The kinetic energy correction is investigated within perturbation theory. We observe that $-\nabla_v^2$ is of the order $1/r_v^2$; i.e., it is of the same size as the perturbing Hamiltonian in (25). If we expand the determinantal function Φ, we find that

$$\int \Phi^*(-\nabla_v^2\Phi)d\mathbf{r}_c=\sum_i \int u_i^{(0)*}(\mathbf{r}_1)(-\nabla_v^2 u_i^{(1)}(\mathbf{r}_1,\mathbf{r}_v))d\mathbf{r}_1 + O(1/r_v^6). \quad (35)$$

The first term is of the order $1/r_v^4$ since $u_i^{(1)}$ is proportional to $1/r_v^2$. Terms of the order $1/r_v^6$ and higher are neglected here, but would have to be included in an evaluation of quadrupole terms. The $1/r_v^4$ term in (35) appears to be of the same order as V_p. Upon substitution of the perturbed wave functions $u_i^{(1)}$ into (35), it is found that the integral vanishes upon integration over solid angle. Consequently there is no contribution from the kinetic energy correction of the same order in r_v as the dipole polarization potential.

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Plasma Losses by Fast Electrons in Thin Films*

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The angle-energy distribution of a fast electron losing energy to the conduction electrons in a thick metallic foil has been derived assuming that the conduction electrons constitute a Fermi-Dirac gas and that the fast electron undergoes only small fractional energy and momentum changes. This distribution exhibits both collective interaction characteristics and individual interaction characteristics, and is more general than the result obtained by other workers. Describing the conduction electrons by the hydrodynamical equations of Bloch, it has been shown that for very thin idealized foils energy loss may occur at a value which is less than the plasma energy, while as the foil thickness decreases below $\sim v/\omega_p$ the loss at the plasma energy becomes less than that predicted by more conventional theories. The net result is an increase in the energy loss per unit thickness as the foil thickness is decreased. It is suggested that the predicted loss at subplasma energies may correspond to some of the low-lying energy losses which have been observed by experimenters using thin foils.

I. INTRODUCTION

THERE has been recently a rather extraordinary amount of experimental and theoretical work on the origin and implications of characteristic energy losses experienced by fast electrons in passing through foils. This effort has received great impetus from the suggestion by Pines and Bohm[1] that some of these energy losses are due to the excitation of plasma oscillations or "plasmons" in the sea of conduction electrons and from their work on the theory of these oscillations.[2] An alternate explanation, which has been advanced many times by various workers, is that these losses are due to interband transitions of individual conduction electrons. Evidence in support of this has been presented[3] showing correlation between the fine structure of x-ray absorption edges and the characteristic loss lines. The plasma interpretation has been strengthened by Watanabe's[4] experimental verification of the Pines-Bohm plasma dispersion relation [Eq. (12) below] in Be, Al, Mg, and Ge. A critical review of the present status of the theory and experiment in this field has been given by Pines.[5]

It is the purpose of this paper to examine theoretically the energy and angular distribution of a fast electron which has lost energy to plasma oscillations in an infinite foil and to consider the effect of the finiteness of the foil. Ferrell[6] has investigated the angular dependence of the characteristic energy losses of fast electrons to an infinite plasma using the theory of Pines and Bohm.[2] He obtains one formula which involves the collective interaction of conduction electrons with the incident electrons and another which includes only the effect of individual interactions between conduction electrons and the incident electrons. A single formula will be derived which includes both collective and individual interactions and which depends upon the momentum distribution of the undisturbed plasma.

Gabor[7] has considered the interaction of a fast electron with a small metallic crystal containing free electrons. He assumes that the electric field is always zero at the surface of the crystal and examines the

* This paper represents a portion of a dissertation submitted by the author to the University of Tennessee in partial fulfillment of the requirements for the Ph.D. degree.
[1] D. Pines and D. Bohm, Phys. Rev. **85**, 338 (1952).
[2] D. Pines and D. Bohm, Phys. Rev. **82**, 625 (1951); **85**, 338 (1952); **92**, 609, 626 (1953).
[3] Leder, Mendlowitz, and Marton, Phys. Rev. **101**, 1460 (1956).
[4] H. Watanabe, J. Phys. Soc. Japan **11**, 112 (1956).
[5] D. Pines, in *Solid State Physics* (Academic Press, Inc., New York, 1955). See also D. Pines, Revs. Modern Phys. **28**, 184 (1956).
[6] R. A. Ferrell, Phys. Rev. **101**, 554 (1956).
[7] D. Gabor, Phil. Mag. **1**, 1 (1956).

probability of interaction in the crystal as it becomes very thin in the direction of the electron beam. It will be shown that his boundary condition leads to unrealistic results. Applying a more realistic boundary condition, the interaction probability in very thin films is derived and the consequences of the results are discussed.

II. DIELECTRIC TREATMENT OF AN ELECTRON GAS

In the following the interaction between an incident fast electron and metal electrons will be considered. The view will be taken that the ensemble of conduction electrons in a metal may be characterized by a dielectric constant which is a function both of the frequency and wave vector of the electromagnetic disturbance in the metal. As long as one considers interelectronic action over distances large compared with the interelectronic spacing, the metal may be treated as though it is a continuous homogeneous medium and can hence be described in terms of its dielectric properties.

The dielectric approach seems to have been first used by Fermi[8] to calculate the stopping power of matter for fast charged particles. Kramers[9] used a similar consideration to calculate specifically the stopping power due to conduction electrons. Both of these authors considered that the dielectric constant is a function of frequency only. A classical theory of the dielectric constant of an assembly of free electrons has been given[10,11] which includes the motion of the undisturbed electrons and results in a dependence of the dielectric constant $\epsilon(\mathbf{k},\omega)$ on the wave vector $\mathbf{k}$ of the disturbance. Lindhard[10] and Hubbard[12] have derived the dielectric constant of conduction electrons by means of quantum perturbation theory. In these treatments it is assumed that there exists a common electric field[13] $\varphi(\mathbf{r},t)$ in space and time in which the separate electrons move and to which they give rise. The development of the wave functions of the electrons is calculated by time-dependent perturbation theory, taking into account that they do not develop independently in time because each electron moves in a field determined by all others.

Hubbard[12] has extended the dielectric treatment to the calculation of the energy-angle distribution of fast electrons in passing through and losing energy to solids, using a method similar to the Weizsäcker-Williams treatment of scattering. The fast electron is represented as an appropriate charge distribution $\rho(\mathbf{r},t) = -e\delta(\mathbf{r}-\mathbf{v}t)$. It is well known[14] that a fast electron which interacts with matter may be considered to be a point charge with a well-defined path as long as one is concerned with processes in which the fractional changes of energy and momentum of the electron are small. The field which is generated is then given by Poisson's equation,

$$\epsilon(\mathbf{k},\omega)\nabla^2\varphi(\mathbf{r},t) = -4\pi\rho(\mathbf{r},t), \tag{1}$$

so that in Fourier space

$$\varphi(\mathbf{k},\omega) = 4\pi\rho(\mathbf{k},\omega)/k^2\epsilon(\mathbf{k},\omega), \tag{2}$$

where $\varphi(\mathbf{k},\omega)$ and $\rho(\mathbf{k},\omega)$ are the four-dimensional Fourier transforms of $\varphi(\mathbf{r},t)$ and $\rho(\mathbf{r},t)$. The field φ is regarded as a perturbation acting on the electrons of the metal. This perturbation causes transitions from occupied to unoccupied levels. An electron making such a transition will acquire $\hbar\omega$ and momentum $\hbar\mathbf{k}$ and the fast electron must suffer a corresponding loss. The probability for absorption of energy $\hbar\omega$ and momentum $\hbar\mathbf{k}$ per unit path length is

$$P(\mathbf{k},\omega) = \frac{1}{\hbar\omega}W(\mathbf{k},\omega),$$

where $W(\mathbf{k},\omega)$ is the energy absorbed per unit volume in $\mathbf{k}$ space per unit frequency interval and per unit path length of the fast electron. Now if one is able to express the energy loss dW/dx per unit path length by the fast electron to the medium as an integral over $\mathbf{k}$, ω space, then the integrand is identically $W(\mathbf{k},\omega)$ and the interaction probability $P(\mathbf{k},\omega)$ is known.

Hubbard considered the energy-angle distribution of fast electrons in plasma but obtained essentially Ferrell's[6] result since he used a simplified form of the dielectric constant. In the present work the approach of Lindhard and Hubbard will be extended to yield a more detailed description of the angle-energy distribution of fast electrons losing energy to plasma.

III. DISTRIBUTION IN ENERGY AND ANGLE OF FAST ELECTRONS IN AN INFINITE PLASMA

The equation for the electric potential due to a point charge, $(-e)$, moving with uniform velocity $\mathbf{v}$ in a uniform infinite plasma, is given by

$$\epsilon(\mathbf{k},\omega)\nabla^2\varphi(\mathbf{r},t) = 4\pi e\delta(\mathbf{r}-\mathbf{v}t), \tag{3}$$

so that in Fourier space

$$\varphi(\mathbf{k},\omega) = -\frac{8\pi^2 e}{\epsilon(\mathbf{k},\omega)}\frac{\delta(\mathbf{k}\cdot\mathbf{v}+\omega)}{k^2}, \tag{4}$$

where

$$\varphi(\mathbf{r},t) = \frac{1}{(2\pi)^4}\int d\mathbf{k}\int d\omega \exp[i(\mathbf{k}\cdot\mathbf{r}+\omega t)]\varphi(\mathbf{k},\omega). \tag{5}$$

[8] E. Fermi, Phys. Rev. **57**, 485 (1940).
[9] H. A. Kramers, Physica **13**, 401 (1947). See also A. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **24**, No. 19 (1948).
[10] J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **28**, No. 8 (1954).
[11] J. Neufeld and R. H. Ritchie, Phys. Rev. **98**, 1632 (1955).
[12] J. Hubbard, Proc. Phys. Soc. (London) **68**, 976 (1955).
[13] In the following it will be assumed that the gauge is chosen so that the vector potential is zero. Also, all velocities are assumed to be nonrelativistic.
[14] E. J. Williams, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **13**, No. 4 (1935).

Then the energy loss per unit path in the medium is given by

$$-\frac{dW}{dx}=eE_x|_{\mathbf{r}=\mathbf{v}t}=\frac{e}{v}\mathbf{v}\cdot\mathbf{E}|_{\mathbf{r}=\mathbf{v}t}, \tag{6}$$

where E_x is the x component of the electric field due to the medium alone. Expressing φ in terms of its Fourier representation

$$-\frac{dW}{dx}=\frac{e^2}{\pi^2 v}\int d\mathbf{k}\int_0^\infty d\omega\cdot\omega\,\mathrm{Im}\left(\frac{1}{\epsilon(\mathbf{k},\omega)}\right)\frac{\delta(\mathbf{k}\cdot\mathbf{v}+\omega)}{k^2}, \tag{7}$$

we may write immediately for the transition probability per unit path length

$$P(\mathbf{k},\omega)=\frac{e^2}{\pi^2\hbar v}\mathrm{Im}\left(\frac{1}{\epsilon}\right)\frac{\delta(\mathbf{k}\cdot\mathbf{v}+\omega)}{k^2}. \tag{8}$$

Now divide $\mathbf{k}$ into $\mathbf{k}_\perp$ and $\mathbf{k}_{\parallel}$, which are perpendicular and parallel, respectively, to $\mathbf{v}$. We may integrate over $k_{\parallel}$ immediately, obtaining

$$P(\mathbf{k}_\perp,\omega)=\frac{e^2}{\pi^2\hbar v^2}\mathrm{Im}\frac{1}{\epsilon}\frac{1}{(k_\perp^2+\omega^2/v^2)}.$$

We have now to obtain an approximate expression for $\epsilon(\mathbf{k},\omega)$. Lindhard[10] gives for the dielectric constant of a sea of free electrons

$$\epsilon(\mathbf{k},\omega)=1-\omega_p^2\sum_n\frac{f(E_n)}{N}\times\left\{\left(\omega-i\gamma+\frac{\hbar}{m}\mathbf{k}\cdot\mathbf{k}_n\right)^2-\frac{\hbar^2k^4}{4m^2}\right\}^{-1}, \tag{9}$$

where γ is essentially the damping constant of the excited states, ω_p is the plasma frequency, N is the density of electrons and $f(E_n)$ is the density of states having energy E_n in the undisturbed plasma.

First, we may expand the denominator of Eq. (9) assuming that ω is large compared with the other terms in the denominator. Then, assuming that the density of states is so great that the sum may be replaced by an integral, we finally obtain

$$\mathrm{Im}\left(\frac{1}{\epsilon}\right)=\frac{2\gamma\omega_p^2\omega}{\{\omega^2-\omega_p^2(1+\delta)\}^2+4\gamma^2\omega^2}, \tag{10}$$

where

$$\delta=\frac{1}{\omega^2}\frac{\hbar^2}{m^2}\left(\frac{k^4}{4}+\frac{3k^2k_0^2}{5}\right),$$

and k_0 is the maximum wave vector of the undisturbed plasma. We now substitute this into Eq. (8) and employ the approximation, good for ϑ small, that $k_\perp=\theta mv/\hbar$ where θ is the angular deviation of the fast electron and mv is its momentum. Then

$$P(\theta,\omega)d\Omega=\frac{\omega_p^2e^2\omega}{\pi^2\hbar v^2}2\gamma[\{\omega^2-\omega_p^2(1+\delta)\}^2+4\gamma^2\omega^2]^{-1}\times\frac{d\Omega}{[\theta^2+(\hbar\omega/mv^2)^2]}, \tag{11}$$

where $d\Omega$ is the element of solid angle around θ. The term in the square brackets has the character of a δ function if the damping constant γ is assumed to be vanishingly small. The resonance occurs when

$$\omega^2=\omega_p^2(1+\delta).$$

Solving this equation by iteration,

$$\omega^2=\omega_p^2+\frac{\hbar^2}{m^2}(k^4/4+3k_0^2k^2/5). \tag{12}$$

This is just the plasma dispersion relation obtained by Pines.[2] Equation (11) contains the major features of the angle-energy losses of fast electrons to free electron plasma. The resonance term yields explicitly the parabolic connection between energy loss and angular distribution found experimentally by Watanabe.[4] The factor in brackets contains the coulomb scattering loss form given by Ferrell.[6] Now to obtain the angular distribution of the fast electrons after losing energy to the plasma, we integrate over all energy losses, obtaining

$$P(\theta)d\Omega=\frac{e^2\omega_p^2}{2\pi\hbar v^2}\left[\omega_p^2+\frac{\hbar^2}{m^2}(k^4/4+3k_0^2k^2/5)\right]^{-\frac{1}{2}}\times\frac{d\Omega}{\theta^2+\frac{\hbar^2}{(2E)^2}\left(\omega_p^2+\frac{\hbar^2}{m^2}\left[\frac{k^4}{4}+\frac{3k_0^2k^2}{5}\right]\right)}, \tag{13}$$

where $k^2=\omega_p^2/v^2+(mv\theta/\hbar)^2$. Now, collecting terms and neglecting higher powers of $\hbar\omega_p$ and $\hbar^2k_0^2/2m$ compared with E, the energy of the incident electron, we find

$$P(\theta)d\Omega=\frac{e^2\omega_p^2}{2\pi\hbar v^2}[\omega_p^2+(6/5)\omega_0\omega_E\theta^2+\omega_E^2\theta^4]^{-\frac{1}{2}}\times\frac{d\Omega}{\theta^2+(\omega_p/2\omega_E)^2}, \tag{14}$$

where $\omega_0=\hbar k_0^2/2m$ and $\omega_E=E/\hbar$.

Now, if one lets $\omega_0\to 0$ and neglects the θ^4 term in this expression, one finds Ferrell's[6] Eq. (8) and Hubbard's[12] Eq. (29) for the angular distribution. Further, one sees that instead of $\epsilon(\mathbf{k},\omega)$ for the free electron gas, one could have used the semiclassical formula for the dielectric constant,

$$\epsilon(\omega)=1+\frac{4\pi Ne^2}{m}\sum_i\frac{f_i}{\omega_i^2+ig_i\omega-\omega^2}, \tag{15}$$

where N is the atomic density of the medium, and f_i, ω_i, and g_i are the oscillator strength, transition frequency and damping constant of the ith interband transition, respectively. Assuming that in the neighborhood of the jth transition only the jth term in this series is important, substituting this ϵ in Eq. (8) and integrating over $\mathbf{k}_{\parallel}$ and ω, one finds, for the probability of scattering through the angle θ, after losing energy to the jth transition,

$$P_j(\theta)d\Omega=\frac{e^2}{2\pi\hbar v^2}\frac{\alpha_j^2}{[\alpha_j^2+\omega_j^2]^{\frac{1}{2}}}\frac{d\Omega}{[\theta^2+(\omega_j^2+\alpha_j^2)/\omega_E^2]}, \quad (16)$$

where $\alpha_j^2=4\pi Ne^2f_j/m$. This again has the same form as Ferrell's Eq. (8) except that in place of the plasma frequency, there appears the one-electron transition frequency ω_j, shifted due to mutual interaction between electrons in the medium. If the frequency $\omega_j\ll\alpha_j$ corresponding to the case of free electrons, then $\alpha_j=\omega_p^2$. Thus if the $\mathbf{k}$ dependence of the dielectric constant of the conduction electrons is neglected, one obtains the same form for the angular distribution, whether the fast electron has excited interband transitions in the solid or whether it has lost energy to plasma oscillations. However, the $\mathbf{k}$ dependence introduces a factor which, although slowly varying, may be verified experimentally.

To examine the origin of the θ^4 term in the expression for $P(\theta)$, suppose that n, the density of conduction electrons, is low so that ω_p and ω_0 are very small. Then

$$P(\theta)d\Omega\rightarrow\frac{4ne^4}{m^2v^4}\frac{d\Omega}{\theta^4}, \quad (17)$$

which is just the small angle approximation to the Rutherford scattering of electrons on free electrons. Thus, the formula (14) displays both collective and individual interaction characteristics. It shows that when the scattering angles are very small the collective behavior of the ensemble of electrons determines the angular distribution. For larger angles or for very low electron densities, individual interaction between the fast electron and the electrons in the medium becomes the determining effect. Even though the quantum dielectric formulation is valid for values of the wave vector which are not too large, it seems somewhat more general than the procedure of Ferrell who used first-order perturbation theory and the cut-off wave vector approach of Pines and Bohm. Thus, he obtains separate formulas valid in the ranges $k<k_c$ and $k>k_c$ where k_c is the cutoff wave vector. The collective treatment was used when $k<k_c$ and individual interaction was assumed for $k>k_c$. The dielectric treatment yields a result which bridges the gap to a certain extent, even though it can be shown that it is also equivalent to first-order perturbation theory.[10, 12]

IV. DISTRIBUTION OF FAST ELECTRONS PASSING THROUGH THIN FOILS

To treat a finite foil, we shall proceed in much the same way as above. The potential throughout space due to a point charge $-e$ moving with velocity $\mathbf{v}$ will be calculated, including the modifying effect of the foil. The energy loss in the foil will be expressed in terms of this potential and as an integral over $\mathbf{k}$ and ω. The integrand again will be interpreted as $\hbar\omega$ times the probability that the electron will lose energy $\hbar\omega$ and momentum $\hbar\mathbf{k}$ in passing through the foil.

Gabor[7] has treated the thin foil problem by making the assumption that the electric field intensity is zero at the boundaries of the foil. This is not in accord with experimental work on the optics of thin films, since it results in the prediction that a metal grain does not interact with electromagnetic radiation originating outside of the grain. His conclusion that the probability for plasma loss should decrease strongly with decreasing foil thickness is a direct consequence of this assumption. The difference which he finds between "coherent" and "incoherent" illumination is due to the same assumption.

To compare with Gabor's treatment, one may expand the field given by Eq. (3) in terms of a Fourier integral in y, z, and t and as a Fourier series involving only cosine functions of the x coordinate, thereby assuming that the electric field intensity is zero both at the boundaries and everywhere outside the foil. Then, using the method described above to calculate the interaction probability $P(\mathbf{k},\omega)$ in the foil, one finds a factor which shows the same limitation obtained by Gabor on x-momentum transfer to the foil. In reality the field is not zero at the surfaces of the foil nor zero outside so that such restrictions on x-momentum transfer do not occur for this reason.[15] More realistic boundary conditions will be used in the treatment given below.

A quantum-mechanical derivation of the dielectric constant of a finite metal seems rather difficult. One might assume that the metal electrons experience a uniform potential inside the foil and that their wave functions vanish at the foil boundaries, rather than taking free electron momentum eigenfunctions appropriate to an infinite medium. One finds in this case that the Fourier components of the field are not proportional to the same Fourier components of the sources

[15] Gabor's unconventional application of first-order time dependent perturbation theory leads to a result which disagrees with the Pines-Bohm formula for the mean free path for plasma loss in thick films. He assumes that the wave function of the incident electron is a plane wave with duration $\tau\gg1/\omega_p$, where $\hbar\omega_p$ is the energy loss of the incident electron. He finds that the total probability of plasma excitation is proportional to the duration τ which leads to an unrealistic result for an infinitely long wavetrain. The more conventional perturbation theory [L. I. Schiff, *Quantum Mechanics*, McGraw-Hill Book Company, Inc., New York, 1949), second edition, p. 200] shows that instead of τ, one should use a/v, the electron "transit time" in the foil. Experimental evidence on the energy variation of mean free path for plasma loss [Blackstock, Ritchie, and Birkhoff, Phys. Rev. **100**, 1078 (1955)] shows good agreement with the Pines-Bohm formula and does not agree as well with Gabor's Eq. (34).

of the field. Hence the dielectric constant as it isusually defined does not exist for the finite foil. However, quantum corrections to the classical expression for the dielectric constant of a free electron gas are not important as long as one considers only small deflections of incident electrons with energies much greater than the plasma energy $\hbar\omega_p$. To show this, we consider the angular distribution for the infinite medium, Eq. (14).

$$P(\theta)d\Omega=\frac{\omega_p e^2}{2\pi\hbar v^2}\left[1+\frac{6\omega_0\omega_E}{5\omega_p{}^2}\theta^2+\frac{\omega_E{}^2}{\omega_p{}^2}\theta^4\right]^{-\frac{1}{2}}$$
$$\times[\theta^2+(\omega_p/2\omega_E)^2]^{-1}d\Omega. \qquad (18)$$

Now ω_0 and ω_p are usually not greatly different, so that the square-root term may be written approximately

$$\left[1+\frac{\omega_E}{\omega_p}\theta^2+\left(\frac{\omega_E}{\omega_p}\theta^2\right)^2\right]^{-\frac{1}{2}}.$$

The term in θ^2 represents the plasma dispersion property which may be obtained classically while the quantum correction is given by the term in θ^4. Now if we take the case of 1.5-kev electrons incident on Al, for which $\hbar\omega_p\sim 14.7$ ev, then $\omega_E/\omega_p\sim 10^2$ and we have

$$P(\theta)\sim\frac{\omega_p e^2}{2\pi\hbar v^2}[1+10^2\theta^2+10^4\theta^4]^{-\frac{1}{2}}[\theta^2+\tfrac{1}{4}\times 10^{-4}]^{-1}.$$

The Coulomb factor (the last one) begins to vary appreciably at $\theta\sim 5$ milliradians, while at this angle the dispersion term represents a correction of only 0.25% and the quantum correction only 10^{-3}%. Furthermore, both of these corrections become less important for higher incident energies. Thus we may assume that a semiclassical treatment of the finite foil case will give a good approximation to the correct energy-angle distribution.

Pines[16] has pointed out that quantum effects show up in finite foils in another way. On the basis of the uncertainty principle one may say that when a fast electron interacts with plasma confined to a foil of thickness a, there must be restrictions on the x-momentum transfer to the plasma, *viz.*, it must take place in multiples of h/a. Again, because of the fact that ϵ is a slowly varying function of $\mathbf{k}$, this quantum effect should introduce only small errors in the semiclassical treatment given below.

We shall now employ the linearized hydrodynamical equations of Bloch[17,18] to describe the behavior of the perturbed conduction electrons. One may show[19] that the dielectric constant of an infinite free-electron gas derived from the Bloch model is a good approximation to the correct one in the case which we are considering. The angular distribution of fast electrons which one obtains is essentially the same as Eq. (14) above except for the quantum correction term.

If one assumes irrotational motion, the linearized Bloch equations may be written

$$g\psi+\frac{\partial}{\partial t}\psi=-\frac{e}{m}\varphi+\frac{P\rho}{m\rho_0}, \qquad (19a)$$

$$\partial\rho/\partial t=\rho_0\nabla^2\psi, \qquad (19b)$$

$$\nabla^2\varphi=4\pi e\rho+4\pi e\delta(x-vt)\delta(y)\delta(z). \qquad (19c)$$

Here ψ is the velocity potential, φ the electric potential, and ρ is the electronic density in the foil, all considered as perturbations around the undisturbed state of the plasma. The three equations (19) are, respectively, the first integral of the equation of motion, the equation of continuity, and Poisson's equation; ρ_0 is the electronic density in the undisturbed state, P/ρ_0 is the pressure change per unit number density in the undisturbed gas, and $P=(3h^3/8\pi)^{\frac{2}{3}}\rho_0{}^{\frac{2}{3}}/3m$. The term $g\psi$ in Eq. (19a) is intended to represent a schematic damping of the electronic motion. The exact value of g is unimportant since we shall assume it to be small. This damping term is quite similar to the damping introduced in the classical derivation of the dielectric constant for free electrons.

We are now in a position to solve for the electric potential everywhere in space. The foil will be assumed infinite in the y and z directions and the boundaries of the foil will be taken at $x=0$ and $x=a$.

Outside the foil it is assumed that the electric potential is the solution of

$$\nabla^2\varphi=4\pi e\delta(x-vt)\delta(y)\delta(z). \qquad (19d)$$

Let

$$\varphi=\begin{cases}\varphi_1, & -\infty<x<0\\ \varphi_2, & 0<x<a\\ \varphi_3, & a<x<\infty.\end{cases} \qquad (20)$$

The boundary conditions are that the electric potential and field intensity must be continuous, and the normal component of velocity must vanish at the foil surfaces. The set of Eqs. (19) subject to the given boundary conditions may be solved by expanding the y, z, and t dependence of φ, ψ, and ρ in Fourier integrals as follows:

$$\varphi(x,y,z,t)=\frac{1}{(2\pi)^3}\int dk_y\int dk_z\int d\omega\,\varphi(x,k_y,k_z,\omega)$$
$$\times\exp[i(yk_y+zk_z+\omega t)]. \qquad (21)$$

Equations (19) are then reduced to linear differential equations and may be solved in a straightforward manner. To be consistent with the assumption that the incident electron velocity is large compared with the Fermi velocity in the plasma, we shall assume that the term involving P in Eq. (19a) is small. This term represents the propagation of disturbances in the hydro-

[16] D. Pines (private communication).
[17] F. Bloch, Z. Physik **81**, 363 (1933); Helv. Phys. Acta **I**, 385 (1934).
[18] H. Jensen, Z. Physik **106**, 620 (1937).
[19] See reference 10, p. 23.

dynamical gas. Retaining only first-order terms in $P/\rho_0 m$ one may readily solve for φ. This is also equivalent to neglecting the dispersion term in Eq. (18) for the angular distribution in the infinite metal. This neglect has been shown above to introduce little error for fast incident electrons.

The total energy loss of the electron is given by

$$W=-e\int_{-\infty}^{\infty}\frac{\partial}{\partial x}\varphi_m(r,t)\,|_{x=vt,\,y=z=0}dx$$
$$=\frac{ie}{(2\pi)^3}\int d\mathbf{k}\int d\omega k_x\delta(k_x+\omega/v)\varphi(\mathbf{k},\omega),\quad (22)$$

where $\varphi_m(\mathbf{r},t)$ represents the field in the medium minus the vacuum field of the incident particle and $\varphi_m(\mathbf{k},\omega)$ is its Fourier transform. One notes that just as in the case of an infinite medium there is a restriction on the x component of momentum loss of the fast electron. This is just an expression of the fact that if the fast electron undergoes momentum change $\hbar k_x$ in the x direction and is deflected through a small angle then from conservation of energy and momentum $\hbar k_x = -\Delta E/v$ where ΔE is its energy loss.

Carrying out the integration over k_x and collecting terms, one finds eventually for the total transition probability

$$P(k_\perp,\omega)=\frac{e^2}{\pi^2\hbar v^2}\left\{\mathrm{Im}\left(\frac{1}{\epsilon}\right)\frac{a}{(k_\perp^2+\omega^2/v^2)}+\frac{2k_\perp}{(k_\perp^2+\omega^2/v^2)^2}\mathrm{Im}\left[\frac{1-\epsilon}{\epsilon}\cdot\frac{2(\epsilon-1)\cos(\omega a/v)+(\epsilon-1)\exp(-k_\perp a)+(1-\epsilon^2)\exp(k_\perp a)}{(\epsilon-1)^2\exp(-k_\perp a)-(\epsilon+1)^2\exp(k_\perp a)}\right]\right\},\quad (23)$$

where $\epsilon=[1-\omega_p^2/(\omega^2-ig\omega)]$.

One sees that the first term is just a times the transition probability per unit path length in an infinite medium. The other terms represent the boundary correction. One may examine first the boundary correction term in the limit of large a. Then

$$P(k_\perp,\omega)\to\frac{e^2a}{\hbar\pi^2v^2}\left\{\frac{\mathrm{Im}(1/\epsilon)}{(k_\perp^2+\omega^2/v^2)}-\frac{2k_\perp}{a(k_\perp^2+\omega^2/v^2)^2}\mathrm{Im}\left(\frac{(1-\epsilon)^2}{\epsilon(1+\epsilon)}\right)\right\}.\quad (24)$$

Now let us define

$$P(k_\perp,\omega)=\{aP_\infty'(k_\perp,\omega)+P_b(k_\perp,\omega)\},$$

where P_∞' is the transition probability per unit foil thickness in an infinite foil and P_b is the term introduced by the boundary effect. Then one may write, inserting the expression for ϵ,

$$P_b=\frac{e^2}{\pi^2\hbar v^2}\frac{2k_\perp}{(k_\perp^2+\omega^2/v^2)^2}\cdot\frac{g\omega_p^4}{\omega}\cdot\left\{\frac{\frac{1}{4}\omega_p^2}{(\omega-\frac{1}{2}\omega_p^2)^2+g^2\omega^2}-\frac{\omega_p^2}{(\omega^2-\omega_p^2)^2+g^2\omega^2}\right\}.\quad (25)$$

One notes that the effect of the boundary is to cause a decrease in loss at the plasma frequency and an additional loss at $\omega=\omega_p/\sqrt{2}$. Call the probabilities for these processes P_{b1} and P_{b2}, respectively. Then integrating over ω,

$$P_{b1}=-\frac{e^2\omega_p^2}{\pi\hbar v^2}\frac{1}{\omega_p}\frac{k_\perp}{(k_\perp^2+\omega_p^2/v^2)^2},\quad (26)$$

$$P_{b2}=\frac{e^2\omega_p^2}{\pi\hbar v^2}\cdot\frac{\sqrt{2}}{\omega_p}\cdot\frac{k_\perp}{(k_\perp^2+\omega_p^2/2v^2)^2}.\quad (27)$$

Again setting $k_\perp=mv\vartheta/\hbar$, one sees that the angular dependence in these terms is very different from that occurring in the angular term for the infinite medium. Carrying out the integrations over $k_\perp$, we find

$$P_{b1}=-\tfrac{1}{2}\pi(e^2/\hbar v),\quad (28)$$

$$P_{b2}=\pi(e^2/\hbar v).\quad (29)$$

The P_{b1} term subtracts from the aP_∞' term, showing that the loss at the plasma frequency should decrease as the foil thickness decreases, while the loss at $\omega_p/\sqrt{2}$ is negligible for thick foils but increases as the thickness decreases.

This shift in the resonance frequency is due to the depolarizing effect of the surfaces of the foil.[20] Jensen[18] has shown that the resonance frequency of plasma contained in a small sphere is less than the value appropriate to an infinite plasma by a factor of $1/\sqrt{3}$ and that this shift is due to the depolarizing effect of surface charge on the sphere. An analogous effect occurs for the plane foil. It may easily be verified using a simple classical analysis that the resonance frequency is indeed $\omega_p/\sqrt{2}$ for electrons in a plane foil so thin that the surface charge determines the field within the foil.

To examine the trend of P_{b1} for small a, one may return to Eq. (20), integrate over ω taking account of only the resonance at $\omega=\omega_p$ and then integrate over $k_\perp$. One finds

$$P_{b1}=\frac{-2e^2}{\hbar v}t\int_0^\infty\frac{x^2dx}{(x^2+t^2)^2}\frac{\cosh x-\cos t}{\sinh x},\quad (30)$$

where $t=a\omega_p/v$.

Now substituting the rational fraction expansion of $\coth x$ and $\mathrm{csch} x$ into this equation and assuming t to

[20] The writer is indebted to Dr. David Pines for pointing out this fact to him.

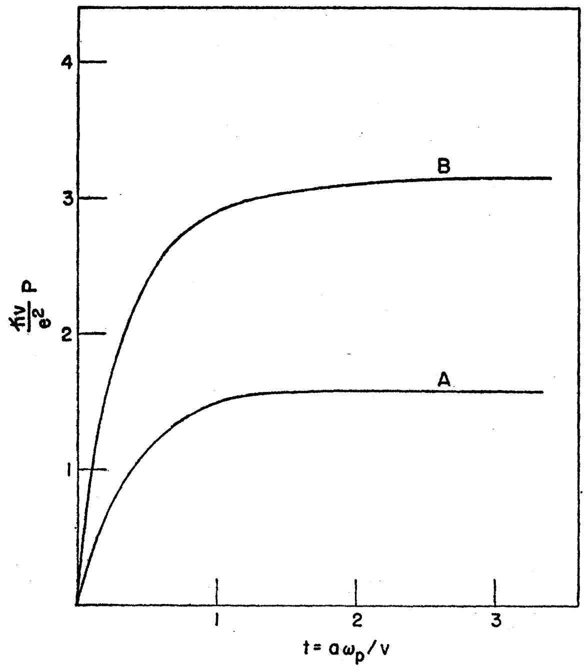

FIG. 1. Interaction probability in thin foil *vs* foil thickness. Curve A is P_{b1}, the decrease in interaction probability at plasma frequency due to finiteness of foil. Curve B is P_{b2}, the probability for loss at "lowered" plasma frequency; a is the foil thickness; v is the electron velocity; and ω_p is the plasma frequency.

be small, one finds

$$P_{b1} = -\frac{e^2}{\pi v} t\left[\ln\left(\frac{1}{t}\right) + 1.57\cdots\right]. \tag{31}$$

Now one may also write[1]

$$aP_\infty' = \frac{e^2}{\hbar v} t\left[\ln\left(\frac{1}{t}\right) + \ln ak_c\right], \tag{32}$$

where k_c is the cut-off wave vector introduced by Pines and Bohm.[2] Then the total transition probability for loss at the plasma energy in very thin films ($t \ll 1$) should be

$$aP_\infty' + P_{b1} = \frac{e^2}{\hbar v} t \ln\left(\frac{ak_c}{4.82\cdots}\right).$$

This formula is obviously wrong when $ak_c < 4.82$. However, the theory used here will surely break down before the foil thickness becomes this small, since k_c is approximately the reciprocal of the mean interelectronic spacing.[3] Figure 1 shows a plot of $P_{b1}(t)$ as a function of t.

The P_{b2} term is interesting in that it shows a coupling between ϑ and ω as a becomes small. This is shown by considering the behavior of the denominator. It is found that resonances occur at

$$\omega = \frac{\omega_p}{\sqrt{2}}[1 \pm \exp(-ak_\perp)]^{\frac{1}{2}} \equiv \frac{\omega_p}{\sqrt{2}}\alpha_\pm^{\frac{1}{2}}. \tag{33}$$

Then

$$P_{b2}(k_\perp)2\pi k_\perp dk_\perp = \frac{e^2}{\hbar v}\frac{\omega_p}{v}\sqrt{2}k_\perp^2 dk_\perp \left\{ \frac{\alpha_+^{\frac{1}{2}}}{\alpha_-}\frac{1-\cos(t\alpha_+^{\frac{1}{2}}/\sqrt{2})}{(k_\perp^2+\omega_p^2\alpha_+/2v^2)^2} + \frac{\alpha_-^{\frac{1}{2}}}{\alpha_+}\frac{1+\cos(t\alpha_-^{\frac{1}{2}}/\sqrt{2})}{(k_\perp^2+\omega_p^2\alpha_-/2v^2)^2} \right\}. \tag{34}$$

The first term in this equation comes from the resonance at $\omega = \omega_p\alpha_+^{\frac{1}{2}}/\sqrt{2}$ and the second from $\omega = \omega_p\alpha_-^{\frac{1}{2}}/\sqrt{2}$. To find the total interaction probability one may now integrate over $k_\perp$. The result is shown in Fig. 1. The distribution of losses in the P_{b2} term is peaked about the energy loss $\hbar\omega_p/\sqrt{2}$ even for $t \sim 1$. This may be easily shown by converting $P(k_\perp)$ to the probability distribution in ω, using the relation [Eq. (33)] between $k_\perp$ and ω.

The net boundary effect is an increase in total energy loss to the conduction electrons above the value which would exist in its absence. The total energy loss to the conduction electrons per unit thickness increases logarithmically as the foil thickness decreases.

The possibility occurs to one that these sub-plasma frequency losses may be identified with the low-lying losses observed by some experimenters using thin foils.[3] It does not seem that the observed values of the losses are $1/\sqrt{2}$ times the "characteristic" losses observed in the same metals. However, it should be noted that thin metallic films may have a strongly granular structure. The strong variation of the grain structure with substrate composition, rate and amount of condensation, etc., of thin evaporated metallic films has been discussed by Heavens.[21] In this reference are given electron micrographs which clearly show the transition from small grain size to a state in which the grains merge to form a nearly uniform film as the amount of material deposited is increased in a series of films. The surface depolarization effect will certainly be larger for a small grain of average dimension a than for the semi-infinite plane foil of thickness a which was treated above. Thus one would expect the "lowered" losses in an actual foil to lie closer to the value $\hbar\omega_p/\sqrt{3}$, appropriate to a spherical grain, than to the value $\hbar\omega_p/\sqrt{2}$. This seems to be true for the low-lying losses which have been observed.

The two most puzzling features about the experimental result on low-lying losses are (a) that such loss lines seem to be narrower and to occur with higher probability than one would expect if they were due to interband transitions of individual electrons,[22] and (b) that some experimenters observe these losses while others do not. The depolarization effect seems to offer a possible explanation of these features.

[21] O. S. Heavens, *Optical Properties of Thin Solid Films* (Academic Press, Inc., New York, 1955).

[22] D. Pines, Revs. Modern Phys. **28**, 198 (1956).

One expects losses at the "lowered" plasma energy to occur whenever the film or grain dimensions approach values $\sim v/\omega_p$ and that in this event the probability of such losses would be comparable with losses at the plasma energy. Since grain size as well as film thickness may vary greatly from one experiment to another, one might expect that the "lowered" plasma losses would not be seen by all experimenters.

Existing experimental evidence from the study of solids by means of fast electron bombardment does not seem to be detailed enough as yet to judge whether or not these "lowered" losses really occur. However, the depolarization effect has been observed in optical investigation of thin solid films. To account for the observed variation in the optical constants of metallic films with film thickness it has been necessary to take into account the depolarization effect of grain boundaries and the depolarizing effect of the grains upon each other. This effect has been calculated classically by David[23] and others.[21] The close relation between the response of a solid to electromagnetic radiation and to bombardment by electrons is well known. Hence one expects that a similar depolarization effect may occur in experiments with fast electrons.

The present observations serve to strengthen the recommendation of Pines[5] that a careful investigation of electron losses in a single metal for various foil thicknesses and bombarding energies should be made.

The detailed mathematical treatment given above applies to a rather idealized model of a metal foil. However, qualitative considerations based upon this treatment should enable one to interpret experimental evidence on the variation of mean free path with foil thickness. If the probability of a low-lying loss is found to decrease linearly as the foil thickness decreases and if it occurs with constant probability relative to the plasma loss probability, then one would conclude that it is due to one-electron interband transitions. On the other hand, if the probability of a low-lying loss is found to increase relative to the probability for plasma loss as the grain size and foil thickness decrease, then the depolarization effect is the more likely explanation.

As noted above, the depolarization effect is expected to have little influence upon the one-electron levels in a solid metallic foil, while this effect may make a large difference in the energy required to produce a plasmon in the foil. If one could determine that the "lowered" losses do exist in certain solids, then the collective nature of the characteristic losses in those solids would be firmly established.

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Note added in proof.—The author has developed a quantum generalization of the dielectric approach of Lindhard and Hubbard which makes unnecessary the concept of the classically prescribed field, $\varphi(\mathbf{r},t)$, which is present in the theory of Lindhard and Hubbard. In a subsequent paper it will be shown that the dielectric approach is capable of describing simultaneously the individual and collective aspects of electronic motion in plasma and is valid for all values of the wave vector in the region of validity of first-order perturbation theory. A formula will be given which is identical with Eq. (11) above for small momentum transfers by fast electrons to plasma and which reduces to the Born scattering formula for large momentum transfers. The correlation energy in metals, excitations in the general dielectric medium including both long- and short-range interactions, and the relativistic generalization of the dielectric theory will also be considered.

[23] E. David, Z. Physik **114**, 389 (1939).