# THEORY OF STIMULATED SCATTERING OF LIGHT BY A LIQUID SURFACE

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The scattering of an intense electromagnetic wave by the fluctuations of a free liquid surface is considered. In contrast with the ordinary theory of spontaneous scattering of light, the nonlinear field action at the interface due to ponderomotive forces is taken into account. At sufficiently high intensities this action causes an appreciable change in the spectrum of thermal capillary waves that determine the scattering of light at the surface of a liquid. An expression for the spectral intensity of the scattered light is obtained for any direction of scattering. Numerical results are given.

1. Light passing through the interface of two media is scattered by the fluctuating deviations of the interface from the equilibrium shape. The spontaneous scattering of light by the liquid surface has long been known[1,2] although the spectral intensity of the scattered light was measured only recently[3,4]. The main contribution to this scattering comes from thermal capillary waves which determine the fluctuations of the free surface of an incompressible liquid. The reverse effect of the electromagnetic field on the interface does not exist in spontaneous scattering of light. However, at sufficiently high intensities of incident radiation the nonlinear field effect becomes significant. This effect was considered previously by the authors[5] for the case of capillary waves that determine the scattering of light in the plane of incidence of the initial monochromatic radiation.

In the present work we obtained the spectrum of scattered light for all directions taking into account its dependence on the intensity of the incident radiation. In Sec. 2 the solution of the electrodynamic boundary problem for a given shape of the liquid surface is used to find the relation between the field intensity of the scattered wave and the deviation of the liquid surface from plane. The obtained formulas are used in Sec. 3 to determine the nonlinear effect of the field at the interface responsible for changing the natural frequencies of the capillary waves. In Sec. 4 an expression is obtained for the spectral intensity of the scattered light in terms of correlation factors of the displacement of the liquid surface, computed on the basis of nonequilibrium thermodynamics.

2. Let a plane monochromatic wave with amplitude $\mathbf{E}_0$, frequency $\omega_0$, and wave vector $\mathbf{k}^{(0)}$ fall from free space ($\epsilon_0 = \mu_0 = 1$) on a surface of a liquid with dielectric permittivity $\epsilon > 1$ and magnetic permeability $\mu = 1$. In the absence of fluctuations the liquid surface is the

$$\mathbf{E}' = \mathrm{Re}\,\{\mathbf{E}_0 \exp[i(\mathbf{k}^{(0)}\mathbf{R} - \omega_0 t)] + \mathbf{E}_1 \exp[i(\mathbf{k}_1\mathbf{R} - \omega_0 t)]\} + \mathbf{E}^{(1)}(\mathbf{R}, t),$$

plane z = 0 deviations from which $\zeta(\mathbf{r}, t)$ are considered small: $|\zeta k^{(0)}| \ll 1$. The boundary problem is postulated at the interface $z = \zeta$ without allowing for relativistic corrections and its solution is sought in the form

$$\mathbf{E} = \mathrm{Re}\,\{\mathbf{E}_2 \exp[i(\mathbf{k}_2\mathbf{R} - \omega_0 t)]\} + \mathbf{E}^{(2)}(\mathbf{R}, t),$$

$$\mathbf{R} = \{x, y, z\}, \quad \mathbf{k}^{(0)} = \{k_{0x}, 0, -k_z\}, \quad \mathbf{k}_1 = \{k_{0x}, 0, k_z\},$$

$$\mathbf{k}_2 = \{k_{0x}, 0, -k_z'\}, \; k_z = \left(\frac{\omega_0^2}{c^2} - k_{0x}^2\right)^{1/2}, \tag{1}$$

where $\mathbf{E}'$ and $\mathbf{E}$ are electric field intensities in free space and in liquid respectively. The amplitudes of the reflected $\mathbf{E}_1$ and transmitted $\mathbf{E}_2$ fields are determined by Fresnel formulas for a plane interface z = 0, while the fields $\mathbf{E}^{(1)}$ and $\mathbf{E}^{(2)}$ are due to the fluctuations and in the present approximation are linear in $\zeta$.

If the initial wave polarization is perpendicular to the plane of incidence (x, z), the continuity of the tangential component of the electric field intensity implies the continuity of the x, y components of the scattered wave field:

$$E_j^{(1)}(\mathbf{k}, \omega) = E_j^{(2)}(\mathbf{k}, \omega), \quad j = x, y, \tag{2}$$

where $E_j^{(n)}(\mathbf{k}, \omega)$ (n = 1, 2) are the coefficients of expansion

$$\mathbf{E}^{(n)}(\mathbf{R}, t) = \mathrm{Re} \int d\mathbf{k} \int_{-\infty}^{+\infty} d\omega\, \mathbf{E}^{(n)}(k, \omega) \exp[i(\mathbf{kr} - (-1)^n k_z^{(n)} z - \omega t)],$$

$$k_z^{(1)} = \left(\frac{\omega^2}{c^2} - k^2\right)^{1/2}, \quad k_z^{(2)} = \left(\varepsilon\frac{\omega^2}{c^2} - k^2\right)^{1/2},$$

$$\mathbf{r} = \{x, y\}, \quad \mathbf{k} = \{k_x, k_y\}. \tag{3}$$

The continuity of the magnetic field intensity means that curl $(\mathbf{E}' - \mathbf{E}) = 0$ for $z = \zeta$. For $\mathbf{E}^{(n)}(\mathbf{k}, \omega)$ this yields

$$\begin{aligned} &i[\mathbf{k}^{(1)}\mathbf{E}^{(1)}(k, \omega)] - i[\mathbf{k}^{(2)}\mathbf{E}^{(2)}(k, \omega)] \\ &= \zeta(\mathbf{k} - \mathbf{k}_0, \omega - \omega_0)\{k_z[\mathbf{k}^{(0)}\mathbf{E}_0] - k_z[\mathbf{k}_1\mathbf{E}_1] - k_z'[\mathbf{k}_2\mathbf{E}_2]\}, \end{aligned} \tag{4*}$$

where $\zeta(\mathbf{q}, \Omega)$ are Fourier transforms of the surface displacements

$$\zeta(\mathbf{r}, t) = \int d\mathbf{q}\, d\Omega\, e^{i(\mathbf{qr} - \Omega t)} \zeta(\mathbf{q}, \Omega),$$

$$\mathbf{k}^{(1)} = \{k_x, k_y, k_z^{(1)}\}, \quad \mathbf{k}^{(2)} = \{k_x, k_y, -k_z^{(2)}\}, \quad \mathbf{k}_0 = \{k_{0x}, 0\}.$$

Solving (2) and (4) together with the transversal condition we obtain

$$E_y^{(n)}(\mathbf{k}, \omega) = iE_{2y} G(\mathbf{k}) \zeta(\mathbf{k} - \mathbf{k}_0, \omega - \omega_0),$$

$$E_x^{(n)}(\mathbf{k}, \omega) = -\frac{k_x k_y}{k_z^{(1)} k_z^{(2)} + k_x^2} E_y^{(n)}(\mathbf{k}, \omega),$$

$$E_z^{(1,2)}(\mathbf{k}, \omega) = \mp \frac{k_z^{(2,1)} E_y^{(n)}(\mathbf{k}, \omega)}{k_z^{(1)} k_z^{(2)} + k_x^2},$$

$$G(\mathbf{k}) = \frac{\omega_0^2 (\varepsilon - 1)[k_z^{(1)} k_z^{(2)} + k_x^2]}{c^2 [k_z^{(1)} + k_z^{(2)}][k_z^{(1)} k_z^{(2)} + k^2]}. \tag{5}$$

Similar formulas but without allowance for dispersion were obtained in the early papers on the theory of spontaneous scattering of light on liquid surface[2].

*$[\mathbf{k}_1\mathbf{E}_1] \equiv \mathbf{k}_1 \times \mathbf{E}_1$

3. Considering the scattering of light on a liquid surface, the latter can be regarded as incompressible with good approximation because the main effect is due to the presence of a fluctuating interface with a sharp change of optical thickness. Experimental results in spontaneous scattering confirm this assumption[3,4].

Hydrodynamic equations for a viscous incompressible liquid taking ponderomotive forces into account have the following form:

$$\frac{\partial \mathbf{v}}{\partial t} = \nu\Delta\mathbf{v} - \frac{\nabla p'}{\rho} - \frac{\partial\varepsilon}{\partial\rho}\frac{\nabla E^2}{8\pi}, \quad \operatorname{div}\mathbf{v} = 0, \tag{6}$$

where $\rho$, $p'$, $\mathbf{v}$, and $\nu$ are the density, pressure, velocity, and coefficient of viscosity respectively. The action of the field on the medium is considered henceforth in a "quasi-stationary" approximation (see[6] for example), i.e., relativistic conditions are neglected.

The boundary conditions at the liquid surface are obtained by the usual method from

$$(\sigma_{ik}' - \sigma_{ik})n_k = -\alpha\Delta\zeta n_i, \tag{7}$$

where $\mathbf{n} = \{-\partial\zeta/\partial x, -\partial\zeta/\partial y, 1\}$ is the unit vector normal to the surface $\zeta(\mathbf{r}, t)$ and $\sigma_{ik}$ and $\sigma_{ik}'$ represent the stress tensor, taking electromagnetic field into account, in liquid and in free space (see for example[6,7]). In this case the explicit form of (7) is

$$n_x\left[p' + \frac{E^2}{8\pi}\left(\varepsilon - \rho\frac{\partial\varepsilon}{\partial\rho} - 1\right)\right] - \eta\left(\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x}\right) = 0,$$

$$n_y\left[p' + \frac{E^2}{8\pi}\left(1 - \varepsilon - \rho\frac{\partial\varepsilon}{\partial\rho}\right)\right] + \frac{E_y}{4\pi}(E_z' - \varepsilon E_z) - \eta\left(\frac{\partial v_y}{\partial z} + \frac{\partial v_z}{\partial y}\right) = 0,$$

$$p' + \frac{E^2}{8\pi}\left(\varepsilon - \rho\frac{\partial\varepsilon}{\partial\rho} - 1\right) + 2\eta\frac{\partial v_z}{\partial z} + \alpha\Delta\zeta = 0, \quad \eta = \rho\nu, \tag{8}$$

where $\alpha$ is the coefficient of surface tension. The term containing only quadratic combinations of $\zeta$ are neglected here.

In the case of incompressible liquid pressure can be redefined in such a way as to eliminate the explicit field-dependence of the volumetric eqs. (6). As a result we have

$$p' = p - \frac{E_{(0)}^2}{8\pi}\left(\varepsilon - \rho\frac{\partial\varepsilon}{\partial\rho} - 1\right) - \rho\frac{\partial\varepsilon}{\partial\rho}\frac{E_{(1)}^2}{8\pi} \tag{9}$$

and (6) and (8) are replaced by

$$\frac{\partial \mathbf{v}}{\partial t} = \nu\Delta\mathbf{v} - \frac{\nabla p}{\rho}, \quad \operatorname{div}\mathbf{v} = 0; \tag{10}$$

$$\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x} = 0,$$

$$n_y(1-\varepsilon)\frac{E_{(0)}}{8\pi} + \frac{E_y}{4\pi}(E_z' - \varepsilon E_z) - \eta\left(\frac{\partial v_y}{\partial z} + \frac{\partial v_z}{\partial y}\right) = 0,$$

$$p + \frac{\varepsilon - 1}{8\pi}E_{(1)}^2 - 2\eta\frac{\partial v_z}{\partial z} + \alpha\Delta\zeta = 0. \tag{11}$$

The indices (0) and (1) denote that only terms that are constant or linear with respect to $\zeta$ respectively are retained in the expansion of $\mathbf{E}^2$ in powers of surface displacement.

Thus we arrive at the usual problem of capillary waves[7], but with different boundary conditions. Therefore the natural frequencies of the capillary waves that can be expected in the solution of this problem are different from those obtainable in the case of zero field.

For the Fourier transforms of velocity and pressure we have

$$\tilde{\mathbf{v}} = (2\pi)^{-3}\int d\mathbf{r}\,dt\,e^{-i(\mathbf{qr}-\Omega t)}\mathbf{v}(\mathbf{R}, t),$$

$$\tilde{p} = (2\pi)^{-3}\int d\mathbf{r}\,dt\,e^{-i(\mathbf{qr}-\Omega t)}p(\mathbf{R}, t),$$

$$\dot{\zeta} = \frac{\partial\zeta}{\partial t} = v_z|_{z=0} \tag{12}$$

and the boundary conditions (11) assume the form

$$\frac{\partial\tilde{v}_x}{\partial z} + iq_x\tilde{v}_z = 0,$$

$$2dq_yE_0^2\tilde{v}_z + \Omega\eta\left(\frac{\partial\tilde{v}_y}{\partial z} + iq_y\tilde{v}_z\right) = 0,$$

$$-i\Omega\tilde{p} + (2iDE_0^2 - \alpha q^2)\tilde{v}_z + 2i\Omega\eta\frac{\partial\tilde{v}_z}{\partial z} = 0, \tag{13}$$

where the dimensionless constants D and d are found from (5):

$$D = \frac{(\varepsilon-1)k_z^2}{8\pi q(k_z + k_z')^2}\{G(\mathbf{k}_0 + \mathbf{q}) - G(\mathbf{k}_0 - \mathbf{q})\},$$

$$d = \frac{k_z^2}{4\pi(k_z + k_z')^2}\left\{\varepsilon - 1 - \frac{1}{2}[g(\mathbf{k}_0 + \mathbf{q}) + g(\mathbf{k}_0 - \mathbf{q})]\right\},$$

$$g(\mathbf{k}) = \frac{k_z^{(2)} + \varepsilon k_z^{(1)}}{k_z^{(1)}k_z^{(2)} + k_x^2}G(\mathbf{k}). \tag{14}$$

Only the terms with capillary wave frequencies $\Omega$ are retained in (13), i.e., quantities of the type $\tilde{v}(\Omega)$ for $\Omega \sim \omega_0$ are dropped.

Since the scattering of light on the surface causes a small variation of frequency $|\omega - \omega_0| \ll \omega_0$, the quantities $k_z^{(n)}$ in (14) are computed for $\omega = \omega_0$.

The dependence of $\tilde{\mathbf{v}}$ and $\tilde{p}$ on z is determined from (10):

$$\tilde{v}_j = A_je^{mz} + Cq_je^{qz}, \quad j = x, y,$$

$$\tilde{v}_z = -\frac{i}{m}(q_xA_x + q_yA_y)e^{mz} - iCqe^{qz},$$

$$\tilde{p} = C\rho\Omega e^{qz}, \quad m = (q^2 - i\Omega/\nu)^{1/2}. \tag{15}$$

A homogeneous system of equations is obtained from (13) for the constants $A_j$ and C. Equating the determinant of this system to zero we find the characteristic equation having the following form in the approximation of weak damping $2\nu q^2 \ll \Omega$:

$$\Omega^2 + 4i\nu q^2\Omega - \frac{\alpha}{\rho}q^3 + 2d\frac{q_y^2}{\rho}E_0^2 + 2iD\frac{q^2}{\rho}E_0^2 = 0. \tag{16}$$

Since our solution concerned a problem linearized with respect to $\zeta$, Eq. (16) coincides with the dispersion equation for the natural capillary wave frequencies when the solution is written in the form

$$\zeta = \int d\mathbf{q}e^{-i(\mathbf{qr}-\Omega t)}\zeta_q$$

with the complex frequency $\Omega$ depending on the wave vector $\mathbf{q}$. From (16) we have

$$\Omega_1 = \Omega_0 + \Delta - i(\gamma_1 - \gamma_2), \quad \Omega_2 = -\Omega_0 - \Delta - i(\gamma_1 + \gamma_2),$$

$$\Delta = -d\frac{q_y^2E_0^2}{\Omega_0\rho}, \quad \gamma_1 = 2\nu q^2, \quad \gamma_2 = -D\frac{q^2E_0^2}{\Omega_0\rho} \quad \Omega = \left(\frac{\alpha q^3}{\rho}\right)^{1/2}. \tag{17}$$

The shift of the real part of the natural frequencies is small in the weak damping approximation, $2\nu q^2 \ll \Omega_0$. At the same time the capillary wave damping determined by the imaginary part of $\Omega$ can vary significantly within the limits of reasonable assumptions about the magnitude of the incident field. If $|\gamma_2| > \gamma_1$ the damping of capillary waves that satisfy this relation changes sign, and the equations of the intermediate waves have solutions that increase in time. This fact was noted in[5] for those capillary waves whose wave vector lies in the plane of incidence of the light beam. It follows from (14) and (17) that in the general case the change of sign of damping is possible only when $\mathbf{k}_0\mathbf{q} > 0$ for $\Omega = \Omega_2$, and when $\mathbf{k}_0\mathbf{q} < 0$ for $\Omega = \Omega_1$ and that the corresponding threshold value of the incident electromagnetic wave intensity is determined by the expression

$$I_0 = \frac{2c\eta q\Omega_0(k_z + k_z')^2}{(\varepsilon - 1)k_z^2[G(\mathbf{k}_0 + \mathbf{q}) - G(\mathbf{k}_0 - \mathbf{q})]}. \tag{18}$$

In this work we consider only the case when the incident light intensity I is less than the threshold value, $I < I_0$.

4. In the presence of field action on the medium, the latter generally speaking cannot be considered in equilibrium. However in our case when only the low-frequency ($\sim\Omega$) component of the nonlinear field-medium interaction is taken into account, we can assume that the medium is in equilibrium and the interaction merely varies its parameters. The spectral intensity of the scattered light is then determined by the level of equilibrium thermal fluctuations of such a modified system and can be expressed by the correlation function of the liquid surface displacement.

In view of the stationary and spatially homogeneous nature of the problem (in the x, y plane) the correlation factors of $\zeta(\mathbf{q}, \Omega)$ and

$$\zeta_q(t) = \int d\Omega\, e^{-i\Omega t}\zeta(\mathbf{q}, \Omega)$$

are related by

$$\begin{gathered}\langle\zeta(\mathbf{q}, t)\zeta^*(\mathbf{q}', t')\rangle = \delta(\mathbf{q} - \mathbf{q}')f_q(t - t'),\\ \langle\zeta(\mathbf{q}, \Omega)\zeta^*(\mathbf{q}', \Omega')\rangle = \delta(\mathbf{q} - \mathbf{q}')\delta(\Omega - \Omega')f(\mathbf{q}, \Omega),\\ f(\mathbf{q}, \Omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dt\, e^{i\Omega t}f_q(t),\end{gathered} \tag{19}$$

where angle brackets denote statistical averaging. The spectral intensity of the scattered light is determined by bilinear combinations of the quantities $E_j^{(n)}(\mathbf{k}, \omega)$ which from (5) and (19) can be expressed in terms of $f(\mathbf{q}, \Omega)$. As a result we obtain a spectral expansion for the scattered light intensity (for $z > 0$):

$$I = \int d\mathbf{k}\int_0^\infty d\omega\, I(\mathbf{k}, \omega), \tag{20}$$

$$I(\mathbf{k}, \omega) = \frac{c|E_2|^2}{16\pi}[f(\mathbf{k} - \mathbf{k}_0, \omega - \omega_0) + f(-\mathbf{k} + \mathbf{k}_0, -\omega + \omega_0)]Z(\mathbf{k}), \tag{21}$$

$$Z(\mathbf{k}) = \frac{\omega_0^4(\varepsilon - 1)^2\{[k_z^{(1)}k_z^{(2)} + k_x^2]^2 + k_y^2[(k_z^{(2)})^2 + k_x^2]\}}{c^4[k_z^{(1)} + k_z^{(2)}]^2[k_z^{(1)}k_z^{(2)} + k^2]^2} \tag{22}$$

Rapidly oscillating terms ($\sim\exp(\pm 2i\omega_0 t)$) and those containing $f(\mathbf{q}, \Omega)$ for $\omega \gg \Omega_0$ are neglected here. The intensity of light scattered into the solid angle $d\varphi d\theta$ within the frequency interval $d\omega$ is given by

$$\begin{gathered}dI = \frac{\omega^2}{2c^2}I(\mathbf{k}, \omega)\sin 2\theta\, d\varphi\, d\theta\, d\omega,\\ k_x = \frac{\omega}{c}\sin\theta\cos\varphi, \quad k_y = \frac{\omega}{c}\sin\theta\sin\varphi.\end{gathered} \tag{23}$$

To compute $f(\mathbf{q}, \Omega)$ we use the well known method[8] based on Onsager's postulate that the time-dependent evolution of fluctuations can be determined from an equation for average values. The solution of macroscopic equations (11) for $\zeta_q(t)$, given initial conditions $\zeta_q(0)$ and $\dot{\zeta}_q(0)$ has the form

$$\zeta_q(t) = \frac{1}{\Omega_1 - \Omega_2}\{(\Omega_1 e^{-i\Omega_2 t} - \Omega_2 e^{-i\Omega_1 t})\zeta_q(0) + i(e^{-i\Omega_1 t} - e^{-i\Omega_2 t})\dot{\zeta}_q(0)\}. \tag{24}$$

The simultaneous correlation factors $\langle\zeta_q(0)\zeta_{q'}^*(0)\rangle$ and $\langle\dot{\zeta}_q(0)\zeta_{q'}^*(0)\rangle$ that are necessary to determine $f(\mathbf{q}, \Omega)$ are computed from Maxwell-Boltzman distribution for capillary waves (see also[1]). The result has the form

$$\begin{gathered}\langle\zeta_q(0)\zeta_{q'}^*(0)\rangle = \frac{\delta(\mathbf{q} - \mathbf{q}')kT}{(2\pi)^2\alpha q^2}\\ \langle\dot{\zeta}_q(0)\zeta_{q'}^*(0)\rangle = 0.\end{gathered} \tag{25}$$

We finally obtain from (24) and (25)

$$f_q(t) = \frac{kT(2\pi)^{-2}}{\alpha q^2(\Omega_1 - \Omega_2)}\{\Omega_1 e^{-i\Omega_2 t} - \Omega_2 e^{-i\Omega_1 t}\}, \quad t > 0, \tag{26}$$

$$f(\mathbf{q}, \Omega) = \frac{kT(2\pi)^{-3}}{\alpha q^2\Omega_0}\left\{\frac{\gamma_1(2\Omega_0 - \Omega) - \gamma_2\Omega_0}{(\Omega - \Omega_0)^2 + (\gamma_1 - \gamma_2)^2} + \frac{\gamma_1(2\Omega_0 + \Omega) + \gamma_2\Omega_0}{(\Omega + \Omega_0)^2 + (\gamma_1 + \gamma_2)^2}\right\} \tag{27}$$

Thus it follows from (21) and (27) that the spectrum of emission scattered in each direction contains the Stokes and anti-Stokes components that are shifted by $\pm\Omega_0$ relative to the incident light frequency $\omega_0$. The spectral width of these maxima is determined by the quantities $\gamma_1 \pm \gamma_2$ and depends on the intensity of the incident light. One of the maxima is sharply narrowed when $|\gamma_2| \to \gamma_1$. In the case of $|k_x| < |k_{0x}|$ the Stokes component narrows down and when $|k_x| > |k_{0x}|$ the anti-Stokes component narrows down.

From the viewpoint of experimental observation of light scattering on liquid surface fluctuations the most desirable seems to be the case when the direction of scattering is close to that of specularly reflected beam $|q| \ll k_0$. The expression for capillary wave damping correction, $\gamma_2$, is in this case reduced with the aid of (14) and (17) to the form

$$\gamma_2 = -\frac{(\varepsilon - 1)^2\omega_0^2(\mathbf{k}_0\mathbf{q})qk_z}{4\pi c^2\Omega_0\rho k_z'(k_z + k_z')^3}|E_0|^2. \tag{28}$$

When scattering is observed in the plane of incidence at an angle of $10^{-2}$ to the direction of regularly reflected beam ($q \sim 10^3$ cm$^{-1}$, $\Omega \sim 10^5$ sec$^{-1}$), and given an angle of incidence of $\pi/6$ and liquid parameters $\eta = 7 \times 10^{-4}$ poise and $\epsilon = 1.7$, Eq. (28) yields the value $I \sim 10^8$ w/cm$^2$ for the critical power of incident radiation at which the observation of significant change in the spectrum of the scattered light ($\gamma_2 \sim \gamma_1$) is possible. Such intensities can be readily obtained from existing sources of coherent monochromatic radiation. However the analysis of the stationary regime is based on the assumption of a sufficiently long pulse $\tau$, since in any case $\tau \gg \Omega_0^{-1} \sim 10^{-5}$ sec. This limitation becomes less rigid ($\tau \gg 10^{-7}$ sec) when the angle of deviation of the scattered beam from the direction of specular reflection is large.

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