

# Complete suppression of metallic reflection upon the resonant excitation of surface plasma waves

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Equations are derived for the reflection of light from a metal surface with a periodic profile near a resonance involving the excitation of surface plasmons. Under certain conditions the reflection of linearly polarized light is completely suppressed.

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The possibility of a resonant excitation of surface plasma waves when light is incident on a rough metal surface underlies several unusual phenomena, including surface-enhanced Raman scattering (a scattering by molecules adsorbed on a surface) and the intensification of second-harmonic generation (see the reviews by Brodskii and Urbakh<sup>1</sup> and Emel'yanov and Koroteev<sup>2</sup> and the bibliographies there). After bombarding a metal target with a laser pulse, Young *et al.*<sup>3</sup> observed surface damage with a periodic structure. To reach a clear understanding of the mechanism for the formation of this structure we need to study the interaction of light with a metal surface with a periodic profile.

In this letter we report results on the reflection of a plane monochromatic light wave of frequency  $\omega$  from a metal surface specified by the equation  $z = b \sin \mathbf{g} \cdot \mathbf{r}$ , where  $z = (\mathbf{n} \cdot \mathbf{r}), (\mathbf{g} \cdot \mathbf{n}) = 0$ ,  $\mathbf{n}$  is the unit vector normal to the unperturbed (plane) surface of the sample, and  $\mathbf{g}$  is the vector of the periodic profile. It is assumed here that the profile depth  $b$  is small in comparison with the wavelength of the light; i.e.,  $bk \ll 1, k = \omega/c$ . To find the relationship between the components of the electric field of the reflected wave,  $E'$ , and of the wave incident on the metal,  $E^0$ , we use the Leontovich boundary condition<sup>4</sup>

$$\left\{ \mathbf{E} - \mathbf{m}(\mathbf{m} \cdot \mathbf{E}) - i \frac{\zeta}{k} [\mathbf{m}, \text{rot } \mathbf{E}] \right\}_{z=b \sin \mathbf{g} \cdot \mathbf{r}} = 0, \quad E = E^0 + E' \quad (1)$$

Here  $\mathbf{m}$  is the normal to the periodic surface, and  $\zeta = \zeta(\omega)$  is the surface impedance, which is assumed to satisfy the inequalities  $|\zeta| \ll 1, \text{Re} \zeta \ll |\text{Im} \zeta|$ .

In principle, we can seek a solution of (1) by formally expanding in powers of the small parameter  $bk$ . In zeroth order in  $bk$ , this expansion reduces to the Fresnel formulas.<sup>4</sup> In first order we find terms  $\sim bk/B(\mathbf{q}_\pm)$ , where  $B(\mathbf{Q}) = \frac{1}{k} \sqrt{k^2 - Q^2} + \zeta, \mathbf{q}_\pm = \mathbf{q} \pm \mathbf{g}_\pm, \mathbf{q}$  is the component of the wave vector of the incident wave which is normal to  $\mathbf{n}, |\mathbf{q}| = k \sin \theta$ , and  $\theta$  is the angle of incidence. If  $\mathbf{g} \neq 0$ , we may have a situation with  $|B(\mathbf{q}_+)| \ll 1$  or  $|B(\mathbf{q}_-)| \ll 1$ —or— $|B(\mathbf{q}_+)|, |B(\mathbf{q}_-)| \ll 1$ . The equation  $B(\mathbf{Q}) = 0$  determines the spectrum and the damping of surface plasmons

with wave vector  $\mathbf{Q}$ , so that this situation corresponds to a resonance with respect to the excitation of surface plasma waves. Since the  $n$ th-order expansion of the field  $E'$  contains terms  $(bk/B)^n$ , we need to sum over the entire perturbation series for  $|B| \lesssim bk$ .

Another solution method—the one we have chosen—is to write the field  $\mathbf{E}'$  as the series

$$E' = \sum_{l=-\infty}^{l=+\infty} \vec{\mathcal{E}}_l \exp [i(\mathbf{q} + l\mathbf{g})r - i\sqrt{k^2 - (\mathbf{q} + l\mathbf{g})^2} z].$$

It turns out that as we raise  $|l|$  to values  $|l| > 1$  the ratio of successive components  $|\mathcal{E}_l|$  is proportional to  $bk$  and does not contain resonant denominators  $B$ . With an accuracy to relative corrections  $\sim (bk)^2$ , the infinite system of algebraic equations which results from (1) can easily be closed. The solution of the system of equations gives us the following relationship between the reflected and incident waves at a single resonance, with  $|B(q_+)| \ll 1$ ,  $|B(q_-)| \sim 1$  [the resonance  $|B(q_-)| \ll 1$  can be reduced to  $|B(q_+)| \ll 1$  through the substitution  $\mathbf{g} \rightarrow -\mathbf{g}$ ]:

$$E'_p = \frac{\cos\theta - \zeta}{\cos\theta + \zeta} \left[ E_p^0 - 2i\omega \frac{|a_p|^2 E_p^0 + a_s a_p^* E_s^0}{F} \right], \quad (2)$$

$$E'_s = -\frac{1 - \zeta \cos\theta}{1 + \zeta \cos\theta} \left[ E_p^0 - 2i\omega \frac{|a_s|^2 E_s^0 + a_s^* a_p E_p^0}{F} \right]. \quad (3)$$

Here the signs of  $p$  and  $s$  correspond to the projections of the amplitudes of the fields  $E^0$  and  $E'$  onto the polarization direction in the plane of incidence and onto the normal to this direction. The other quantities are

$$F = \omega^2 - \omega_p^2(q_+) + i\omega(\gamma_d + \gamma_r), \quad \omega_p(q_+) = c|q_+| \left( 1 - \frac{|\zeta|^2}{2} + A(bk)^2 \right),$$

$$\gamma_d = -\omega \operatorname{Im} \zeta^2, \quad \gamma_r = |a_p|^2 + |a_s|^2,$$

$$a_p = \left( \frac{|\zeta| \omega \cos\theta}{2} \right)^{1/2} bg \frac{\cos\varphi}{\cos\theta + \zeta}, \quad a_s = \left( \frac{|\zeta| \omega \cos\theta}{2} \right)^{1/2} bg \frac{\sin\varphi}{1 + \zeta \cos\theta},$$

where  $\omega_p(q_+)$  is the renormalized frequency of a surface plasmon with wave vector  $q_+$ ;  $A$  is a dimensionless function of  $\mathbf{q}$ ,  $\mathbf{g}$ , and  $k$ , on the order of unity;  $\varphi$  is the angle between  $\mathbf{g}$  and  $\mathbf{q}$ ; and  $\gamma_d$  and  $\gamma_r$  are, respectively, the dissipative and radiative parts of the plasmon damping. The radiative part results from the conversion of a plasmon into a photon.

At the double resonance [ $|B(q_+)|, |B(q_-)| \ll 1$ ] we have

$$E'_p = \frac{\cos\theta - \zeta}{\cos\theta + \zeta} E_p^0, \quad E'_s = -\frac{1 - \zeta \cos\theta}{1 + \zeta \cos\theta} \left[ 1 - 4i\omega \frac{|a_s|^2}{\widetilde{F}} \right] E_s^0. \quad (4)$$

Here  $\tilde{F} = \omega^2 - \omega_p^2(q_+) + i\omega(\gamma_d + \tilde{\gamma}_r)$   $\tilde{\gamma}_r = 2|a_s|^2$ .

The first terms in square brackets in Eqs. (2)–(4) correspond to reflection from a plane surface (the non-resonant contribution). The second terms in the square brackets give the contribution of the resonant excitation of surface plasmons. Because of this excitation, the intensity of the reflected light is a strong function of  $\omega$ ,  $\theta$ ,  $\mathbf{g}$ , and  $b$ . The most obvious manifestation of this dependence is the possible complete suppression of reflection from a metal surface with a periodic profile, because the resonant and nonresonant contributions cancel out. For linearly polarized light under the condition  $\cos\theta \gg |\zeta|$  the complete-suppression effect sets in under the conditions

$$\omega = \omega_p(q_+), \quad \varphi = \arctg(h \cos\theta) \pm \frac{\pi}{2}, \quad h = \frac{E_p^0}{E_s^0}, \quad (5)$$

$$bk = 2 \left( \frac{\operatorname{Re} \zeta}{\cos^3 \theta} \right)^{1/2} \left( 1 \pm \frac{h \sin \theta}{\sqrt{1+h^2}} \right) \times \begin{cases} 1 & \text{for } h \sin \theta \gg \frac{\gamma_d}{2\omega} \\ 2^{-1/2} & \text{for } h \sin \theta \ll \frac{\gamma_d}{2\omega} \end{cases}$$

It is important to note that the profile depths found from these conditions satisfy the smallness condition  $bk \ll 1$ . The angle  $\varphi$  in (5) corresponds to an orientation of the vector  $\mathbf{g}$  along the projection of the electric field of the incident wave onto the unperturbed surface. It follows from (5) that the reflection for  $p$ -polarized light ( $E_s^0 = 0$ ) and  $s$ -polarized light ( $E_p^0 = 0$ ) is completely suppressed under the conditions  $\mathbf{g} \parallel \mathbf{q}$  and  $\mathbf{g} \perp \mathbf{q}$ , respectively. The periods of the sinusoidal profile,  $d = 2\pi/g$ , are related to the light wavelength  $\lambda = 2\pi c/\omega$  by

$$d = \frac{\lambda}{1 \pm \sin\theta} \quad \text{for } E_s^0 = 0 \quad d = \frac{\lambda}{\cos\theta} \quad \text{for } E_p^0 = 0. \quad (6)$$

The periods of the damage structure observed in Ref. 3 agree precisely with (6). On this basis it may be suggested that the structures may form as a result of the onset of an unstable process which gives rise to a surface profile having the property of maximum light reflection.

Formulas analogous to (2) and (3) were derived in Ref. 5 through a formal expansion, in which terms of up to second order in  $bk$  were retained. However, those formulas do not contain terms of order  $(bk)^2$  in the denominator  $F$  and thus become physically meaningless under the conditions  $\gamma_r \gtrsim \gamma_d, |\omega - \omega_p|$ : The intensity of the reflected light predicted by these formulas may become greater than that of the incident light.

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<sup>1</sup>A. M. Brodskii and M. I. Urbakh, Usp. Fiz. Nauk **138**, 413 (1982) [Sov. Phys. Usp. **25**, 810 (1982)].

<sup>2</sup>V. I. Emel'yanov and N. I. Koroteev, Usp. Fiz. Nauk **135**, 345 (1981) [Sov. Phys. Usp. **24**, 864 (1981)].

<sup>3</sup>I. F. Young, J. S. Preston, H. M. van Driel, and J. E. Sipe, Phys. Rev. **B 27**, 1155 (1983).

<sup>4</sup>L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Nauka, Moscow, 1982, p. 414.

<sup>5</sup>G. S. Agarwal and S. S. Iha, Phys. Rev. **B26**, 482 (1982).

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