretically investigated. Expression (7) can yield the $h_L(x)$ dependence. In particular, for a rectangular beam-intensity profile this dependence can be obtained by substituting (5) in (7).

3. NONLINEAR EXCITATION OF SEW AT THE STOKES FREQUENCY

We assume that the laser-pulse duration is sufficiently long and that the amplitude fields at the initial frequency ω_L and at the frequency ω_S of the first Stokes component have reached their stationary values. The effect of anti-Stokes and higher Stokes components on stimulated Raman scattering can be disregarded. We describe the SRS using the macroscopic theory[14,15] and assume that the lower medium in Fig. 1 has a nonlinear Raman susceptibility

$$\chi \propto \frac{1}{\Omega_0^2-\Omega^2+2i\Omega\Gamma}, \tag{9}$$

where $\Omega = \omega_L - \omega_s$, Ω_0 is the natural frequency, and $\Gamma \ll \Omega_0$ is the damping factor. The source of the wave at the frequency ω_S is a nonlinear polarization in the form

$$\mathbf{P}_S^{NL}=(4\pi)^{-1}\mathbf{D}_S^{NL}(z)\exp(-i\omega_S t+i\mathbf{k}_S\boldsymbol{\rho}),$$

where

$$\mathbf{D}_S^{NL}(z)=\chi\mathbf{E}_L(\mathbf{E}_S\mathbf{E}_L^*)\exp(2q_{1L}+q_{1S})z, \tag{10}$$

E_S and E_L are the electric-field amplitudes in the lower medium at $z=0$ and at frequencies ω_S and ω_L, respectively. The main difficulty in the theoretical investigation of nonlinear interactions involving SEW is that the signal wave and the nonlinear polarization depend differently on z, so that the Maxwell equations in the lower medium cannot be solved directly. This difficulty can be overcome, however, by solving first the problem for a nonlinear polarization proportional to $\delta(z-z')$ where δ is the Dirac delta function.[11] Next, summing the contributions to the signal wave from sources located at different distances from the film, we obtain the following equation which is valid in the approximation of slowly varying amplitudes:

$$\frac{1}{k_S''}\frac{d\mathscr{H}_S(x)}{dx}+\mathscr{H}_S(x)=-\frac{1}{2}\frac{i\omega_S}{c}\frac{\varepsilon_3}{\varepsilon_1}\frac{\tau_S}{ik_{\perp S}s_S}$$
$$\times\int_{-\infty}^{0}[q_{1S}D_{xS}^{NL}(z')\cos\varphi+ik_S D_{zS}^{NL}(z')]\exp(q_{1S}z')dz', \tag{11}$$

where the x axis is directed along the vector $\mathbf{k}_S$; the subscript S means that the corresponding quantity is determined not for ω_L and k_L as above, but for ω_S and $k_s = |k_s|$; $\mathscr{H}_s(x)$ is the SEW magnetic-field amplitude at the Stokes frequency for $z=0$; φ is the angle between the vectors $\mathbf{k}_L$ and $\mathbf{k}_S$. Expressing the electric field at $z=0$ in terms of $\mathscr{H}_s(x)$ and $\mathscr{H}_L(x)$ and integrating in the right-hand side of (11) with allowance for (10), we transform (11) into

$$\frac{1}{k_S''}\frac{d\mathscr{H}_S}{dx}+(1-G_S|\mathscr{H}_L|^2)\mathscr{H}_S=0, \tag{12}$$

where

$$G_S=-\frac{1}{4}\chi\frac{\varepsilon_3}{\varepsilon_1^4}\frac{c^2}{\omega_L^2}\frac{(k_S k_L+q_{1S}q_{1L}\cos\varphi)^2}{ik_{\perp S}(q_{1L}+q_{1S})}\frac{\tau_S}{s_S}. \tag{13}$$

An analysis of (12) shows that for $\operatorname{Re} G_s|\mathscr{H}|^2<1$ the signal-wave intensity is damped as a function of the coordinate x. If, however, the initial laser-beam intensity $I_L=\varepsilon_3^{-1/2}|H_L|^2$ exceeds a certain threshold $I_{L\,\mathrm{thr}}$, the Stokes-wave intensity increases. To determine $I_{L\,\mathrm{thr}}$ we assume as before that a rectangular laser beam illuminates the film area $0<x<\mathscr{D}$. The signal wave is initiated by an SEW spontaneously excited on the film boundary and having an amplitude $\mathscr{H}_S(0)$ at $x=0$. As noted in the preceding section, during the stage of linear excitation of the SEW we can neglect size effects and assume that $\mathscr{H}_L=\tau_L H_L$ for $0<x<\mathscr{D}$. The SRS threshold intensity is then

$$I_{L\,\mathrm{thr}}=\operatorname{Re}(\varepsilon_3^{1/2}G_S|\tau_L|^2)^{-1}. \tag{14}$$

When the beam intensity I_L exceeds the threshold, $|\mathscr{H}_S|^2$ increases exponentially as a function of x:

$$|\mathscr{H}_S|^2(x)=|\mathscr{H}_S|^2(0)\exp\left[(M-1)\frac{x}{L_S}\right], \tag{15}$$

where $L_s=1/2k_S''$ is the SEW range at the frequency ω_S and $M=I_L/I_{L\,\mathrm{thr}}$ is the intensity-to-threshold ratio. It can be seen, however, that if M and the transverse beam dimension $\mathscr{D}$ are large enough, expression (15) describes an unbounded increase of the signal-wave intensity. We shall show in the next section that (15) describes correctly the behavior of our system only if the inequality $|\mathscr{H}_s|^2 \ll |\mathscr{H}_L|^2$ is satisfied. If, on the other hand, the signal SEW becomes comparable with $|\mathscr{H}_L|^2$ it becomes necessary to take into account the reaction of the signal to the distribution of the field of frequency ω_L in all the media. This effect is similar in many respects to pump depletion in the case of stimulated scattering of bulk waves,[14,15] but exhibits some differences when nonlinear interactions involving SEW are considered.

4. PUMP DEPLETION

As shown earlier, even allowance for size effects leads to the conclusion that the equalities $\mathscr{H}_L=\tau_L H_L$ can be violated. The only factor preventing the signal intensity from growing without limit is the decrease of $|\mathscr{H}_L|^2$ [see (12)]. This is due to the presence, in the lower medium at the frequency ω_L, of nonlinear polarization

$$\mathbf{P}_L^{NL}=(4\pi)^{-1}\mathbf{D}_L^{NL}(z)\exp(-i\omega_L t+i\mathbf{k}_L\boldsymbol{\rho}),$$

where

$$\mathbf{D}_L^{NL}=\chi^*\mathbf{E}_S(\mathbf{E}_L\mathbf{E}_S)\exp(2q_{1S}+q_{1L})z, \tag{16}$$

which can be neglected only at low signal-wave amplitudes. Since the signal increases faster at $\varphi=0$ [see (12) and (13)], we assume for the time being that the vector $\mathbf{k}_L$ is also directed along the x axis. By analogy with (12) we obtain then the following equation, which is a generalization of (3) to include the presence of the nonlinear polarization in the lower medium:

$$\frac{1}{k_L''}\frac{d\mathscr{H}_L}{dx}+(1+G_L|\mathscr{H}_S|^2)\mathscr{H}_L=\tau_L H_L. \qquad (17)$$

The expression for G_L is similar to (13) with χ replaced by $-\chi^*$ and the subscripts L and S interchanged.

Simultaneous solution of (12) and (17) yields the functions $\mathscr{H}_L(x)$ and $\mathscr{H}_S(x)$, and determines in particular the values of $\mathscr{H}_{L0}$ and $\mathscr{H}_{S0}$ that are formed along x. Since the choice of the phase of one of the waves is arbitrary, we can take $\mathscr{H}_{L0}$ to be real. Then, equating the derivatives in (12) and (17) to zero, we obtain the desired values

$$\mathscr{H}_{L0}=G_S^{-1/2};\quad \mathscr{H}_{S0}=\left(\frac{\tau_L H_L G_S^{1/2}-1}{G_L}\right)^{1/2}e^{i\psi}\ \text{for}\ I_L>I_{L\,\text{thr}}$$

$$\mathscr{H}_{L0}=\tau_L H_L;\ \mathscr{H}_{S0}=0\ \text{for}\ I_L<I_{L\,\text{thr}}, \qquad (18)$$

where ψ is an arbitrary phase that depends on the phase of the seed SEW. We emphasize that in contrast to zero-threshold nonlinear interactions, the amplitudes $\mathscr{H}_S$ and $\mathscr{H}_L$ are formed by the spot only as a result of pump depletion. Note that the fastest growth of the Stokes wave obtains at $\omega_s=\omega_L-\Omega_0$ [see (9)] and to a wave vector $\mathbf{k}_S$ of length corresponding to the maximum of Re G_S. When these conditions are met, G_S is in general complex. In the steady state, however, as follows from (18), G_S is real. This is accomplished by slight shifts of ω_S and k_S from their optimum values, so that G_S assumes the maximum positive-definite value. Without dwelling on an analysis of Eqs. (12) and (17) in detail, we note that even for real $G_{S,L}$ and $k''_{S,L}$ the signal and pump intensities approach their limit as a function of distance in an oscillatory manner. In particular, for $L_S=L_L$ the oscillatory intensity regime approaches a steady value $8-\sqrt{48}<\sqrt{M}<8+\sqrt{48}$ above the threshold.

For large M and $\mathscr{D}$ size effects, which are due to the derivatives in (12) and (17), need be taken into account only to determine the SRS threshold, since the amplitudes $\mathscr{H}_S$ and $\mathscr{H}_L$ have over most of the illuminated area the steady-state values (18). If, however, the excess of beam intensity above threshold is small, the formation length can become quite large. It is known from the theory of stimulated scattering that the signal intensity becomes comparable with the pump intensity at a spontaneous seed-signal gain $10^{13}\approx e^{30}$ (Ref. 15). It follows from this reference that such a gain is obtained for SRS of SEW over a distance $d=30L_S/(M-1)$.

Thus, slightly above threshold ($M\gtrsim 1$) the formation length d of a Stokes wave is much larger than the path length L_S and can be comparable with or even larger than the transverse dimension $\mathscr{D}$ of the laser spot.

We have dealt so far with SEW amplitudes $\mathscr{H}_L$ and $\mathscr{H}_S$ on the lower boundary of the film. Experimentally, however, one can record only a bulk wave at frequency ω_S and a beam reflected from the film and having an initial frequency ω_L. Their amplitudes h_S and h_L at $z=l$ can be found from expression (7), which remains valid notwithstanding the nonlinear polarization in the lower medium. Since no Stokes-frequency bulk wave is incident on the film ($H_S=0$), we obtain from (7) the following relation between the amplitudes:

$$h_S=s_S\mathscr{H}_S. \qquad (19)$$

The intensity I_S of the bulk signal wave recorded in experiment is then

$$I_S=\varepsilon_3^{-1/2}|h_S|^2=\varepsilon_3^{-1/2}|s_S|^2|\mathscr{H}_S|^2.$$

5. EXPERIMENT

The experimental setup is identical with that described in Ref. 10 (see the inset of Fig. 2). A surface wave of initial frequency ω_L was excited on the interface between benzene and a silver film. Benzene was chosen as the nonlinear medium since it has a high nonlinear Raman susceptibility χ and at the same time does not react chemically with silver. A silver film 450–550 Å thick was evaporated on a diagonal face of a rectangular glass prism ($\varepsilon_3=(1.775)^2$). The SEW were excited by a p-polarized neodymium-laser beam ($\lambda_L=1.06\,\mu$m) incident on the film at an angle θ_L through the glass. The laser beam diameter at the entrance to the prism was several millimeters. The bulk radiation at the wavelength $\lambda_S=1.185\,\mu$m corresponding to the first Stokes component of SRS in benzene was recorded with a photomultiplier. The observation angle θ_S, i.e., the angle between the signal-wave propagation direction and the normal to the surface, was somewhat smaller than θ_L (see the experimental setup in Fig. 2). Besides the signal-beam energy, it was also possible in the experiment to record the energy and duration t_p of each laser pulse. We measured t_p using two-photon luminescence in a rhodamine-6G dye solution.

Experiment has shown that SRS of SEW has a strongly pronounced threshold. Thus, for a fixed laser pulse length t_p, a threshold intensity excess of only 5% increased the signal intensity I_S by more than a factor of 300. The SRS threshold $I_{L\,\text{thr}}$ could be exceeded only using picosecond pulse durations t_p. The short pulse length made possible high intensities $I_L>I_{L\,\text{thr}}$ in the laser beam at an energy density insufficient to damage the silver film. Since the time for the SRS to reach steady state at picosecond pulse lengths becomes comparable with t_p, it is necessary to determine what stage of time evolution the process has reached in the course of the experiment. This was done by measuring the threshold intensity $I_{L\,\text{thr}}$ as a function of the laser pulse length t_p. It can be seen from Fig. 2 that $I_{L\,\text{thr}}$ is decreased by

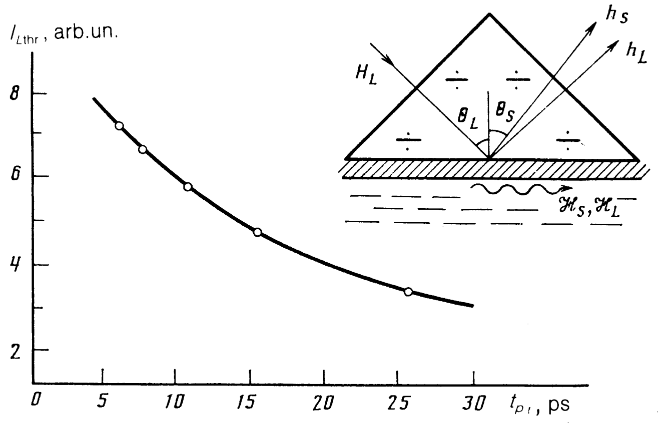

FIG. 2. Dependence of the laser-wave threshold intensity $I_{L\,\text{thr}}$ on the duration of the laser pulse.

one-half when the pulse is shortened from 25 to 7 ps. The SRS process can thus be regarded as stationary only if $t_p \geqslant 25$ ps, for according to Eq. (14) the threshold intensity of a process that evolves to a steady state should be independent of t_p. At pulse lengths 7 ps $< t_p$ 20 ps, which also occur in the experiments, the SRS has no time to reach steady state. For such pulse lengths t_p, however, there is likewise no pronounced nonstationary regime, for otherwise the SS threshold would be proportional to the total energy density in the beam,[11,15] a result not confirmed by experiment.

6. ENERGY DEPENDENCE OF SIGNAL INTENSITY

At values of I_L slightly above threshold, no Stokes-wave field is formed over a distance corresponding to the size of the area illuminated by the laser beam. The signal intensity averaged over the spot increases exponentially with the laser intensity I_L. This can be verified by integrating (15), with allowance for (19), over the laser-spot area. As I_L increases, however, the distance at which the signal is formed decreases; the exponential dependence of I_S on I_L no longer holds, since the field of the Stokes waves corresponds over most of the spot to a stationary regime with respect to the coordinate x [see (18)]. The dependence of I_S on the laser intensity I_L, obtained after transforming (18) with allowance for (19), is:

$$I_S=|s_S|^2\left(\frac{G_S|\tau_L|^2}{\varepsilon_3^{1/2}|G_L|^2}\right)^{1/2}(I_L^{1/2}-I_{L\,\mathrm{thr}}^{1/2}). \qquad (20)$$

At sufficiently large $\mathscr{D} \gg L_S$, the steady state corresponds to values which are not far above the threshold: $I_L - I_{L\,\mathrm{thr}} \ll I_{L\,\mathrm{thr}}$. A linear relation $I_S \propto (I_L - I_{L\,\mathrm{thr}})$ should then be observed according to (20). Thus, for a stationary SRS regime the theory predicts for the intensity I_S a rather complicated energy dependence containing exponential, linear,and square-root components. Figure 3 shows the experimental energy dependence of the signal intensity, obtained for $t_p = 25$ ps corresponding to stationary SRS. Slightly above threshold, as already mentioned in the preceding section, the signal intensity increases rapidly as I_L increases. Unfortunately, the large experimental errors of I_S and I_L, due to fluctuations of the laser pulse length, made it impossible to plot the energy dependence of I_S in this laser-energy interval. Nonetheless, the theoretical results, which predict an exponential dependence of I_S on I_L, can explain the experimental rapid increase of the intensity I_S. With further increase of the laser intensity, a linear energy dependence is observed (see Fig. 3), likewise in agreement with the theoretical results. The behavior of the energy dependence with further increase of the laser intensity I_L at $t_p = 25$ ps could not be investigated for fear of damaging the silver film.

The energy dependence of the signal intensity was obtained also for $t_p = 7$ ps (see Fig. 3). Although the SRS at this pulse length cannot be regarded as stationary, the result does not differ qualitatively from that discussed above. Both a rapid increase of the intensity I_S for a small excess above threshold and a linear stage are observed. At higher intensities I_L it is possible to observe also a deviation from linearity, which can interpreted as a change to a square-root dependence. The experimental energy dependences of the intensity I_S are thus in qualitative agreement with the theoretical results.

7. ANGULAR DEPENDENCE OF THE SRS THRESHOLD

Returning to expression (14) for the threshold intensity of the laser beam we note that $|\tau_L|^2$ depends strongly on

$$k_L=\varepsilon_3^{1/2}(\omega_L/c)\sin\theta_L,$$

hence also on the incidence angle θ_L. As shown by many studies of nonlinear SEW interactions, a connection exists between the angular dependence of $|\tau_L|^2$ and the linear reflection coefficient $R_L = |r_L|^2$. If the SEW are effectively excited, in particular, the maximum of $|\tau_L|^2$ is reached at the incidence angle $\theta_L = \theta_{L0}$ corresponding to the minimum reflection coefficient R_L. Figure 4 shows the angular dependence of the threshold intensity $I_{L\,\mathrm{thr}}$ obtained at a fixed pulse length, and of the reflection coefficient R_L. The plot of R_L was obtained at low laser-beam intensities. It can be seen that the experimental angular dependence of $I_{L\,\mathrm{thr}}$ agrees qualitatively with the theoretical expression (14). Indeed, $I_{L\,\mathrm{thr}}$ has a minimum at $\theta_L = \theta_{L0}$ and increases steeply when the incidence angle θ_L deviates from the optimal θ_{L0}. Note that the angular dependence of the reflection coefficient R_L can yield the dielectric constant $\varepsilon_2(\omega_L)$ of the silver and the film thickness l. The results can be used to obtain the theoretical angular dependence of $|\tau_L|^2$ and thereby compare quantitatively the experimental and theoretical results.[7,8] In the investigation of SRS, however, many factors hinder this comparison, and we shall discuss it presently in greater detail.

It can be seen from Eq. (13) that G_S depends both on the length and on the direction of the wave vector $\mathbf{k}_S$, and this should be suitably manifested in the angular dependence of the signal intensity I_S. Note that from among the factors in the right-hand side of expression (13) for G_S the quantity most strongly dependent on the length of the wave vector $\mathbf{k}_S$ is τ_S. The maximum G_S corresponds then to a wave-vector length

$$k_{S0}=\varepsilon_3^{1/2}(\omega_S/c)\sin\theta_{S0}.$$

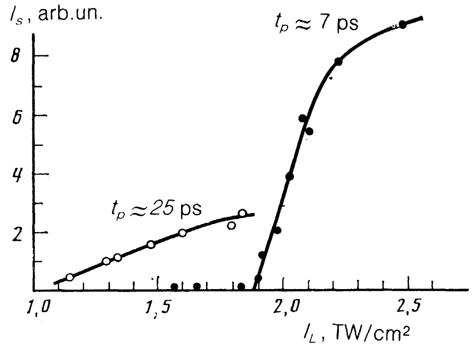

FIG. 3. Energy dependence of the signal-wave intensity I_S for different pulse lengths.

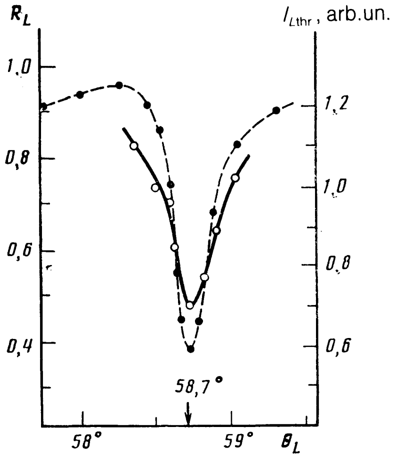

FIG. 4. Dependence of the threshold intensity $I_{L\,thr}$ of a laser wave (solid curve) and of the linear reflection coefficient R_L (dashed) on the angle of incidence of the laser wave on the plasma.

The angle θ_{S0} can in turn be determined from the experimental angular dependence of the linear reflection coefficient $R_S(\theta_S)$ shown in Fig. 5. This dependence was obtained by using the first Stokes component of the bulk SRS in benzene, which has the same frequency ω_S as the investigated signal. Figure 5 shows a plot of the signal intensity I_S versus the observation angle θ_S for fixed I_L and t_p and for a constant incidence angle $\theta_L = \theta_{L0}$ of the laser beam on the film. It can be seen that I_S is a maximum at the angle θ_{S0} corresponding to the maximum G_S. Note that θ_{S0} is somewhat smaller than θ_{L0}, owing to the dispersion of the silver. In agreement with (13), the signal intensity I_S decreases sharply when the observation angle θ_S deviates from the optimal $_{S0}$.

According to (13), G_S depends also on the angle θ between the vectors $\mathbf{k}_L$ and $\mathbf{k}_S$. Figure 6 shows the experimental plots of I_S versus φ at fixed I_L, t_p, and $\theta_L = \theta_{L0}$. They are, in agreement with (13), quite smooth, and the maximum of the intensity I_S corresponds to $\varphi = 0$. Note that the signal-intensity angular dependences of Figs. 5 and 6 were obtained experimentally by using a slit diaphragm, and agree thus qualitatively with the theory.

An additional experiment was devoted to the tendency of the SRS to build up as a function of x. We have considered up to now a situation corresponding to large transverse dimensions of the laser spot. Let us examine illumination of the film by a narrow strip with length much larger than width, in a direction making an angle φ_0 with the vector $\mathbf{k}_L$. Slightly above threshold, the gain length is insufficient for the intensity of a Stokes wave propagating across the strip to become high enough [see (15)]. The signal intensity will be a maximum when the vector $\mathbf{k}_S$ is parallel to the strip, i.e., when $\varphi = \varphi_0$, even though $G_s(\varphi_0) < G_s(0)$. In fact, experiment has shown that the signal beam direction follows up the strip orientation. The illuminated region was formed in this case by focusing the laser beam on the film with a cylindrical lens. Note that this last experiment corroborates the decisive influence of size effects on SRS.

Thus, most experimental data agree qualitatively well with the theoretical results. A quantitative comparison of theory with experiment is hindered by the following circumstances. The SRS threshold can be exceeded at laser energies 1.5–2 times lower than the film damage threshold. Heating the film alters its properties, especially the dielectric constant of the silver. The path length of the SEW is therefore decreased at both the initial and the Stokes frequencies, and the angular dependence of $|\tau_{S,L}|^2$ and $R_{S,L}$ is distorted. This leads in turn to distortion of the angular dependence in Figs. 4 and 5. In particular, when I_L is increased the plot of I_S versus the angle in Fig. 5 is broadened. The changes in the dielectric constant of silver, which depend on the laser intensity I_L, thus make quantitative comparison of the experimental data with the theory difficult. The main factor preventing a quantitative comparison of the theoretical and experimental results, however, is the stimulated temperature scattering which, as we shall show below, accompanies the SRS.

Investigation of the dependence of the reflection coefficient R_L on the intensity I_L in a laser beam at fixed θ_L has revealed threshold-dependent changes of R_L, with the thresholds of these changes lower than the SRS threshold. We emphasize that this behavior of the reflection coefficient for nanosecond pulses is due to stimulated temperature scattering (STS).[11] A check on the possibility of this scattering

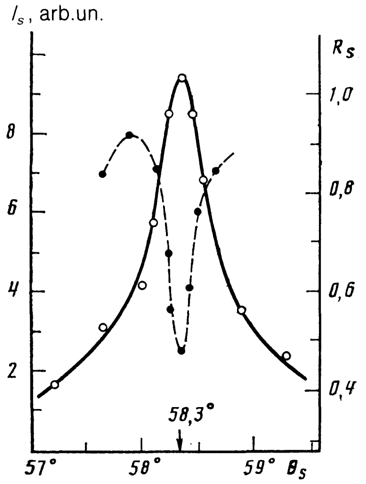

FIG. 5. Angular dependences of the signal-wave intensity I_S (solid curve) and of the linear reflection coefficient R_S (dashed).

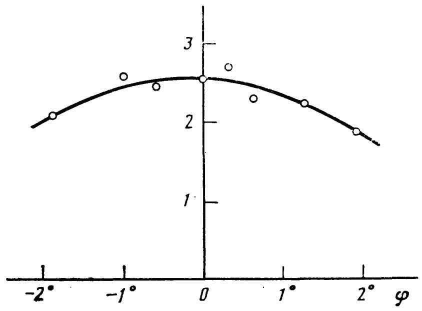

FIG. 6. Dependence of the signal-wave intensity I_S on the angle φ between the vectors $\mathbf{k}_L$ and $\mathbf{k}_S$.

for picosecond pulses required the following additional studies. We obtained the energy dependence of the reflection coefficient R_L following SEW excitation on the interface between silver and glycerine. Owing to the low nonlinear susceptibility of glycerine, the SRS threshold was in this case appreciably higher than the pump intensity, so that no SRS developed. Threshold-dependent changes of the reflection coefficient, however, were observed also in this case, attesting to the presence of STS under these experimental conditions. When the laser beam damages the silver film side exposed to air, just as in the case of nanosecond pulses,[9,11] formation of surface periodic structures is observed; this also attests to the presence of STS at picosecond pulse durations. Stimulated temperatures scattering thus accompanies the stimulated Raman scattering of surface electromagnetic waves, but makes quantitative comparison of experiment with theory very difficult. In addition, the STS-induced threshold-dependent changes of the reflection coefficient R_L make it impossible to obtain the additional experimental data needed to record the changes produced in R_L by pump depletion in STS of SEW [see (7)].

8. CONCLUSION

The need for taking size effects into account in investigations of stimulated Raman scattering of surface electromagnetic waves causes SRS to differ radically from zero-threshold nonlinear interactions in which SEW participate. Its indisputable similarity to bulk stimulated scattering, namely an exponential growth of the signal intensity and formation at a greater distance, distinguish this SRS also from stimulated scattering of bulk waves. These peculiarities are reflected, in particular, in the set of equations (12) and (17). The right-hand side of (17), which corresponds to pumping of SEW at the initial frequency by an external source, is missing from the analogous set of equations describing bulk STS. It is also mandatory, in investigations of SRS, to take into account the SEW absorption represented by the unity terms in the left-hand sides of (12) and (17). Expression (20), in particular, corresponds to a square-root dependence of the signal intensity I_S on the pump intensity I_L far above threshold. We emphasize that in the case of stimulated scattering of bulk waves far above threshold, I_S depends linearly on I_L. It is the peculiarities of the investigated effect which distinguish it both from zero-threshold nonlinear interactions with SEW participation and from stimulated scattering of bulk radiation, which make the study of stimulated Raman scattering of electromagnetic waves interesting.

The need for taking pump depletion into account in the case of stationary SS with SEW participation has stimulated the rather detailed investigations of this effect in the present paper. This makes our results useful for the study of other nonlinear effects in which SEW participate. Foremost is stimulated temperature scattering, since the pump-depletion effect, as mentioned in Ref. 11, can also influence strongly nonstationary stimulated scattering. In experiments on zero-threshold nonlinear interactions with SEW participation, the signal intensity is as a rule much less than the laser intensity,[1-6,8] so that pump depletion can be neglected. But in degenerate four-wave mixing, for example, a diffraction efficiency exceeding one percent was achieved in experiment.[7] At these and higher values of the diffraction efficiencies, effects similar to pump depletion can influence strongly also the zero-threshold nonlinear interactions with SEW participation.

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