Stimulated thermal scattering into a surface electromagnetic wave (experimental)

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Stimulated thermal scattering into a surface electromagnetic wave has been studied experimentally. The results show that the scattering is by a grating of the dielectric constant of silver.

Several nonlinear optical effects involving surface electromagnetic waves (SEWs) have now been studied experimentally.[1–3] However, the strong absorption of the SEWs means that the surface will suffer damage at comparatively low SEW intensities. The effect is to hinder an experimental study of nonlinear optical effects, in particular, certain types of stimulated scattering which occur after a threshold is reached. It has been shown that in addition to the well-studied stimulated thermal scattering (due to absorption; STS-A) from a bulk wave into a bulk wave,[4,5] there can be a stimulated thermal scattering of a bulk wave into a SEW.[6] A study has been made of scattering by capillary waves excited at the surface of a liquid metal or semiconductor. In the present experiments we use a metal film a few hundred angstroms thick, so that as the film melts we obtain a liquid layer of sufficient depth, and another mechanism is responsible for the effect.

Single-mode light from a neodymium laser with a pulse length of 50 ns at half-maximum, polarized in the plane of incidence, is incident from air on a silver film deposited on a diagonal face of a rectangular glass prism. The angle of incidence, θ, is varied in the course of the experiment. In most cases, a new part of the film is illuminated in each laser flash. If a SEW is excited at the silver-air interface, it is partially reradiated into the interior of the glass. As it passes through a lateral face of the prism, this bulk wave is incident on a photodetector, where it can be detected. The appearance of a signal at the photodetector is therefore evidence of the excitation of SEWs at the boundary of the silver. To insure that a nonlinear scattering is occurring, we find the energy of this bulk wave, E_S, for various energies of the initial laser beam, E_L, and various angles of incidence θ. Figure 1 shows the ratio E_S/E_L (after subtraction of a linear noise) versus E_L for $\theta = 55°$ (triangles) and $\theta = 68°$ (circles). The unit along the abscissa corresponds to the threshold energy E_{thr} for laser damage to the film at the given angle of incidence. By "damage threshold" here we mean the minimum value of the energy in the beam which leaves damage on the film that is visible under a microscope. The detected beam is directional; it is propagating along the ray which lies in the plane of incident in the initial beam and makes an angle ψ with the normal to the surface. Since the value ψ agrees with the known dispersion relation for SEWs,[1] i.e., since the relation $n \sin \psi \cong [\epsilon_1/(\epsilon_1 + 1)]^{1/2}$ holds, we can conclude that the source of the detected signal is an SEW. Here n is the refractive index of the glass, and $\epsilon_1 = \operatorname{Re} \epsilon$ is the real part of the dielectric constant of silver at the frequency of the laser

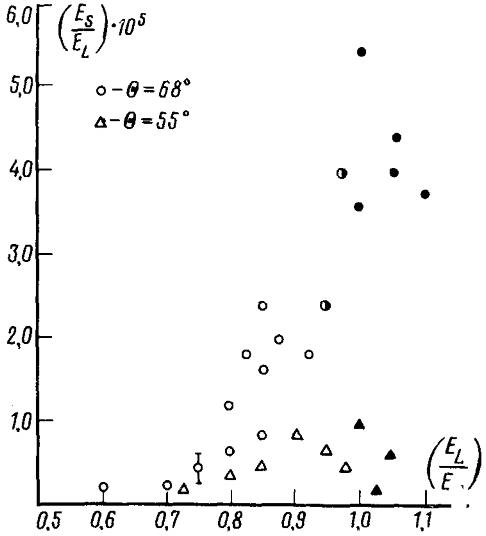

FIG. 1.

light. To calculate ϵ and the film thickness d, we work from the angular dependence of the reflection coefficient found during incidence of the laser beam from the side of the glass. The shape of this dependence depends strongly on the quantities which we wish to determine.[1–3]

In the course of the experiment it was found that the efficiency of the nonlinear scattering depends strongly on the angle of incidence, reaching a maximum at $\theta = 68$–$70°$. This scattering could not be observed outside the region $50° < \theta < 75°$. It can be seen that at $\theta = 68°$ the detected signal increases markedly with the occurrence of damage (the dark symbols in Fig. 1), exceeding the linear noise by a factor of several units. In the absence of damage, we checked the grating responsible for the appearance of the directional nonlinear scattering for reversibility. For this purpose, the energy of the laser beam was attenuated to a value which would definitely not cause a nonlinear scattering, and we exposed the same part of the film to another laser flash. As a rule, the signal energy decreased to the level of the linear noise in this case, providing evidence that the grating is reversible. At energies $E_L \geqslant 0.95 E_{\text{thr}}$, however, we do not find the same result (the half-filled symbols in Fig. 1). We thus have evidence that changes in the film which are invisible under a microscope are irreversible.

Now that we are convinced that we are dealing with a nonlinear scattering, we wish to examine its nature. We know[2] that the derivative of the dielectric constant of silver with respect to the temperature is quite high, and the observed scattering could be interpreted as a stimulated thermal scattering by a dielectric-constant grating, according to calculations. We would then have a complete explanation for the reversibility of the grating responsible for the effect. The changes caused in the dielectric constant of the surface layers of the metal by other mechanisms are negligible. For thermal gratings with a large period Λ, the rise time is far longer than the pulse length, so that such gratings do not occur.[4,5] On the other hand, for gratings with a small period, for which the process may be regarded as in a steady state, a further decrease in Λ leads to a decrease in the efficiency of the stimulated thermal scattering.[4,5] Since we were able to continuously vary the angle of incidence and thus the period of the thermal grating, we establish that there exists an optimum angle θ_{opt} corresponding to the highest scattering efficiency. To check whether $\theta_{opt} \cong 70°$ corresponded quantitatively to the thermal and optical constants of the film, we carried out a theoretical study of the effect.

At energies $E_L > E_{thr}$ the damage occurs first at the maxima of the thermal grating, and normal periodic structures should remain at the surface of the film.[6,7] This conclusion is supported by the experiments. Figure 2 shows photomicrographs of a film for $\theta = 68°$ for cases in which E_L is just slightly (Fig. 2a) and well (Fig. 2b) above the damage threshold. Like the nonlinear scattering, the surface structures are observed at $50° < \theta < 75°$. Their period Λ corresponds to the scattering of a laser beam into an SEW; i.e., the relation $\Lambda^{-1} = \lambda^{-1}\{[\epsilon_1/(\epsilon_1 + 1)]^{1/2} - \sin\theta\}$, where $\lambda = 1.06$ μm holds very accurately. After the appearance of the surface structures, the scattering is a scattering by a damage grating which has a high diffraction efficiency.[8] As a result, there is a further increase in the signal energy (Fig. 1). From Fig. 2 we can extract further information on the stimulated thermal scattering, since the thermal grating is the nucleating region for the observed surface structure. We see that the stimulated thermal scattering does not give rise to a common SEW with a phase which is constant over the entire spot. Because of the energy transport along the wave vector

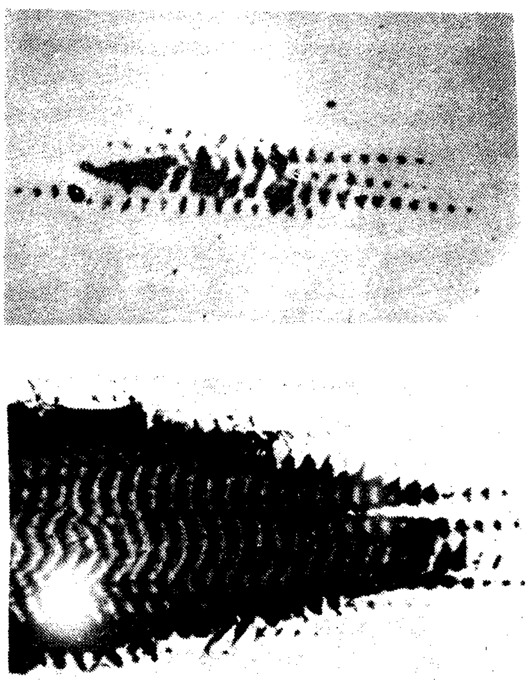

FIG. 2.

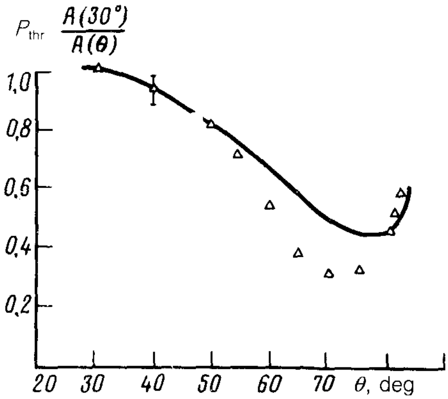

FIG. 3.

of the SEW (from left to right in Fig. 2), its phase is correlated in this direction. In the perpendicular direction, the phase of the SEW relaxes over a distance of a few tens of microns. The result is the formation of several SEWs, which propagate at small angles with respect to each other. The effect is to slightly increase the divergence of the bulk wave that is detected.

Only a small fraction of the SEW energy is reradiated into the interior of the glass, since most of the energy is absorbed. Because of the additional absorption, the parts of the film where SEWs are excited through stimulated thermal scattering may suffer damage earlier (Fig. 2). Figure 3 shows, in relative units, the experimental angular dependence of the threshold energy density $P_{thr} = E_{thr} \cdot \cos\theta$ (triangles). In addition, we calculated the angular dependence of the fraction of the energy absorbed by the film, $A(\theta)$, assuming that the energy of the SEW is small. Since the damage is determined by specifically the absorbed energy density, we would have a constant product $A(\theta)P_{thr} = \text{const}$ in the absence of nonlinear scattering. The solid line in Fig. 3 shows the angular dependence $A(30)/A(\theta)$, which coincides with the curve for P_{thr} at $\theta = 30°$. The substantial discrepancy between the curves at $50° < \theta < 75°$ indicates that an additional absorption due to a nonlinear scattering into SEWs is having a significant effect on the threshold for damage to the film. The error that might occur in the determination of ϵ and d has little effect on the shape of the $A(30)/A(\theta)$ curve, despite the fact that the absolute value of A may change. We thus have confirmation of our last conclusion. In summary, we have carried out an experimental study of a stimulated thermal scattering into surface electromagnetic waves at a dielectric-constant grating.

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