THEORY OF CHARACTERISTIC ENERGY LOSSES IN THIN FILMS

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The energy losses of a fast charged particle moving through a plasma layer are computed by quantum field-theoretic methods. The losses due to excitation of volume and surface plasmons are considered as well as those due to binary collisions at the surface and inside the plasma layer. Primary attention is given to the surface plasmons, which have not been studied before in any detail. We consider the dependence of the plasmon spectrum on the thickness in thin films; the dependence reflects the discreteness of the plasma electron momentum. Certain features of the energy spectrum of the surface plasmons in a plasma with diffuse boundaries have also been studied.

THE characteristic losses arising in the passage of a fast charged particle through a plasma have been treated many times, both classically and by quantum-theoretic methods [1-3] (Silin and Rukhadze [2] give an extensive bibliography on this subject). In all of this work the plasma has been regarded as an unbounded system and edge effects have been neglected; this approach to the problem leads to a number of difficulties. In the first place, neglecting edge effects means that the so-called surface plasmons do not appear, but in recent years the effect of surface plasmons has become extremely important from both the theoretical [4-7] and experimental [8-9] points of view. It was first pointed out by Ritchie [4] that the losses due to the excitation of surface plasmons in very thin films can be decisive in certain cases. [1)] Second, the edge effect is manifest in the reduction in the characteristic losses due to the excitation of volume plasmons. Finally, the existence of surface plasmons implies a modification of the volume-plasmon spectrum in thin films.

The present work is devoted to an investigation of the above-mentioned features of the problem. The stopping power of the plasma layer is expressed in terms of an electromagnetic field correlation function, which is simply related to the retarded Green's function. In the analysis we shall only take account of the Coulomb interaction between particles. The resulting expression then contains the losses of the charged particle due to the excitation of both volume and surface plasmons (collective losses) as well as those due to binary collisions at the surface and within the film. When spatial dispersion is neglected (collective losses only) our results coincide with the corresponding results given by Ritchie; [4] if edge effects are neglected our results coincide with those given by Larkin. [3]

Thin metal films (~ 100 Å) exhibit an experimentally observable dependence of plasma frequency on film thickness; this effect is due specifically to the quantum nature of the electrons in the film, more precisely, the discreteness of the electron momentum. The discreteness in electron momentum is much more important than the discreteness in the plasmon wave vector considered by Ichikawa. [14]

Finally, we consider surface plasmons in a plasma with diffuse boundaries. An important factor in this case is the specific plasma damping of surface plasmons due to the conversion of the energy of the surface plasmon into the energy of a volume plasmon in regions in which volume plasmons can exist i.e., in the region $\epsilon(\omega, \mathbf{r}) = 0$.

[1)] It should be noted that the solutions given by Garibyan, Silin, and Fetisov in the analysis of transition radiation and the skin effect [10-12] contain fields corresponding to the surface plasmons, since these are exact solutions. However, since the surface plasmons are neither excited by transverse waves nor radiate transverse waves [6] they actually drop out of the analysis in these problems. The statement by Ferrell [5] that surface plasmons make a contribution to the transition radiation [11] in thin films is also erroneous. The contribution to this radiation is actually due to the so-called optical plasmons, which only exist in thin films; [13] the net effect is to make the intensity of the transition radiation higher in thin films than in thick films, a result that has been verified experimentally.

1. TRANSITION PROBABILITY

We consider a plasma slab of thickness d ($0 < x_3 < d$) bounded on both sides by a nonabsorbing dielectric medium whose dielectric constant is set equal to unity. Assume that the particle moving through the slab has mass M and velocity v and that the velocity is high enough so that the interaction with the particles in the plasma can be treated by perturbation-theoretic methods. The system Hamiltonian is

$$\hat{H} = \hat{H}_n + \hat{H}_0 + \hat{H}_1, \tag{1}$$

where $\hat{H}_n$ is the Hamiltonian for the plasma slab, which contains all interactions between the particles, $\hat{H}_0$ is the Hamiltonian for the freely moving particle, and $\hat{H}_1$ is the Hamiltonian that describes the interaction between the incident particle and the plasma:

$$\hat{H}_1 = -\int \hat{j}_\mu(x)\,\hat{A}_\mu(x)\,d^3x, \tag{2}$$

where $\hat{j}_\mu(x)$ is the operator characterizing the current associated with the moving particle; it will be assumed hereinafter that the current operator commutes with the operators of the plasma particles (exchange effects are neglected). To first order in $\hat{H}_1$, in the usual way we now obtain the probability for transition of the external particle from state p into state p′ by summing over final states and statistically averaging over initial plasma states (the states p and p′ are not necessarily plane-wave states)

$$W_{p\to p'} = i\int \langle p|j_\mu(x)|p'\rangle\langle p'|j_\nu(x')|p\rangle D_{\mu\nu}^{+}(x,x')\,d^4x\,d^4x', \tag{3}$$

where

$$D_{\mu\nu}^{+}(x, x') = \langle \hat{A}_\mu(x)\,\hat{A}_\nu(x')\rangle \tag{4}$$

is the electromagnetic field correlation function; the Fourier components of this function are related to the retarded Green's function $D^R_{\mu\nu}(\mathbf{x}, \mathbf{x}', \omega)$ by the expression:[15]

$$D_{\mu\nu}^{+}(\mathbf{x},\mathbf{x}',\omega) = \frac{2i}{1-e^{-\omega\beta}}\,\mathrm{Im}\,D_{\mu\nu}^{R}(\mathbf{x},\mathbf{x}',\omega). \tag{5}$$

The average in (4) is taken over a Gibbs distribution.

Assuming that the initial and final electron states are stationary states with momenta $\mathbf{p}$ and $\mathbf{p}' = \mathbf{p} - \mathbf{k}$ and corresponding energies $\epsilon_\mathbf{p}$ and $\epsilon_{\mathbf{p}'}$ we obtain the transition probability per unit time from (3):

$$W_{\mathbf{p}\to\mathbf{p}'} = -\frac{2a_{\mu\nu}(\mathbf{p},\mathbf{p}')\,\mathrm{Im}\,D_{\mu\nu}^{R}(\mathbf{k},\mathbf{k},\omega)}{1-e^{-\omega\beta}}; \tag{6}$$

$$D_{\mu\nu}^{R}(\mathbf{k},\mathbf{k},\omega) = \int D_{\mu\nu}^{R}(x,x')\,e^{-i[\mathbf{k}(\mathbf{x}-\mathbf{x}')-\omega(t-t')]}\,d^3x\,d^3x'\,dt,$$

$$\langle \mathbf{p}|j_\mu(x)|\mathbf{p}'\rangle\langle\mathbf{p}'|j_\nu(x')|\mathbf{p}\rangle = a_{\mu\nu}(\mathbf{p},\mathbf{p}')\,e^{-i[\mathbf{k}(\mathbf{x}-\mathbf{x}')-\omega(t-t')]}, \tag{7}$$

$\omega = \epsilon_\mathbf{p} - \epsilon_{\mathbf{p}-\mathbf{k}}$. Thus the entire problem has been reduced to that of finding the retarded Green's function for the electromagnetic field of the system.

In an infinite uniform plasma

$$D_{\mu\nu}^{R}(\mathbf{k},\mathbf{k},\omega) = VD_{\mu\nu}^{R}(\mathbf{k},\omega) \tag{8}$$

(V is the volume of the system) and the results of Larkin[3] and Alekseev[16] are obtained.

Only the Coulomb interaction of the incoming particle with the plasma is considered. In this case (6) yields

$$W_{\mathbf{p}\to\mathbf{p}'} = \frac{2e^2}{1-e^{-\omega\beta}}\,\mathrm{Im}\,D^R(\mathbf{k},\mathbf{k},\omega) \qquad (D^R = D_{44}^R). \tag{9}$$

2. ENERGY SPECTRUM

In finding $D^R(\mathbf{k}, \mathbf{k}', \omega)$ it will be found convenient to start with the equation for the temperature Green's function $\bar{D}(x, x')$ which is of the form[15]

$$\Delta\bar{D}(x,x') - \int \Pi(x,y)\,\bar{D}(y,x')\,d^4y = \delta^4(x-x'), \tag{10}$$

where $\Pi(x, y)$ is the polarization operator of the system while the Fourier components $R^R(\mathbf{x}, \mathbf{x}', \omega)$ and $\bar{D}(\mathbf{x}, \mathbf{x}', k_4)$ obey the relations

$$\bar{D}(k_4, \mathbf{x}, \mathbf{x}') = D^R(\omega = ik_4, \mathbf{x}, \mathbf{x}'), \qquad k_4 > 0. \tag{11}$$

Since the exact value of $\Pi(x, y)$ can not be found we must make a number of simplifying assumptions at this point.

1. We assume that the following inequalities are satisfied

$$|d\lambda_e(\mathbf{x})\,/\,dx| \ll 1, \qquad r_D\,/\,r_0 \ll 1, \tag{12}$$

where $\lambda_e(\mathbf{x})$ is the DeBroglie wavelength, r_D is the Debye radius, and r_0 is the characteristic scale length of the inhomogeneity in the system (hereinafter this distance will be set equal to the thickness of the plasma slab). The semiclassical approximation[17] can then be used to describe the individual electron characteristics.

2. In the usual way we assume for simplicity that the ion charge is uniformly smeared over the thickness of the film. Then the space charge, which is due primarily to the contact potential difference and free carriers, is localized at the boundary (for example, near a pn junction). The size of this region (L) is an inverse function of the carrier concentration. For example, in metals $L \ll 10^{-7}$ cm; in semiconductors used in tunnel diodes $L \sim 10^{-6}$ cm.

Several different limiting cases exist depending on the relation between L, d, and λ_f (the distance over which the electromagnetic field changes significantly, say the wavelength): a) the geometric-optics approximation

$$d \gtrsim L \gg \lambda_f \tag{13}$$

or in more general form

$$|d\lambda_f(\mathbf{x}) / d\mathbf{x}| \ll 1, \tag{13'}$$

b) the approximation in which the boundary separating the two uniform media is sharp

$$L \ll \lambda_f; \qquad L \ll d, \tag{14}$$

c) the inhomogeneous system

$$d \gg L \sim \lambda_f. \tag{15}$$

The energy spectrum of the plasma in case a) has been investigated fairly completely in[18]. Here we shall be concerned with case b).

The condition in (14) states that the nature of the charge distribution in the region L is unimportant. Thus, we assume that the surplus charge in this region is distributed over the surface. This refers to electrons at the Tamm surface levels, adsorption states, and other states localized close to the surface. For simplicity we assume that the relative contribution of these surface electrons is small in the process at hand i.e., these electrons are shunted by the internal electrons. Under these assumptions the solution of Eq. (10) is not especially difficult. Since the solution is a complex one we shall not reproduce it here; instead we write the expression for the probability (9)

$$W = \frac{4e^2}{1 - e^{-\omega\beta}} \times \operatorname{Im}\Big\{ \frac{1}{\alpha_c + 1/k_\parallel} \Big[\frac{2}{d} k_\perp \sin\frac{k_\perp d}{2} \sum_{k_3^c} \frac{1}{(k_3^2 - k_\perp^2)\mathbf{k}^2\varepsilon(\mathbf{k}, \omega)} - \frac{k_\perp \sin(k_\perp d/2) - k_\parallel \cos(k_\perp d/2)}{k_\parallel (k_\perp^2 + k_\parallel^2)} \Big]^2$$
$$+ \frac{1}{\alpha_a + 1/k_\parallel} \Big[\frac{2}{d} k_\perp \cos\frac{k_\perp d}{2} \sum_{k_3^a} \frac{1}{(k_3^2 - k_\perp^2)\mathbf{k}^2\varepsilon(\mathbf{k}, \omega)} - \frac{k_\perp \cos(k_\perp d/2) + k_\parallel \sin(k_\perp d/2)}{k_\parallel (k_\perp^2 + k_\parallel^2)} \Big]^2$$
$$- k_\perp^2 \Big[\frac{2}{d} \sin^2\frac{k_\perp d}{2} \sum_{k_3^c} \frac{1}{(k_3^2 - k_\perp^2)^2\mathbf{k}^2\varepsilon(\mathbf{k}, \omega)} + \frac{2}{d}\cos^2\frac{k_\perp d}{2} \times \sum_{k_3^a} \frac{1}{(k_3^2 - k_\perp^2)^2\mathbf{k}^2\varepsilon(\mathbf{k}, \omega)} \Big] \Big\}. \tag{16}$$

Here, $\epsilon(\mathbf{k}, \omega)$ is the plasma dielectric constant while $k_\perp$ and $k_\parallel$ are the perpendicular and parallel components of k (with respect to the surface of the film)

$$\alpha_{c,a} = \frac{2}{d} \sum_{k_3^{c,a}} \frac{1}{\mathbf{k}^2\varepsilon(\mathbf{k}, \omega)}, \tag{17}$$

$$k_3^c = 2n\pi / d, \quad k_3^a = (2n+1)\pi / d, \qquad n = 0, \pm 1, \pm 2\ldots \tag{18}$$

It is evident from (16) that the energy loss of a fast particle traversing a plasma slab appears at the absorption poles and at discrete frequencies given by the dispersion relations

$$\varepsilon(\mathbf{k}_\parallel, k_\perp) = 0, \tag{19}$$

$$\alpha_{c,a}(\mathbf{k}_\parallel, \omega) + 1 / k_\parallel = 0. \tag{20}$$

The dispersion relation in (19), which corresponds to the so-called volume plasmon, has been thoroughly investigated by many authors. The dispersion relations in (20) correspond to tangential ($\alpha = \alpha_c$) and normal ($\alpha = \alpha_a$) plasmons. It is evident from (18) and (20) that the normal and tangential plasmons are due to electron transitions of various kinds. The normal plasmons arise from electron transitions between states of opposite parity (summation over k_3^a) while the tangential plasmons arise from transitions between states of the same parity (the summation is taken over k_3^c).

It is interesting to note that odd electron states do not appear in the application of periodic boundary conditions and this leads to the absence of normal plasmons. The problem at hand is a case in which the energy spectrum of a system depends qualitatively on the concrete form of the boundary conditions. The correct boundary conditions are especially important when the film is very thin because in this case the normal plasmon energy spectrum is considerably different from that of tangential plasmons; moreover, in thin films greatest interest attaches to the normal plasmons because these make the chief contribution to transition radiation[13] (here we are concerned with optical normal plasmons).

For the case of a one-component plasma in the free electron approximation when

$$\omega^2 c^{-2} \ll k_\parallel^2 \ll \omega^2 v_T^{-2} \tag{21}$$

the dispersion relations (20) yield the two branches

$$\omega_{n,\tau} \approx \frac{\omega_0 (1 \pm e^{-k_\parallel d})^{1/2}}{\sqrt{2}} + \frac{\omega_{n,\tau}^2}{\omega_0^2} a_{n,\tau} k_\parallel v_T - i \frac{|\varepsilon(\omega_{n,\tau})|^2}{\sqrt{\pi}\,|\bar{\varepsilon}|^2} k_\parallel v_T, \tag{22}$$

$$a_{n,\tau} = a(\omega_{n,\tau}/\omega_0, d), \qquad \bar{\varepsilon} = \varepsilon(\mathbf{k}, \omega)\big|_{\omega \sim k v_T}, \tag{23}$$

where v_T is the thermal velocity of the plasma electrons and ω_0 is the plasma frequency. The meaning of the various terms in (22) is discussed in[13] and will not be considered further here.

In thin films, ($k_{||}d \ll 1$) (22) gives

$$\omega_n \approx \omega_0 (1 - k_{\parallel} d/4) + a_n k_{\parallel} v_T - i \frac{(k_{\parallel} d)(k_{\parallel} v_T)}{2\sqrt{\pi}\,|\bar{\varepsilon}|^2},$$

$$\omega_\tau \approx \omega_0 \sqrt{k_{\parallel} d/2} + {}^1/_2 a_\tau (k_{\parallel} d)(k_{\parallel} v_T) - i(2v_T/d)/\sqrt{\pi}\,|\bar{\varepsilon}|^2. \qquad (22')$$

In thick films ($k_{||}d \gg 1$)

$$\omega_n \approx \omega_\tau \approx \omega_0/\sqrt{2} + {}^1/_2 a_{n,\tau} k_{\parallel} v_T - i k_{\parallel} v_T / 2\sqrt{\pi}\,|\bar{\varepsilon}|^2. \qquad (22'')$$

The dispersion relation in (22″) is interesting: because of spatial dispersion the energy loss due to the excitation of surface plasmons is a linear function of the scattering angle ϑ:

$$\Delta E \approx \omega_0/\sqrt{1+\varepsilon_0} + a_{n,\tau} p v_T \vartheta / 2(1+\varepsilon_0^{-1}) \qquad (24)$$

(ϵ_0 is the dielectric constant of the medium surrounding the film, for example an oxide of the film, which has not been considered before); this is in contrast with the quadratic dependence of the loss due to the excitation of volume plasmons. To verify (24) we have taken experimental points for Al from the work of Kunz[9] which appear to give a good fit to the line (24) with $a_{n,\tau} \approx 0.8$. It should be noted that very few data points were available and it would be desirable to carry out more careful experiments in order to check this result.

To treat high-momentum excitations $\epsilon(\mathbf{k}, \omega)$ should be given by the expression:[2]

$$\varepsilon(\mathbf{k}, \omega) = 1 - \frac{\omega_0^2}{\omega^2 - (\mathbf{k}^2/2m)^2}. \qquad (25)$$

Substituting (25) in (20) we obtain two kinds of excitations: surface excitations, characterized by the frequencies

$$\omega_{s,n}^2 \approx \omega_0^2 + \frac{k_{\parallel}^4}{4m^2}\left(1 - \frac{\omega_0^4}{2\omega_{s,n}^4[1 + \text{th}(k_{\parallel}d/2)]^2}\right) \qquad (26)^*$$

(normal surface plasmons) and

$$\omega_{s,\tau}^2 \approx \omega_0^2 + \frac{k_{\parallel}^4}{4m^2}\left(1 - \frac{\omega^4}{2\omega_{s,\tau}^4[1 + \text{cth}(k_{\parallel}d/2)]^2}\right) \qquad (27)^\dagger$$

(tangential surface plasmons) and volume excitations, characterized by the frequencies

$$\omega_V^2 \approx \omega_0^2 + \left\{\frac{1}{2m}\left[k_{\parallel}^2 + \left(\frac{n\pi}{d}\right)^2\right]\right\}^2, \quad n = 1, 2, \ldots, \qquad (28)$$

where odd n corresponds to normal excitations and the even n corresponds to tangential excitations.

Surface plasmons with high momenta $k_{||}$ (in contrast with plasmons with small $k_{||}$) are damped much more rapidly as a function of distance from the boundary into the surrounding medium in the direction into the plasma. The plasmon, which represents a single completely defined ensemble of electron excitations and the electromagnetic field, consists primarily of electron excitations at high $k_{||}$. Because of the boundary conditions that have been used these electron excitations are localized within the film; however, the localization of the plasmon outside the film is due only to the electromagnetic field, and the electromagnetic contribution to the plasmon is relatively small.

3. STOPPING POWER OF THE PLASMA LAYER

We first consider losses due to collective excitations (transfer of small momenta to the medium) i.e., $k_{||}v_T \ll \omega_0$. Under these conditions the total loss is not very sensitive to the spatial dispersion of the dielectric constant; neglecting spatial dispersion we have from (16)

$$W = -\frac{2e^2}{(1-e^{-\omega\beta})\mathbf{k}^2}\,\text{Im}\left\{\left[d - \frac{2k_{\parallel}}{k^2}\left(\sin^2\frac{k_\perp d}{2}\,\text{cth}\,\frac{k_{\parallel}d}{2} + \cos^2\frac{k_\perp d}{2}\,\text{th}\,\frac{k_{\parallel}d}{2}\right)\right]\frac{1}{\varepsilon(\omega)} + \frac{8k_{\parallel}}{k^2(1-e^{-2k_{\parallel}d})} \times\left(\frac{\sin^2(k_\perp d/2)}{\varepsilon(\omega) + \text{th}(k_{\parallel}d/2)} + \frac{\cos^2(k_\perp d/2)}{\varepsilon(\omega) + \text{cth}(k_{\parallel}d/2)}\right)\right\}. \qquad (29)$$

It is evident from (29) that the edge effect tends to reduce the loss due to the excitation of volume plasmons.[4,19] (The temperature factor is neglected for reasons of simplicity.)

Let us now consider losses due to binary collisions. Substituting (25) in (16) and omitting the intermediate calculations we have

$$W \approx \frac{4e^2\omega_0^2\pi k_\perp^2}{m\omega^2 d}\,\frac{\sin^2[(k_\perp d - n\pi)/2]}{[k_\perp^2 - (n\pi/d)^2]^2}\,\delta(\omega - \omega_V) + \frac{2e^2\omega_0^4 m k_{\parallel}}{\omega^2[k_\perp^2 + (\omega_0/\omega)^4 k_{\parallel}^2/16]}\,\delta(\omega - \omega_s). \qquad (30)$$

It is apparent from (30) that terms describing the intersection of the volume excitations with the incoming electron contain the factor

$$\frac{2\sin^2[(k_\perp d - n\pi)/2]}{\pi d(k_\perp - n\pi/d)^2} \xrightarrow[d\to\infty]{} \delta\left(k_\perp - \frac{n\pi}{d}\right). \qquad (31)$$

This factor results from the fact that momentum is only conserved approximately in a system of finite dimensions (the conservation improves as the dimensions of the system get larger). The particle passing through the film transfers part of its momentum to the volume excitation and part to the dividing surface. The situation is reminiscent of energy conservation when an interaction is switched on and off.

Let us introduce the quantity

*th = tanh.

†cth = coth.

$$\Delta E = \frac{1}{v} \int (\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p-k}}) W_{\mathbf{p}\to\mathbf{p}'} \frac{d^3k}{(2\pi)^3}, \quad (32)$$

which defines the mean energy lost by the particle passing through the film and consider separately the loss to collective volume excitations (ΔE_1), the individual volume excitations (ΔE_2), the collective surface excitations (ΔE_3) and the individual surface excitation (ΔE_4).

Because of the exponential nature of Landau damping the transition from collective volume excitations to individual excitations is rather sharp. Let the vector corresponding to this boundary be

$$k = k_m. \quad (33)$$

Then, using (29), (30), and (32) and assuming that the imaginary part of the dielectric constant is infinitesimally small, for normal incidence we have

$$\Delta E_1 \approx \frac{e^2\omega_0^2 d}{4\pi v^2} \left[\ln \frac{k_m v}{\omega_0} - \sin^2\alpha \int_0^\beta \frac{x^2 \operatorname{cth} x dx}{(x^2+\alpha^2)^2} - \cos^2\alpha \int_0^\beta \frac{x^2 \operatorname{th} x dx}{(x^2+\alpha^2)^2} \right]; \quad (34)$$

$$\alpha = \omega_0 d/2v, \qquad \beta = [k_m^2 - (\omega_0/v)^2]^{1/2} d/2, \quad (35)$$

$$\Delta E_2 \approx \frac{4\pi e^2 \omega_0^2}{mvd} \int_{k>k_m} \frac{d^3k}{(2\pi)^3} \frac{k_\perp^2 \sin^2[(k_\perp d - n\pi)/2]}{\omega[k_\perp^2 - (n\pi/d)^2]^2}. \quad (36)$$

The relation (34) coincides with the corresponding expressions obtained by other authors.[4,19]

The situation is different for the surface excitations. It is evident from (22) (cf.[13]) that the damping of the surface plasmons is a smooth function of the wave vector so that the transition from collective excitations to single particle excitations is a smooth one. In this case the integration over k cannot be divided into two parts, one corresponding to collective effects and the other to the single particle excitations. Evidently, in computing both ΔE_3 and ΔE_4 the integration over k can extend over the entire region. However, in both cases there is an error of $\sim (\hbar\omega_0/mv^2) \ll 1$. Thus the losses to excitation of normal plasmons are

$$\Delta E_{3n} \approx \frac{e^2\omega_0^2 d}{2\pi v^2} \left\{ \frac{\sin^2\alpha}{4\alpha^2(1+4\alpha^2)} + \sin^2\frac{\alpha}{\sqrt{2}} \times \left[\frac{1}{2+4\alpha^2} + \frac{\sqrt{2}}{4\alpha}\left(\frac{\pi}{2} - \operatorname{arc\,tg}\frac{1}{\sqrt{2}\alpha} \right) \right] \right\}, \quad (37)^*$$

while the losses to the excitation of tangential plasmons

$$\Delta E_{3\tau} \approx \frac{e^2\omega_0^2 d}{2\pi v^2} \left\{ \int_0^1 \frac{x^3 \cos^2(\alpha x/2)}{x^2+\alpha^2} dx + \cos^2\frac{\alpha}{\sqrt{2}} \times \left[\frac{1}{2+4\alpha^2} + \frac{\sqrt{2}}{4\alpha}\left(\frac{\pi}{2} - \operatorname{arc\,tg}\frac{1}{\sqrt{2}\alpha} \right) \right] \right\}. \quad (38)$$

Finally,

$$\Delta E_4 \approx \frac{2e^2 m\omega_0^4}{v} \int \frac{k_\parallel \delta(\omega-\omega_s)}{\omega k^4 [k_\perp^2 + k_\parallel^2 (\omega_0/2\omega)^4]} \frac{d^3k}{(2\pi)^3}. \quad (39)$$

Now let us consider thick films

$$\omega_0 d/v \gg 1. \quad (40)$$

The relations (34)—(39) now yield

$$\Delta E_1 \approx \frac{e^2\omega_0^2 d}{4\pi v^2} \ln\frac{k_m v}{\omega_0} - \frac{e^2\omega_0}{8v},$$

$$\Delta E_2 \approx \frac{e^2\omega_0^2 d}{4\pi v^2} \ln\frac{2mMv}{(m+M)k_m},$$

$$\Delta E_3 \approx \frac{e^2\omega_0}{4\sqrt{2}v}, \qquad \Delta E_4 \approx \frac{2^{3/5} e^2 \omega_0^{6/5} \operatorname{cosec}(0{,}3\pi)}{10\pi v^{7/5} m^{1/5} (1+m/M)^{3/5}}. \quad (41)$$

The total losses to volume excitations are thus

$$\Delta E_V = \Delta E_1 + \Delta E_2 \approx \frac{e^2\omega_0^2 d}{4\pi v^2} \ln\frac{2mMv^2}{(m+M)\omega_0} - \frac{e^2\omega_0}{8v}. \quad (42)$$

The first term in (42), which does not take account of edge effects, coincides with the result obtained by Larkin[3] by means of a more complicated calculation.

For the surface excitations we have from (41)

$$\frac{\Delta E_4}{\Delta E_3} \sim \frac{1}{3}\left(\frac{\omega_0}{mv^2} \right)^{1/5} < 1, \quad (43)$$

that is to say, the surface losses are due primarily to collective excitations. Obviously this statement holds only within the limits of the model used in Sec. 2; it does not take account of the excitations of absorbed states and the other surface states considered there.

Now let us consider thin films

$$\omega_0 d/v \ll 1, \qquad k_m d \gg 1. \quad (44)$$

To logarithmic accuracy, we have from (34)

$$\Delta E_1 \approx \frac{e^2\omega_0^2 d}{4\pi v^2} \ln\frac{k_m d}{7}. \quad (45)$$

For still thinner films

$$k_m d \ll 1 \quad (46)$$

the relation in (34) yields

$$\Delta E_1 \approx \frac{e^2\omega_0^2 d}{4\pi v^2} \sin^2\alpha \ln\frac{k_m v}{\omega_0}. \quad (47)$$

*arctg = $\tan^{-1}$.

It is evident from (45) and (47) [2)] that the losses to volume excitations per unit thickness vanish in a very thin film.

This effect, due to the plasma boundaries, was first pointed out by Ritchie[4] and by Faĭnberg and Khizhnyak.[19] We shall not take account of losses at the absorption poles which, as shown in[19], are independent of film thickness in the linear approximation. Losses per unit thickness described by ΔE_2 are also reduced as the thickness is reduced. However, this reduction appears at very small thicknesses, in which case the model used here no longer applies.

For the losses due to surface excitations using (37) and (38) for the case given by (44) we have

$$\Delta E_{3n} \approx \frac{e^2}{2\pi d}\sin^2\alpha, \qquad \Delta E_{3\tau} \approx \frac{e^2\omega_0^2 d}{2\pi v^2}\cos^2\frac{\alpha}{\sqrt{2}}. \quad (48)$$

The last relations show that the total losses to the excitation of normal and tangential surface plasmons are of the same order. It should be noted, however, that the distribution of loss over energy is considerably different in the two cases. Specifically, as is evident from (22) the losses due to normal plasmons in thin films exhibit a resonance effect near ω_0 whereas the losses due to tangential plasmons, which are characterized by a continuous spectrum, extend from 0 to $\omega_0/\sqrt{2}$, smearing out the resonance effect near ω_0. Furthermore, the losses per unit thickness due to surface plasmons in very thin films ($\alpha \ll 1$) are independent of thickness and consequently appear as the basic collective losses. In this respect they are analogous to the losses at absorption poles[20] and differ from the losses to volume excitations, which are density dependent. This dependence arises because the losses at frequencies corresponding to zero dielectric constant occur only over the path that lies within the slab whereas the particle losses at the absorption poles and at frequencies of the surface plasmons are primarily outside the slab.

The losses per unit thickness described by ΔE_4 and those described by ΔE_2 are weak functions of thickness and, within the limits of applicability of the model being used here, are described essentially by (41).

At the present time surface plasmons have not been investigated to any great extent, either theoretically or experimentally. For this reason we wish to list the basic features of the losses due to surface plasmons: 1) The absence of a density effect. 2) A more pronounced (compared with volume plasmons) angular dependence of the scattering probability of incoming electrons [cf. (29)]. 3) A linear dependence of energy loss on scattering angle, in contrast with the quadratic dependence for volume plasmons. This dependence arises as a result of spatial dispersion and is determined by (24) in the case of thick films. This dependence changes as the thickness is reduced. 4) The width of the absorption line (in a collisionless plasma) increases linearly with ϑ for thick films [cf. (22)].

A number of additional surface-plasmon effects arise in case of oblique incidence of the fast particle; these also have certain characteristic features, but will not be discussed here.

4. FREQUENCY OF THE PLASMA OSCILLATIONS AS A FUNCTION OF THICKNESS IN THIN FILMS

We have seen above that electrons passing through a plasma lose part of their energy to the excitation of volume plasmons; this process is characterized by a dispersion relation of the form $\epsilon(\mathbf{k}, \omega) = 0$, or

$$\omega^2 \approx \omega_0^2 + k^2\overline{v^2}, \quad (49)$$

where $\overline{v}$ is the mean square velocity of the plasma electrons. In films $k_{min} \sim \pi/d$ and (49) yields

$$\omega_{min}^2(d) \approx \omega_0^2 + (\pi/d)^2\overline{v^2}. \quad (50)$$

The dependence of plasma oscillation frequency on film thickness, which appears when $d < 50$ Å, has been pointed out by Ichikawa.[14] For Al, for example, the contribution due to the second term in (5) amounts to $\sim(38/d^2)$ eV where d is given in Angstroms. The experimental data given in[14], however, indicate that the thickness effect appears much earlier. For example, for Al ($\omega_0 \approx 15$ eV) with $d = 100$ Å

$$\Delta\omega(d) = \omega_{min}(d) - \omega_0 \approx 0.1 \text{ eV}, \quad (51)$$

which clearly disagrees with (49). This rather strong dependence of plasma frequency on thickness can be attributed to the quantum nature of electrons in thin films, specifically to the discreteness of the electron momentum. We shall see below that this discreteness is much more important than the discreteness of the plasma wave vector.

The semiclassical approximation used in the preceding sections is not suitable for solving the problem at hand. For this reason we return to (10), writing this relation for a plasmon wave function $\varphi(x)$

2) It should be noted that (47) is only qualitative since the basic contribution in ΔE_1 in this case comes from plasmons with $k \sim k_m$ for which it is necessary to take account of spatial dispersion and the quantum nature of the electrons, both of which are neglected in the present section.

$$\Delta\varphi(x) - \int \Pi(x, x')\varphi(x')d^4x' = 0. \qquad (52)$$

To find $\Pi(x, x')$ we must at least know the free Green's functions of the plasma electron components. The determination of these functions in actual crystals is an extremely complicated problem. Since we are only interested in determining the effect of the discreteness of the electron momenta on the plasma spectrum we shall consider a much simpler model. Specifically, we regard the plasma slab as a one-dimensional potential well with impenetrable walls. We neglect surface electrons, whose effect on the volume plasmon spectrum is insignificant, in view of their relative smallness.

In the model we have chosen the wave functions of the electrons are $\sim \sin p_3 x_3$ where $p_3 = \nu\pi/d$, $\nu = 1, 2, \ldots$ and the free Green's function for the electrons; consequently the polarization operator can be determined easily. Furthermore, substituting the following expansion in (52)

$$\varphi(x) = \sum_{k_\parallel k_3} \varphi(k_\parallel, k_3)\cos k_3 x_3\, e^{i(k_\parallel x - \omega t)}, \qquad (53)$$

we obtain the dispersion relation for the volume plasmons:

$$k^2 + \frac{e^2}{d}\sum_{p_3}\int \frac{dp_\parallel}{(2\pi)^2}\frac{(\varepsilon_p - \varepsilon_{p-k})(n_p - n_{p-k})}{\omega^2 - (\varepsilon_p - \varepsilon_{p-k})^2}\varphi(\mathbf{k})$$

$$= \frac{e^2}{d}\sum_{p_3}\int \frac{dp_\parallel}{(2\pi)^2}\frac{(\varepsilon_p - \varepsilon_{p-k})(n_p - n_{p-k})}{\omega^2 - (\varepsilon_p - \varepsilon_{p-k})^2}\varphi(\mathbf{k}_\parallel\, |2p_3 - k_3|). \qquad (54)$$

The summation is taken over both positive and negative values of p_3.

When $d \to \infty$ the right side of (54) vanishes and we obtain the usual dispersion relation for volume plasmons in an infinite plasma. For finite d we regard the right side as a perturbation, noting that all of the terms are of equal value. The $p_3 = k_3$ term contains a factor $\sim (p_F d)^{-1}$ (p_F is the Fermi momentum); the remaining terms contain this factor plus an additional factor due to the fact that the plasmon "shape" is not harmonic. It is evident that the latter is also $\sim (p_F d)^{-1}$.

Thus, limiting ourselves to terms containing the first order ratio of the electron Fermi wavelength to the film thickness, we have

$$k^2 + \frac{e^2}{d}\sum_{p_3 \neq k_3}\int \frac{dp_\parallel}{(2\pi)^2}\frac{(\varepsilon_p - \varepsilon_{p-k})(n_p - n_{p-k})}{\omega^2 - (\varepsilon_p - \varepsilon_{p-k})^2} = 0. \qquad (55)$$

The basic difference between the dispersion relation in (55) and the usual relation for an infinite plasma (aside from the fact that the summation becomes an integration) is the absence of terms with $p_3 = 0$, k_3. This is a direct consequence of the application of the uncertainty relation to the electron momentum and coordinate, which yields

$$p_{min} \sim \pi / d, \qquad (56)$$

and this is presumably a universal result not associated with the choice of the electron wave functions in the film. (A $p_3 = k_3$ term would correspond to electron transitions from a $p_3 = 0$ state to a $p_3 = k_3$ state and vice versa; by virtue of (56) these must be eliminated as must the $p_3 = 0$ term.)

Solving Eq. (55) in the usual approximation ($k^2\overline{v^2} \ll \omega^2$) we have

$$\omega^2 \approx \omega_0^2\left[1 + \frac{1}{4d}\left(\frac{9\pi}{n}\right)^{1/3}\right] + a^2k^2, \qquad (57)$$

where $a^2 \sim \overline{v^2}$, and n is the plasma electron density. If the film is not too thin the dependence of frequency on thickness is thus given essentially by

$$\frac{\Delta\omega(d)}{\omega_0} \approx \frac{1}{8d}\left(\frac{9\pi}{n}\right)^{1/3}. \qquad (58)$$

For Al with d = 100 Å (58) gives $\sim 1/150$ in agreement with (51). Thus the basic dependence of the plasmon spectrum on thickness can be explained completely by the fact that the electron momenta must assume discrete values.

An analogous dependence of the energy spectrum on thickness is found for surface plasmons. However, this dependence is completely masked by the stronger dependence on the shape of the plasma surface and has not been observed experimentally.[8,9]

Our dispersion relation (57) is in good agreement with experiment and serves to verify the collective nature of the characteristic losses in thin films since the indicated dependence of loss energy on thickness cannot be explained by interzone transitions of individual electrons. The latter effect, due to the broadening of the forbidden zones, also connected with (56), is given by

$$\Delta E \sim \pi^2 / md^2, \qquad (59)$$

which is small since it is quadratic in 1/d.

5. SURFACE PLASMON CHARACTERISTICS IN A PLASMA WITH DIFFUSE BOUNDARIES

Up to this point we have assumed that the plasma boundary is sharp. Actually, however, the boundary between two media will always be diffuse to some extent. The diffuseness of this boundary is especially important for surface plasmons, because they are localized near the boundary. The important point is that when one assumes a sharp boundary, for example, a vacuum-plasma boundary, the electromagnetic field of a surface plasmon with wave vector parallel to the separation surface $k_\parallel$ and frequency $\sim\omega_0/\sqrt{2}$ (ω_0 is the Langmuir frequency) is damped exponentially on going away from the boundary, falling off as $\sim \exp(-k_{\parallel}|x_3|)$. In other words, the surface plasmon is localized near the boundary (within a layer of thickness $\sim k_{\parallel}^{-1}$). Now let us consider the case of a diffuse boundary characterized by the dimension L. If L is such that $k_{\parallel}L \gg 1$ it is clear from general considerations that the surface plasmon must either vanish altogether or must be considerably different from that which is obtained in the case of a plasma with a sharp boundary. Furthermore, since the surface plasmon is characterized by $\epsilon(\omega) = -1$, in a diffuse vacuum-plasma transition, for a given frequency there always exists a spatial region in which $\epsilon(\omega, x_3) = 0$, in which a volume plasmon can exist. The latter circumstance means that in this region part of the energy of the surface plasmon is converted into energy of the volume plasmon; this is due to the same specific damping mechanism of the surface plasmon in a plasma with smeared-out boundaries. This damping is stronger the larger the region in which the volume plasmons exist, i.e., the smoother the transition region.

Proceeding now to a quantitative discussion, we confine ourselves to a semi-infinite plasma with a linear electron density variation

$$n(x_3) = \begin{cases} 0 & x_3 < 0 \\ n_0 + (N - n_0)(1 - x_3/L) & 0 < x_3 < L. \\ n_0 & x_3 > L \end{cases} \quad (60)$$

The dielectric constant is

$$\varepsilon(\omega,\ x_3) = 1 - \omega_0^2(x_3) / \omega(\omega + i\nu), \quad (61)$$

$$\omega_0^2(x_3) = 4\pi n(x_3) e^2 / m, \quad (62)$$

and in the transition region we can write

$$\varepsilon(\omega,\ x_3) = a - x_3 / x_0, \quad (63)$$

$$a = 1 - \frac{4\pi Ne^2}{m\omega(\omega + i\nu)}, \quad x_0 = \frac{\omega(\omega + i\nu)m}{4\pi(n_0 - N)e^2}L. \quad (64)$$

To find the energy spectrum we start with the Laplace equation for the scalar potential using the appropriate boundary conditions.[3)] The solution is written in the form

$$\varphi(x,\ t) = e^{i(\mathbf{k}_{\parallel}\mathbf{x} - \omega t)}\varphi(x_3). \quad (65)$$

Introducing the new variable

$$\zeta = x_0\varepsilon(\omega,\ x_3) = ax_0 - x_3, \quad (66)$$

we obtain in the transition region an equation

$$\frac{\partial^2\varphi(\zeta)}{\partial\zeta^2} + \frac{1}{\zeta}\frac{\partial\varphi(\zeta)}{\partial\zeta} - k_{\parallel}^2\varphi(\zeta) = 0, \quad (67)$$

the solution of which is

$$\varphi(\zeta) = AK_0(k_{\parallel}\zeta) + BI_0(k_{\parallel}\zeta), \quad (68)$$

where K_0 and I_0 are Bessel functions of imaginary argument.

A field outside the transition region which vanishes at $\pm\infty$, is defined by a potential

$$\varphi(x_3) = \begin{cases} e^{k_{\parallel}x_3} & x_3 < 0 \\ Ce^{-k_{\parallel}x_3} & x_3 > L \end{cases}. \quad (69)$$

By joining the solutions at the points $x_3 = 0$ and L we find the constants of integration and the dispersion relation. This relation is

$$\frac{\varepsilon(x_3 = 0)I_1(\delta) + I_0(\delta)}{\varepsilon(x_3 = 0)K_1(\delta) - K_0(\delta)} = \frac{I_1(\delta') - I_0(\delta')}{K_1(\delta') + K_0(\delta')}, \quad (70)$$

$$\delta = ax_0k_{\parallel}, \qquad \delta' = (ax_0 - L)k_{\parallel}. \quad (71)$$

Let us consider two particular cases.

1. N = 0. In this case we have from (70)

$$\omega \approx \frac{\omega_p(L)}{\sqrt{2}}\left(1 - i\pi k_{\parallel} \Big/ 4\left(\frac{\partial\varepsilon}{\partial x_3}\right)_{\varepsilon(\omega,\ x_3) = 0} - i\frac{\nu}{2}\right). \quad (72)$$

When $k_{\parallel}L \gg 1$ (70) does not have a solution and this indicates the disappearance of the surface plasmons, as expected.

The most interesting feature of the dispersion relation (72) is the appearance of damping even when $\nu \to 0$, i.e., in the absence of dissipation. This damping, as indicated above, is due to the conversion of the energy of the surface plasmon into the energy of the volume plasmon in the region $\epsilon(\omega, x_3) = 0$. We neglect this damping temporarily. Then the width of the resonance line $\Delta\omega = \nu$ and the width of the region in which $\epsilon(\omega, x_3) = 0$ is $\Delta x_3 = \nu L/\omega$. However, the electromagnetic field in this region $E_{x_3}(\zeta) \approx k_{\parallel}L/2\zeta \approx k_{\parallel}\omega/\nu$. When $\nu \to 0$, $\Delta x_3 \to 0$ but $E_{x_3} \to \infty$. Hence the heat generated primarily in the region $x_3 = L/2$ is finite and given by $Q \approx \pi\omega k_{\parallel}L/4$ regardless of ν. It is important to emphasize that in the limit $\nu \to 0$ this heat is only generated in the region $x_3 = L/2$ i.e., in the region in which the volume plasmon exists. (Actually, because of the specific damping mechanism this region expands to $\Delta x_3 \approx \pi k_{\parallel}L^2/8$.)

The damping of the surface plasmon due to conversion is of the form

$$\gamma = Q / 2W \approx {}^1\!/_8\pi\omega k_{\parallel}L, \quad (73)$$

which coincides with (72) and verifies the statement above. The indicated damping mechanism is of interest in that it can be used for generating energy at high density in a small volume through excita-

[3)] In the interest of simplicity we neglect spatial dispersion, although it could actually become important, especially when $\nu \to 0$.

tion of surface plasmons by whatever method is feasible for example, incident electrons.

2. $N \gg n_0$. In this case (70) has the solution

$$\omega(\omega + i\nu) \approx \begin{cases} {}^1/_2\, \omega_p^2 (L)(1 + Nk_\parallel L/2n_0), & Nk_\parallel L/n_0 \ll 1 \\ {}^1/_2\, \omega_p^2 (0)(1 + O(1/k_\parallel L)), & k_\parallel L \gg 1 \end{cases}. \quad (74)$$

There is no specific damping in this case because of the absence of a region $\epsilon(\omega, x_3) = 0$ for these frequencies. The entire diffuseness effect reduces to deformation of the dispersion curve of the surface plasmons.

The surface-plasmon features considered here are not important in metals, (since L is small) but can be important in gas plasmas or semiconductor plasmas, particularly when there are pn junctions in the latter.

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311