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PLASMA RADIATION BY ROUGH SURFACES*

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The recent observation of a peak of scattered radiation at the plasma frequency from thin metal foils irradiated with light can be explained in terms of a small amount of surface roughness of the foils.

Recent experiments[1-3] have measured a peak of electromagnetic radiation at the plasma frequency excited by an electromagnetic wave incident on metal foils. An interesting aspect of these experiments is that the plasma radiation occurs at angles differing from the transmitted or reflected angles. As is well known in optics, and as has been shown for this particular problem,[4] when a plane wave is incident on a plane parallel slab the emitted electromagnetic radiation is only in the transmitted and reflected directions. Even at the plasma frequency where the plasma mode is excited by the incident radiation, the emitted radiation is only in the two directions. The effect of the plasma mode is to produce a dip in the transmission and a peak in the reflection as has been experimentally verified.[5] The explanation of the recent experiments requires that the foil not be a plane parallel slab—also noted by the authors of Ref. 2. In practice, experimental foils do not have perfectly plane surfaces but somewhat rough ones, and we show here that this surface roughness can explain the experimental results. One contribution to the surface roughness might be surface phonons but a recent experiment[6] has shown that this contribution is negligible by showing that there is no appreciable frequency shift in the scattered radiation.

One expects for good experimental foils that the roughness will be small. An incident electromagnetic wave would then set up approximately the same fields and currents that are present in a perfectly smooth foil of the same average thickness. Consider the difference in the currents for the cases of a perfectly smooth foil and a good experimental one of the same average thickness. This difference will be appreciable only at the surfaces of the foils where they do not overlap. It is just this difference in current which produces the difference in emitted radiation between the rough and smooth foils. Thus our problem is reduced to finding the radiation from a distribution of surface currents. We are interested in the plasma mode, which can be excited only by radiation with an electric field normal to the surface.[7,8] Only surface currents normal to the boundary produce normal electric fields, and we limit ourselves to calculating the effects of such currents.

We assume that the height of the rough structure on the foil is much less than both the wavelength of the radiation and the thickness of the film. In such a case the variation of the current in the direction normal to the surface does not enter into the problem and we only have to consider current variations along the surface. The radiation by these currents from a small element is the same as that from a small dipole on the surface of a smooth foil. The electromagnetic properties of the foil are determined by the frequency-dependent dielectric constant ϵ. We assume that the foil is surrounded on either side by vacuum and the surfaces of the foil are at $z=\pm\frac{1}{2}\tau$. The radiation from such a dipole has been previously determined in the course of calculating the transition radiation from smooth foils.[8] In this calculation the radiation from dipoles on either side of the boundary has been found, and a discontinuity of this radiation occurs when the boundary is crossed. The radiation from a dipole straddling the boundary can be found by averaging the contribution from dipoles just on either side of the boundary. By integrating the contributions from these surface currents, we find that the radiated power per unit solid angle, per unit frequency range, and per unit area of the foil is given by[8,9]

$$T_{\Omega,\omega} = (2\pi)^2(\omega^4/Ac^3)\sin^2\theta\cos^2\theta\,|U_{k_\perp\omega}|^2, \tag{1}$$

where $k = (\omega/c)\sin\theta$,

$$U_{k_\perp\omega}^{(r)} = \int dx_0 dy_0\, j(x_0y_0)\exp[-i(k_y y_0 + k_x x_0)]W_{k_\perp\omega}, \tag{2}$$

and the integration is performed over both surfaces of the foil. Here θ is the angle between the normal to the foil and the direction of the radiation, $d\Omega$ is a solid angle element, and A is the area of the foil. In addition, $W_{k_\perp\omega}$, for the surface nearest to the observer, is given by

$$W_{k_\perp'}^{+} = \frac{(\epsilon+1)e^{-K(z-\frac{1}{2}\tau)}(e^{K'\tau}-fe^{-K'\tau})}{2\pi c(\epsilon K+K')(e^{K'\tau}-f^2e^{-K'\tau})}. \tag{3}$$

For the other surface,

$$W_{k_\perp\omega}^{-} = \frac{e^{-K(z+\frac{1}{2}\tau)}}{\pi c}\,\frac{K'(\epsilon+1)}{(e^{K'\tau}-f^2e^{-K'\tau})(\epsilon K+K')^2}, \tag{4}$$

where

$$K = [k_\perp^2 - \epsilon(\omega/c)^2]^{1/2},$$
$$k_\perp^2 = k_x^2 + k_y^2,$$
$$K' = [k_\perp^2 - \epsilon(\omega/c)^2]^{1/2},$$
$$f = (\epsilon K - K')(\epsilon K + K')^{-1}. \tag{5}$$

The dipole is a current element of magnitude $j(x_0, y_0)dA$ at $(x_0, y_0, \frac{1}{2}\tau)$ on the surface, c is the velocity of light, and the radiation from a single dipole is obtained by letting $U_{k_\perp\omega} = W_{k_\perp\omega}$ in (1).

The current density $j(x_0y_0)$ is driven by the incident electromagnetic radiation and thus will be in phase with it. We separate out this phase factor by writing

$$j(x_0y_0) = j_0(x_0y_0)\,e^{iqx_0}, \tag{6}$$

where now the variation of $j_0(x_0y_0)$ along the surface depends only on the properties of the surfaces. By use of (2) and (6) we find

$$|U_{k_\perp\omega}|^2/A = |W_{k_\perp\omega}|^2 \int dt ds\, g(ts)\exp\{i[q-k_x)t - k_y s]\}, \tag{7}$$

where $t = x_0 - x_0'$, $s = y_0 - y_0'$, and

$$g(t,s) = \frac{1}{A}\int dx_0 dy_0\, j_0(x_0y_0)j_0^*(x_0-t, y_0-s). \tag{8}$$

The function $g(t,s)$ is a measure of the average correlation of two current elements a distance $\rho = (t^2+s^2)^{1/2}$ apart and for a homogeneous film has cylindrical symmetry. The correlation distance over which $g(t,s)$ remains appreciable is a measure of the average dimension along the surface of the pits or bumps. For lack of any more detailed knowledge, we assume the physically reasonable form

$$g(t,s) = I_0^2\frac{4}{\pi\sigma^2}\exp\left(\frac{-(t^2+s^2)}{\sigma^2}\right). \tag{9}$$

Here σ is a measure of the average dimensions along the surface of the surface imperfections, and

$$I_0^2 = \langle j_0^2\rangle \pi\sigma^2/4, \tag{10}$$

where $\langle j_0^2\rangle$ is the mean-square average of j_0 over the foil surface. We see from (1) and (7) that the radiation depends on two separate factors, one being the radiation pattern from a single dipole and the other being the Fourier transform of the surface-current spatial-correlation function. The Fourier transform of $g(t,s)$ with the special form given in (9) is easily performed and we find that

$$|U_{k_\perp\omega}|^2/A = 4I_0^2 \exp\{-[(q-k_x)^2+k_y^2]\sigma^2/4\}(|W_{k_\perp\omega}^+|^2+|W_{k_\perp\omega}^-|^2). \tag{11}$$

All quantities are now determined except for I_0. The determination of I_0 requires a detailed knowledge of the roughness of the surface such as the shape of the bumps and pits. Such information is not available, but we can use some general arguments to obtain a sufficiently precise estimate of I_0. Firstly, I_0 will be proportional to the foil current densities in the vicinity of and normal to the surface. From our assumption of small roughness, this normal current density should be closely given by that at the surface of a smooth film. Secondly, I_0 will be proportional to the height of the bumps and the depth of the pits, and to their number per unit area. In fact, using (10) I_0^2 can be written as

$$I_0^2 = (a\omega^2|\epsilon-1|^2|E_n|^2/64\pi)\delta^2\sigma^2, \tag{12}$$

where $|E_n|^2$ is the square of the magnitude of the normal component of the electric field inside and near the surface of a smooth foil, δ^2 is the mean square height of the deviations of the rough surface from the smooth foil, and a is a geometric factor of order 1, which should not depend very strongly on ϵ.

A typical experiment would consist of observing the scattered light as a function of the frequency, normalizing the result to a constant incident power. Denoting the magnitude of the incident electric field by E_i, the incident power is given by $|E_i|^2c$. Thus the ratio of the scattered power $T_{\Omega,\omega}$ to the incident power T_i is, using (1), (11), and (12),

$$\frac{T_{\Omega,\omega}}{T_i} = \frac{\pi}{4c^4}\omega^6 a|\epsilon-1|^2\left|\frac{E_n}{E_i}\right|\sigma^2\delta^2\sin^2\theta\cos^2\theta\exp\{-[(q-k_x)^2+k_y^2]\sigma^2/4\}(|W_{k_\perp,\omega}^+|^2+|W_{k_\perp\omega}^-|)^2, \tag{13}$$

where

$$\left|\frac{E_n}{E_i}\right|^2 = \frac{\sin^2\theta_0\cos^2\theta_0}{|(\sin^2\theta_0-\epsilon)|}\left|\frac{(1-f_0)\left(e^{K_0'\tau}-f_0e^{-K_0'\tau}\right)}{e^{K_0'\tau}-f_0^2e^{-K_0'\tau}}\right|^2$$

$$K_0' = (\omega/c)(\sin^2\theta_0-\epsilon)^{1/2},$$

$$k_x = \omega/c\sin\theta\cos\varphi,$$

$$k_y = \omega/c\sin\theta\sin\varphi,$$

$$q = (\omega/c)\sin\theta_0,$$

and $f_0 = [-i\epsilon(\omega/c)\cos\theta_0 - K_0'][-i\epsilon(\omega/c)\cos\theta_0+K_0']^{-1}$. Here θ_0 is the angle between the foil normal and the direction of the incident light, and φ is the angle between the polarization planes of the incident and emitted light. The expression for $|E_n/E_i|$ is obtained from a straightforward application of Maxwell's equation.

The expression for the radiation given by (13) is rather involved and it is worthwhile to consider the limit of a thin film. In this limit $\omega\tau/c$ and $K'\tau \ll 1$. Assuming an ideal situation where ϵ_2, the imaginary part of ϵ, is small and constant in the vicinity of the plasma frequency, one finds that $T_{\Omega\omega}/T_i$ has a peak at the plasma frequency ω_p determined by the real part of $\epsilon=0$, and its peak value is

$$\left(\frac{T_{\Omega\omega}}{T_i}\right)_{\max} \approx \frac{\sin^2\theta\,\sin^2\theta_0\cos^2\theta\cos^2\theta_0\,\omega_p{}^4 a\delta^2\sigma^2}{2\pi c^4\beta^2\beta_0{}^2}\exp\{-[(q-k_x)^2+k_y{}^2]\sigma^2/4\}, \tag{14}$$

where $\beta=(\omega_p\tau/c)\sin^2\theta+2\epsilon_2\cos\theta$, $\beta_0=(\omega_p\tau/c)\sin^2\theta_0+2\epsilon_2\cos\theta_0$, and ϵ_2 is the imaginary part of ϵ. Its frequency halfwidth $\Delta\omega$ is

$$\Delta\omega=\frac{\sqrt{2}}{BB_0}\{[(B^2+B_0{}^2)^2+4B^2B_0{}^2]^{1/2}-(B^2+B_0{}^2)\}^{1/2}\left|\frac{d\epsilon_1}{d\omega_p}\right|^{-1}, \tag{15}$$

where

$$B=2\cos\theta/\beta,$$

$$B_0=2\cos\theta_0/\beta_0,$$

and $d\epsilon_1/d\omega_p$ is the derivative of the real part of ϵ with respect to frequency evaluated at $\omega=\omega_p$. In good experimental foils one expects that the angular variation of the exponential in (14) is small and that the angular dependence comes mainly from the other factors.

The theory presented here is capable of explaining the experiments.[1-3] The maximum in the plasma radiation[1] at a thickness of 300 Å could be understood by assuming that the roughness $\sigma^2\delta^2$ increases with thickness. By (14), for small τ the radiation intensity increases as $\sigma^2\delta^2$, while for larger τ it decreases as $\sigma^2\delta^2/\tau^4$ and presumably reaches its maximum at 300 Å. Further comparisons with experiment are given by Kretschmann and Raether,[10] where the agreement between theory and experiment is verified in more detail. Wilems and Ritchie[11] present a quantum calculation of the same problem treated here but limited to a thin slab of a free-electron gas. Such a treatment is not as appropriate as the one given here for a metal like Ag which deviates greatly from free-electron behavior.

In summary, the peak in scattered radiation at the plasma frequency can be quantitatively understood to be caused by a small amount of surface roughness of the experimental foils. This suggests that such measurements can give quantitative information on the surface roughness of foils.

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