STIMULATED SCATTERING OF LIGHT FROM THE SURFACE OF A HIGHLY VISCOUS LIQUID

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Stimulated light scattering from the surface of a highly viscous liquid is analyzed theoretically. Stimulated light scattering in this case differs greatly from all other known types of stimulated scattering in that the Stokes and anti-Stokes frequency shifts of the scattered radiation depend on the intensity of the incident light. The instability threshold of capillary waves on the surface of a liquid is calculated.

STIMULATED scattering (SS) of light on the surface of a liquid was predicted theoretically in[1]. In the present work we have calculated the threshold intensity I_0 of the scattered light. This threshold determines the onset of capillary wave instability on the surface of a highly viscous liquid for which

$$2\nu q^2 \ll \Omega_0. \tag{1}$$

Here $\nu = \eta/\rho$ is the kinematic viscosity, ρ is the density, and $\mathbf{q}$ is the wave vector of a capillary wave. The frequency Ω_0 is determined from the dispersion equation for capillary waves:

$$\Omega_0 = (\alpha q^3/\rho)^{1/2} \tag{2}$$

where a is the coefficient of surface tension.

It is characteristic of SS on the surface of a low-viscosity liquid[1] that the frequencies of the capillary waves on which SS occurs (and therefore the frequencies $\omega_0 \pm \Omega_0$ of the Stokes and anti-Stokes scattered components, where ω_0 is the incident light frequency) do not depend on the incident light intensity I up to a threshold I_0 but are determined from Eq. (2). The intensity I governs only the logarithmic decrement γ of the capillary wave:

$$\zeta \sim \exp[i(\mathbf{qr} - \Omega t)] = \exp(-\gamma t)\exp[i(\mathbf{qr} \pm \Omega_0 t)].$$

In[1] the complex frequency Ω is given by

$$\Omega = \pm\Omega_0 - i2\nu q^2[1 \mp BI/(2\Omega_0\rho\nu q^2)], \tag{3}$$

where $B = B(\mathbf{k}_0, \mathbf{q}, \Psi, \epsilon)$ is a certain function of only the incident light wave vector $\mathbf{k}_0$, the capillary wave vector $\mathbf{q}$, the incident light polarization $\psi (\cos\psi = E_y/|\mathbf{E}|)$, and the dielectric constant ϵ of the liquid.[1)] The threshold value

$$I_0 = 2\eta q^2\Omega_0/|B| \tag{4}$$

corresponds to the condition $\gamma = \mathrm{Im}\,\Omega = 0$.

In the present work we consider SS on the surface of a highly viscous liquid when

$$(\Omega_0/2\nu q^2)^2 < 0{,}145. \tag{5}$$

Subject to (5) and without including the ponderomotive action of the radiation, the capillary wave frequency Ω is purely imaginary $(-i\Omega_0^2/2\nu q^2)$; this corresponds to exponentially damped motion of the liquid surface without time-dependent oscillations.[2,3] The spectrum of light scattered on the thermal fluctuations of this liquid surface contains only an unshifted component.[3]

The ponderomotive action of the field of an intense light wave on the surface of a highly viscous liquid can be taken into account in exactly the same way as this was done in[1] for the case of a low-viscosity liquid. Thus for arbitrary viscosity of the liquid and arbitrary polarization of the incident light we obtain the following characteristic equation determining the complex capillary wave frequency Ω:

$$\Omega_0^2 + (2\nu q^2 - i\Omega)^2 + i\frac{2B}{\rho}I = (2\nu q^2)^2\sqrt{1 - i\frac{\Omega}{\nu q^2}} \tag{6}$$

This equation differs from the usual characteristic equation for capillary waves in a viscous liquid[2] only by the presence of the last term on the left-hand side; this term includes the light intensity. Here $B(\mathbf{k}_0, \mathbf{q}, \psi, \epsilon)$ is the same function as in (3). In the case of an extremely viscous liquid, which we shall be considering henceforth, we have $2\nu q^2 \gg \Omega_0$, and the solution of (6) is[2)]

$$\Omega = \frac{BI}{\rho\nu q^2} - i\frac{\Omega_0^2}{2\nu q^2}\left[1 - \frac{3}{2\Omega_0^2}\left(\frac{BI}{\rho\nu q^2}\right)^2\right]. \tag{7}$$

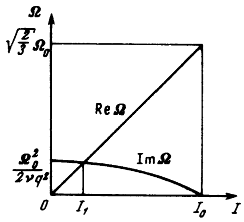
The accompanying figure shows the real and imaginary parts (Re Ω and Im Ω) of the frequency belonging to the capillary wave on which SS takes place, as functions of the light intensity I. The threshold intensity I_0, determined from the condition Im $\Omega = 0$, is given by

$$I_0 = \sqrt{2/3}\,\eta q^2\Omega_0/|B|. \tag{8}$$

The frequency of the excited capillary wave is $BI_0/\rho\nu q^2 = \sqrt{2/3}\,\Omega_0$. It is of interest to compare the thresholds for low- and high-viscosity liquids under identical conditions of excitation, i.e., identical values of $\mathbf{k}_0$, $\mathbf{q}$, and ψ. If we assume here an identical dielectric constant ϵ for both types of liquids, then the values of $B(\mathbf{k}_0, \mathbf{q}, \psi, \epsilon)$ will also coincide. The ratio

1) For the special case, considered in [1], of polarization perpendicular to the plane of incidence, we have $B = (8\pi/c)q^2 D$, where D is determined from (5) of [1].

2) The second root of (6) corresponds to extremely stronger damping of the wave and will therefore be disregarded.



of the thresholds obtained from (4) and (8) [with the indices (1) and (2) designating the low- and high-viscosity liquid, respectively] will then be

$$\frac{I_0^{(2)}}{I_0^{(1)}} = \frac{1}{\sqrt{6}} \frac{\eta_2}{\eta_1} \left(\frac{\rho_1 \alpha_2}{\rho_2 \alpha_1} \right)^{1/2}. \quad (9)$$

The SS effect on the surface of a highly viscous liquid is basically different from all other known forms of stimulated light scattering in that the frequency Re Ω of the excited capillary wave does not depend on the wave vector **q** of this wave (as in SS on the surface of a low-viscosity liquid, or similarly in the case of Mandel'shtam-Brillouin SS). Equation (7) shows that the given frequency depends on the intensity I of the incident light.

In a given direction of observation (i.e., for given **q**) the following qualitative picture of the scattering appears. At low intensities I the scattered light spectrum contains, as already mentioned, only the unshifted line of width $\Omega_0^2/2\nu q^2$. With increasing I we have the corresponding "running" capillary wave because of a real addition to the frequency, Re $\Omega = BI/\rho\nu q^2$. Then either a Stokes component ($\omega_0 - |\mathrm{Re}\,\Omega|$) or an anti-Stokes component ($\omega_0 + |\mathrm{Re}\,\Omega|$) appears depending on the direction of scattering (the sign of B). At the light intensity $I_1 = \rho\Omega_0^2/2|B|$ the scattered line is shifted from ω_0 by a distance equal to its (previously given) width $\Omega_0^2/2\nu q^2$. With further increase of I this shift grows, but the width of the line decreases until it vanishes at $I = I_0$, which corresponds to the onset of capillary wave instability. For $I > I_0$ a special quantitative analysis of SS is needed.

We conclude with a numerical estimate of the threshold I_0. When the incident light wave is polarized in the incident plane at the incident angle $\theta = 80°$, with $q \approx 10^3/\mathrm{cm}^{-1}$, $\Omega_0 \approx 10^5\ \mathrm{sec}^{-1}$, $k_0 \approx 10^5\ \mathrm{cm}^{-1}$, and $\eta \approx 10$ poise, we have $I_0 \approx 6 \times 10^8\ \mathrm{W/cm}^2$.

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[1] F. V. Bunkin, A. A. Samokhin, and M. V. Fedorov, ZhETF Pis. Red. 7, 431 (1968) [JETP Lett. 7, 337 (1968)].

[2] V. G. Levich, Fiziko-khimicheskaya gidrodinamika (Physico-chemical Hydrodynamics), Fizmatgiz, 1959.

[3] R. H. Katyl and U. Ingard, Phys. Rev. Lett. 19, 64 (1967).

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