Experimental observation of stimulated Raman scattering of surface electromagnetic waves

A. N. Sudarkin and K. N. Ushakov
Institute of Problems of Mechanics, Academy of Sciences of the USSR

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A stimulated Raman scattering of surface electromagnetic waves has been seen experimentally for the first time. The experimental results are compared with the theory at a qualitative level. It is shown that the stimulated Raman scattering of surface electromagnetic waves is substantially different from other nonlinear interactions involving surface waves.

Several nonlinear optical effects involving surface electromagnetic waves have recently been studied experimentally.[1,2] It is important to note that these effects do not involve thresholds, and the only factors limiting their observation are the sensitivity of the measurement system and the presence of a linear noise. The observation of nonlinear effects which do involve a threshold, such as stimulated scattering effects, on the other hand, is complicated for the following reasons. Since the mean free path (L_S) of the signal surface electromagnetic wave is short, an intense pump laser beam is required in order to exceed the threshold for the stimulated scattering. Meeting that condition, however, is hindered by the low thresholds for damage to the surface of the metal. On the other hand, stimulated scattering effects have several properties which sharply distinguish them from other nonlinear interactions involving surface electromagnetic waves. Stimulated thermal scattering is the only type of stimulated scattering of surface waves which has been studied to date.[3,4] In this letter we report the observation and study of a stimulated Raman scattering of surface electromagnetic waves.

In the experiment, a surface electromagnetic wave at the frequency ω_S is excited in a nonlinear fashion at an interface between a silver film and benzene. A surface electromagnetic wave at the laser frequency ω_L is excited by bulk radiation by the method of frustrated total internal reflection.[1,2] The p-polarized beam from a single-mode (in terms of transverse indices) neodymium laser ($\lambda_L = 1.06\,\mu$m) is incident at an angle θ_L on the other boundary of the film, from the side of a rectangular glass ($n = 1.775$) prism (Fig. 1). The signal surface electromagnetic wave is partially radiated into the bulk wave ($\lambda_S = 1.185\,\mu$m), which is detected in the experiment (Fig. 1). The observation of stimulated Raman scattering is facilitated by the brevity of the relaxation to a steady state, which takes only a few tens of picoseconds. At a pulse length of about 10 ps, it thus becomes possible to exceed the threshold for a stimulated Raman scattering at an energy density which does not damage the film.

In the absence of a nonlinearity, the amplitude of the magnetic field at the silver-benzene interface, $\mathscr{H}_L$, is proportional to the amplitude of the wave incident on the film, $H_L :: \mathscr{H}_L = \tau_L H_L$, where τ_L depends on the dielectric constants of the media at the frequency ω_L and the projection of the wave vector onto the surface, $k_L = (\omega/c) n \sin\theta_L$, and reaches a maximum in absolute value at $k_L = \mathscr{K}_L$, where $\mathscr{K}_L$ is the length of the wave vector of the surface electromagnetic wave. A theoretical analysis of

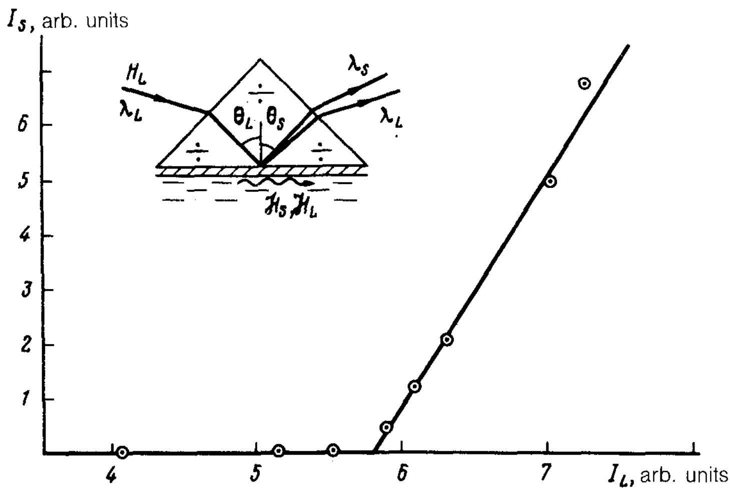

FIG. 1.

the effect shows that in a steady state in terms of the time the field $\mathscr{H}_L$ and the amplitude of the signal wave at the silver-benzene interface, $\mathscr{H}_S$, obey the equations

$$\frac{1}{k''_S}\frac{d\mathcal{H}_S}{dx} + (1 - G_S|\mathcal{H}_L|^2)\mathcal{H}_S = 0 \tag{1}$$

$$\frac{1}{k''_L}\frac{d\mathcal{H}_L}{dx} + (1 + G_L|\mathcal{H}_S|^2)\mathcal{H}_L = \tau_L H_L \ , \tag{2}$$

where $k''_{S,L} = 1/2L_{S,L}$, and $G_S(\omega_S, \omega_L, k_L, k_S)$ is a positive real quantity which is proportional to the nonlinear susceptibility of benzene (the vectors $\mathbf{k}_L$ and $\mathbf{k}_S$ run parallel to the x axis). Analysis of (1) and (2) shows that in describing the stimulated Raman scattering we need to consider the transport of electromagnetic energy along the surface, which is essentially not manifested in other nonlinear interactions. In the latter case, the signal is established over a distance L_S, which does not exceed a few tens of microns (usually much shorter than the size of the laser spot). We can thus assume that the signal amplitude and the interaction efficiency are constant over the spot. Furthermore, one usually ignores the inverse effect of the signal on the amplitude of the pump wave, under the assumption that the relation $\mathscr{H}_L = \tau_L H_L$ holds as before.

Stimulated Raman scattering has some different properties. Let us consider a laser spot which is confined spatially to $0 < x < D$. We assume that a spontaneous seed with an amplitude $\mathscr{H}_{S0}$ is applied to the "entrance" of the medium at $x = 0$. The seed will grow exponentially along the coordinate x if the laser beam intensity $I_L = 1/n|H_L|^2$, exceeds the threshold $I_{L0} = (nG_S|\tau_L|^2)^{-1}$. If the extent to which the threshold is exceeded is only small, the establishment length may be hundreds of microns and may be comparable to the spot size D. In this case the efficiency of the stimulated Raman scattering will depend on D. If the threshold is exceeded by an ample margin, and D is large, the establishment of a signal (which cannot grow without bound) nevertheless occurs, but it is a consequence of a decrease in $|\mathscr{H}_L|^2$. This effect is simllar to the depletion of pump waves during the stimulated scattering of bulk waves. By solving Eqs. (1) and (2) jointly, one can find the steady-state values of the amplitudes:

$$\mathcal{H}_S^y = \left(\frac{\tau_L H_L G_S^{1/2} - 1}{G_L}\right)^{1/2} e^{i\psi} \ ; \qquad \mathcal{H}_L^y = G_S^{-1/2} \ (\neq \tau_L H_L).$$

The experimental results show that the stimulated Raman scattering of surface electromagnetic waves does indeed clearly involve a threshold. When the threshold intensity is exceeded by a few percent, the signal increases by three orders of magnitude. Figure 1 shows the experimental results on the signal intensity I_S as a function of I_L at a fixed pulse length. We see that I_S is proportional to $(I_{L0} - I_L)$, in accordance with the theory. Figure 2 shows the angular dependence of the threshold I_{L0} obtained at a fixed pulse length (the solid line). Shown for comparison in this figure is the angular dependence of the linear reflection coefficient R of the film for the laser beam

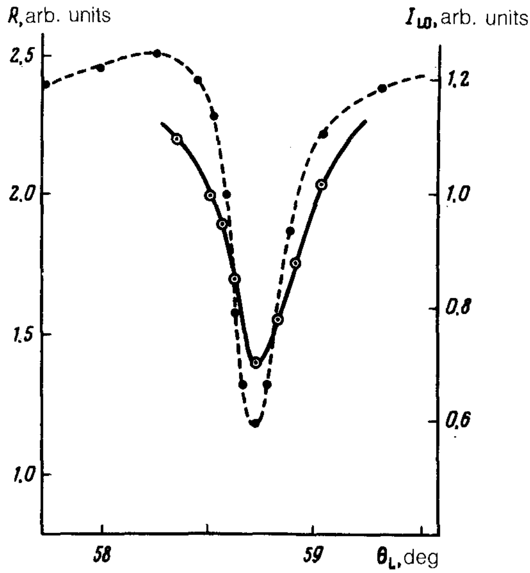

FIG. 2.

at small values of I_L (the dashed line). Near the optimum angle, the theory predicts a proportionality $I_{L0} \sim (1/|\tau_L|^2) \sim R$. The curves in Fig. 2 are somewhat different in shape, apparently because of an increase in the imaginary part of the dielectric constant of the silver during the heating of the film and the weak angular dependence of G_S which is present.

In the case of stimulated Raman scattering, in contrast with other nonlinear interactions, neither the length nor the direction of the wave vector $\mathbf{k}_S$ is imposed by the pump waves; each is selected by the system spontaneously. The value of G_S reaches a maximum at $k_S = \mathscr{K}_S$ and $\varphi = 0$, where φ is the angle between the vectors $\mathbf{k}_S$ and $\mathbf{k}_L$. The experiments show that at large beam sizes the signal does indeed propagate in the plane of incidence ($\varphi = 0$), and the angle between its propagation direction and the normal to the surface, θ_S (Fig. 1), satisfies the expression $\theta_S = \arcsin c/\omega n \mathscr{K}_S$. If the laser spot has the shape of a long, narrow stripe, which makes an angle φ_0 with $\mathbf{k}_L$, the angle φ between $\mathbf{k}_S$ and $\mathbf{k}_L$ is the same: $\varphi = \varphi_0$. This equality results from the circumstance that the process builds up over space. Despite the slightly smaller value of G_S, the signal wave can undergo an unhindered spatial growth in this direction.

Plans call for a more detailed study of the observed effect. It may be possible to find the energy dependence of I_S near the threshold, to identify the role played by the anti-Stokes components of the stimulated Raman scattering, and to study how the laser pulse length τ_p influences the effect. At our pulse lengths, we observe a slight dependence of the threshold on τ_p, so the process cannot be judged a completely steady-state process in time. We also hope to make a quantitative comparison of the experimental and theoretical results.

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