Predicted Radiation of Plasma Oscillations in Metal Films*

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Because of their highly collective nature it is predicted that plasma oscillations in a thin metal film should, under the proper circumstances, give off ultraviolet radiation. The plasma oscillations can be excited by fast electrons, incident normal to the film and inelastically scattered by it. Surface effects are essential, and of the special types of oscillations which can occur in a plane parallel slab of electron gas, only that involving motion normal to the slab can radiate. The yield is computed to be one photon for every one thousand electrons incident at 10 kev. The radiation is at the plasma frequency, ω_p, or at 2100 A for a sodium film. Its identification should be facilitated by the characteristic $\cos\theta$ dependence of the intensity, where θ is the angle between the foil normal and the direction of emission of the photon. Straightforward computation yields a radiative mean life of $\omega_p^{-1}(\lambda_p/\pi\tau)\times(\cos\theta/\sin^2\theta)$, which is generally shorter than that due to intrinsic interband damping, except at small angles. λ_p is the photon wavelength and τ the film thickness. From the competition of the two decay modes it should be possible to determine the intrinsic damping rate, and hence the product of the optical constants nk. The radiative lifetime is so short as to produce appreciable line-broadening, and thereby provide an independent check on the experiment. In the appendix the inelastic electron scattering coefficient is derived for the excitation in a thin film of the radiative-type plasma oscillations.

I. INTRODUCTION

ONE of the most striking of the electronic properties of a metal is the ability of its electron gas to undergo plasma oscillations. These oscillations, in which all of the conduction electrons participate, are a consequence both of the inertia of the electrons and of their repulsive Coulomb interactions. Definite evidence for these oscillations is obtained from experiments in which a metal foil is bombarded with fast electrons (of, say, 20 kev) whose energy is precisely measured both before and after they have passed through the foil. The energy lost is found not to have random values, which might be expected from chance collisions with conduction electrons of various velocities in the metal, but on the contrary to be quite discrete. For some metals (such as Al, Mg, and the alkali metals) the energy loss spectrum consists of especially sharp lines—the so-called characteristic energy losses or eigenlosses. These lines appear at multiples of a basic quantum of energy which is generally found to be equal to Planck's constant times the classical frequency of oscillation of the electron plasma in the metal. For this, and other reasons, the energy loss experiment is most naturally interpreted in terms of plasma excitation[1,2] and the basic unit of energy has been termed the plasmon.[3]

The accuracy and resolution of the energy loss experiments are limited by the necessity of using high-energy electrons. (The mean free path of the incident electrons must be of the order of magnitude of, or greater than, the thickness of the foil. Otherwise more complicated multiple scattering processes will occur.) The energy loss is of the order of a few electron volts and quite small by comparison. The quantity of interest is thus unfortunately the difference of two large and practically equal numbers. It would consequently be highly desirable to find an alternative method of measuring the frequency of the oscillations. Since accelerated electric charges in general radiate, it is only natural to hope that under suitable circumstances the plasma oscillations will give off electromagnetic radiation. The wavelength of this ultraviolet light would then yield a precise value for the frequency. It is immediately clear that the oscillations must have wavelengths at least as long as the radiation arising from them. As a consequence the electric field, which acts between different portions of a plasma wave and sustains its oscillation, may suffer important retardation corrections. Although these corrections have not yet been thoroughly studied,[4] we want to present in this paper several conclusions which seem to be independent of retardation. In particular, we find that fast electrons passing through thin metal foils should yield some fluorescent radiation via plasma oscillations.

Although the sharp eigenlosses observed in many metals are quite generally attributed to collective oscillations of the electron plasma, there seems to have been no attention, either experimental or theoretical, to the possibility that these oscillations might give off detectable radiation, of wavelength corresponding to the characteristic frequency of the plasma.[5] This situation is without doubt attributable to the longitudinal nature of plasmons in a bulk electron gas. In such a case the electron motion is in the direction of the plasmon momentum and is therefore not coupled to the transverse waves of the electromagnetic field. This restriction does not, however, apply to the thin foils which are used in the transmission-type eigenloss experiments. The image force at the surfaces of a foil constrains the electrons to remain inside, and

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[1] R. A. Ferrell, Phys. Rev. **101**, 554 (1956).
[2] R. A. Ferrell and J. J. Quinn, Phys. Rev. **108**, 570 (1957).
[3] D. Pines, Revs. Modern Phys. **28**, 184 (1956). This survey article should be consulted for further references in the field.
[4] They are presently being investigated by the present author and E. A. Stern of this university.
[5] A report on these ideas has been given at the Eugene, Oregon meeting of the American Physical Society [R. A. Ferrell, Bull. Am. Phys. Soc. Ser. II, **1**, 244 (1956)].

contributes an uncertainty to the momentum normal to the foil. The plasmon thereby acquires a degree of transversality, which enables it to radiate. Experimental detection of this radiation should not be difficult, and promises to be a useful tool for studying the plasma oscillations in metals. Not only would it constitute decisive evidence for the collective nature of these oscillations, but, as mentioned above, it would provide a means of measuring the plasma frequencies with an accuracy in excess by orders of magnitude of that attained by the conventional method of electron energy loss. As we have seen above, the energy loss method requires taking the difference of two large quantities, each of which exceeds the quantity of interest by a factor of the order of 10^3. The optical method, on the other hand, would measure the difference directly. Both methods are familiar in the analogous field of nuclear physics. The "optical" method in the latter case is, of course, the general technique of gamma-ray spectroscopy, and is an indispensable tool for investigating the excited states of nuclei. It may be expected that the corresponding technique in solid-state physics could assume comparable importance. To push the analogy further, the experiment we are here advocating for the metal foils[6] corresponds, for the nuclei, to the well-known Coulomb excitation by incident charged particles followed by detection of the fluorescent gamma ray.

The plasmons which radiate must have essentially zero momentum parallel to the foil, since the wavelength of the optical radiation, $\lambda_p=2\pi c/\omega_p$, is very long (of the order of 10^3 A). (c is the velocity of light and ω_p the plasma frequency.) As will be shown below, the type of oscillation which radiates involves electron motion primarily normal to the foil, and consists essentially, over any particular region of the foil, of a vibrating double layer with phase appropriate to the region. The phase varies gradually as one passes along the foil from one region to another, and is specified by the factor $\exp i(k_x x+k_y y)$, where the front surface of the foil has been taken in the $x-y$ plane and $\hbar k_{x,y}$ are components of the plasmon momentum in this plane. This vector, which we designate by $\hbar\mathbf{k}$, determines the direction of the photon emitted by the plasmon. Letting the photon wave number be of magnitude $k_p=2\pi/\lambda_p=\omega_p/c$ and have its direction at angle θ with the z axis (direction of motion of the incident electrons), conservation of momentum in the $x-y$ plane requires $k_p \sin\theta=k$, or

$$\sin\theta=k/k_p. \qquad (1)$$

(Throughout this paper we deal only with the case of normally incident electrons.) Thus, plasmons of momentum greater than $\hbar k_p$ cannot radiate and can only decay by ordinary electronic damping. Those with $k<k_p$ decay with both modes of damping competing. According to estimates arrived at in Sec. IV below, the effective lifetime due to radiation alone is surprisingly short. Therefore, in relatively ideal metals, such as aluminum, magnesium, and sodium, radiation competes so favorably with electronic damping that the decay can be expected to go almost entirely by radiation, provided momentum conservation permits. The radiation lifetime determined below is, to be sure, angle dependent and increases as θ^{-2} for small θ. Therefore, in the limit of small angles electronic damping is bound to dominate, and no radiation will be detected. The angle at which the radiation intensity begins to fade provides therefore a measurement of the intrinsic electronic damping rate.

By assuming that every plasmon which is allowed to radiate will do so, we can very easily determine the angular distribution of the radiation. As explained in the preceding paragraph, this assumption can be expected to be valid except at the small angles, where the intensity determined here is an overestimate. From reference 1, Eq. (8), we see that the probability of creating a plasmon by inelastically scattering a fast incident electron is independent of plasmon momentum $\hbar k$, for small values of k. Equation (8) must by modified for very thin foils, as is shown in Appendix I, but its constancy for small k remains unchanged. Let us define the scattering coefficient $\mu(\alpha;\tau)$ by writing the probability of inelastically scattering the incident electron by angle α into solid angle $d\Omega$ as $\mu(\alpha;\tau)d\Omega$, where τ is the thickness of the foil.[7] Conservation of momentum for the plasmon creation process determines $d\Omega$ as $(\hbar/p)^2$ times the element of area in the k_x-k_y plane, where p is the momentum of the incident electrons. The element of solid angle corresponding to photon emission into a cone between θ and $\theta+d\theta$ is obtained by referring to Fig. 1 of reference 1 and to Eq. (1) above. One finds

$$\begin{aligned} d\Omega &= (\hbar k_p/p)^2 2\pi \sin\theta d \sin\theta \\ &= (\hbar k_p/p)^2 \cos\theta (2\pi \sin\theta d\theta), \end{aligned} \qquad (2)$$

where the quantity in parentheses is now the differential solid angle for the photons. The probability per unit solid angle per incident electron of emitting a photon at angle θ to the z axis can consequently be written as

$$\mu_p(\theta;\tau)=[\mu(0;\tau)/2](\hbar k_p/p)^2 \cos\theta, \qquad (3)$$

where $\mu_p(\theta;\tau)$ is the scattering coefficient for photon emission. A factor of one-half has been included because any given plasmon will radiate equally both above and below the $x-y$ plane.

The photon yield per incident electron, $Y(\tau)$, can be found by integrating Eq. (3) over all photon solid

[6] Efforts to detect the radiation from sodium foils are being made by E. J. O'Brian, R. M. Talley, and E. P. Trounson of the Naval Ordnance Laboratory.

[7] $\mu d\Omega$ is τ times the "differential inverse mean free path" of reference 1. We prefer to avoid introducing an equivalent cross section per electron, as done by some authors, since the scattering is a property of the interacting electron plasma and does not exist for individual electrons isolated in space.

angles,

$$Y(\tau)=\mu(0;\tau)\pi(\hbar k_p/p)^2. \tag{4}$$

The scattering coefficient $\mu(0;\tau)$ is calculated in Appendix I, but here a few qualitative considerations will suffice for our purpose. The main point to be noted is that, for optimal yield, the incident electrons should be of as high velocity as possible. This is seen by considering the equation $\Delta E=\mathbf{v}\cdot\Delta\mathbf{p}$, essentially one of Hamilton's canonical equations of motion, where ΔE and $\Delta\mathbf{p}$ are the energy and momentum lost by the fast electron and $\mathbf{v}$ is its velocity. Setting $\Delta E=\hbar\omega_p$, the plasmon energy, we find the minimum value of Δp by taking the two vectors parallel:

$$\Delta p=\hbar\omega_p/v=2\pi\hbar\lambda_p^{-1}(v/c)^{-1}. \tag{5a}$$

This momentum change corresponds to a difference in the wave numbers of the initial and final states of the electron of $\Delta k=\Delta p/\hbar$. Consequently, using the Born approximation to describe the interaction of the electron with the plasma, we see that the electron causes the excitation by means of an effective potential which has a variation in space corresponding to the wavelength

$$\lambda_{\text{ex}}=2\pi/\Delta k=(v/c)\lambda_p. \tag{5b}$$

If the foil is too thick compared to λ_{ex} the effective potential will act on different parts of the plasma with different phases and be ineffective for the present purpose. For $v/c=\frac{1}{5}$ (about 10-kev electrons) and $\lambda_p=2100$ A (Na plasma radiation), Eq. (2) gives $\lambda_{\text{ex}}=420$ A. The optimal thickness of the foil would generally be approximately one-half a wavelength, which in this case amounts to about 200 A. In carrying out the experiment care should be taken not to exceed this limit, which, however, can be raised by using higher energy electrons.

Having determined the optimal film thickness, we now need an explicit expression for the scattering coefficient $\mu(\alpha;\tau)$. As a first approximation we can take over the expression for the bulk electron gas [Eq. (8) of reference 1]:

$$\tau\frac{d(1/\lambda)}{d\Omega}=\frac{\tau\hbar\omega_p}{2\pi mv^2a_0}\left(\frac{1}{\alpha^2+\theta_E^2}\right).$$

(Here we use α instead of θ for the angle of scattering, to avoid confusion with the angle of photon emission.) By re-examining the derivation of this expression it is easily seen that it is relativistically correct, provided θ_E is defined as $\Delta p/p=\hbar\omega_p/vp$. In Appendix I it is shown that as a consequence of the finite thickness of the film two correction factors arise. The first is $(\pi\tau/\lambda_{\text{ex}})^{-2}\sin^2(\pi\tau/\lambda_{\text{ex}})$ and expresses quantitatively the effect discussed in the preceding paragraph. The second is $\theta_E^2/(\alpha^2+\theta_E^2)$ and results from the special nature of the plasma oscillations in thin films. Thus we have, after substituting from Eq. (5b)

$$\mu(\alpha;\tau)=\frac{e^2}{\pi\hbar v}\left(\frac{\theta_E^2}{(\alpha^2+\theta_E^2)^2}\right)\left(\frac{\sin^2(\pi\tau/\lambda_{\text{ex}})}{\pi\tau/\lambda_{\text{ex}}}\right). \tag{6a}$$

It is interesting that the total scattering coefficient integrated over all angles of scattering yields simply

$$\int_0^\infty \mu(\alpha;\tau)2\pi\alpha d\alpha=\frac{e^2}{\hbar v}\left(\frac{\sin^2(\pi\tau/\lambda_{\text{ex}})}{\pi\tau/\lambda_{\text{ex}}}\right), \tag{6b}$$

and does not contain the logarithmic factor of Pines'[8] expression for the mean free path for plasmon excitation in a bulk electron gas. Equation (6b) is consequently independent of the short wavelength cutoff for plasmons.[9] This is because only plasmons of a certain type are included in Eq. (6a). Setting $\alpha=0$, making the choice $\tau=\lambda_{\text{ex}}/2$ of the preceding paragraph, and substituting into Eq. (4) yields

$$Y(\lambda_{\text{ex}}/2)=\frac{2}{\pi}\frac{e^2}{\hbar c}\frac{v}{c}. \tag{7a}$$

This result can be increased somewhat by choosing the somewhat smaller thickness $\tau=0.37\lambda_{\text{ex}}$, giving

$$Y=0.0053v/c. \tag{7b}$$

Equation (7b) gives one-tenth of one percent, or one photon for every thousand incident electrons, for the case of $v/c=\frac{1}{5}$. As already emphasized, the yield will be increased to the extent that it is possible to use electrons of higher incident velocity and films of appropriately greater thickness. In this way the scattering pattern can be contracted more into the very small angles which correspond to the very long wavelength radiative-type plasma oscillations. Equation (7b) predicts a maximum yield of one-half of one percent.

A further item which we mention here only in passing is that, in the preparation of these relatively thin foils, the metal generally aggregates on the substratum in the form of small spherical droplets.[10,11] Special experimental techniques are required (e.g., deposition at low temperature) to obtain a uniform flat slab of metal with plane parallel surfaces. Being the simplest, this is the only case dealt with below. It is hoped, however, that in the future some detailed calculations can be made on the radiation to be expected from the nonideal case of separate spheres distributed at random over the substratum. If the spheres are sufficiently isolated from one another they will all vibrate independently, but in unison, at the reduced frequency $\omega_p/\sqrt{3}$.

[8] D. Pines, Phys. Rev. **92**, 626 (1953).
[9] R. A. Ferrell, Phys. Rev. **107**, 450 (1957).
[10] O. S. Heavens, *Optical Properties of Thin Solid Films* (Butterworths Scientific Publications, Ltd., London, 1955).
[11] H. Mayer, *Physik Dünner Schichten* (Wissenschaftliche Verlag Sgesellschaft, Stuttgart, 1950), Vol. I.

Since they are all excited at the same time by the incident electron they will oscillate in phase and contribute coherently to some detectable radiation at this reduced frequency.[12]

In the following pages, Sec. II gives a treatment of the long wavelength plasma oscillations by means of the frequency dependent dielectric constant. Of the two different possible types, the type which can actually radiate is selected in Sec. III, which discusses the effect of retardation. The radiation lifetime of these "normal" oscillations is computed in Sec. IV and shown to be much shorter than the ordinary interband damping lifetime, for relatively ideal metals. Section V concludes this paper with a brief summary.

II. DIELECTRIC THEORY

The dynamic electrical properties of a medium are expressed by the frequency dependent dielectric constant $\epsilon(\omega)$. Generally ϵ is also a function of wave number, but in the present problem we are interested only in long wavelengths, i.e., in the limit of vanishing wave number. For the free electron gas

$$\epsilon(\omega)=1-\omega_p^2/\omega^2. \tag{8}$$

ϵ is defined as the ratio of the electric displacement vector $\mathbf{D}=\mathbf{E}+4\pi\mathbf{P}$, (where $\mathbf{P}$ is the polarization), to the electric field $\mathbf{E}$. Thus,

$$\mathbf{D}=\epsilon\mathbf{E}. \tag{9}$$

For a bulk gas in the absence of external sources $\mathbf{D}=0$, which requires $\epsilon\mathbf{E}=0$. The condition for a nonzero electric field is therefore $\epsilon=0$, which according to Eq. (3) gives

$$\omega=\omega_p, \tag{10}$$

just the classical frequency of oscillation. The case of a bounded slab of gas extending from the plane $z=-\tau/2$ to $z=+\tau/2$ is more complicated. In the absence of external charge we can no longer conclude that the displacement vanishes, but only that it satisfies the continuity equation

$$\nabla\cdot\mathbf{D}=0. \tag{11}$$

This equation has the well-known consequence that the normal component of $\mathbf{D}$, in this case D_z, is continuous across the boundaries. Substitution of Eq. (9) into Eq. (11) gives $\nabla\cdot\mathbf{E}=0$, except at the boundaries, where ϵ is discontinuous. Setting $\mathbf{E}=-\nabla\varphi$, where φ is the electric scalar potential, we find Laplace's equation. Because of the symmetry about the $z=0$ (i.e., $x-y$) plane the solutions must be either even or odd functions of z, and it is necessary to consider only positive values of z. For $0<z<\tau/2$ we have

$$\varphi=\varphi_0\cos(\mathbf{k}\cdot\mathbf{x}+\delta)(e^{kz}\pm e^{-kz}),$$

where $\mathbf{k}$ and $\mathbf{x}$ are vectors in the $x-y$ plane and δ is an arbitrary phase shift, while for $z>\tau/2$ we have

$$\varphi=\varphi_0\cos(\mathbf{k}\cdot\mathbf{x}+\delta)e^{-kz}(e^{k\tau}\pm 1).$$

The time dependence can be included either in φ_0 (standing wave) or δ (running wave). The normal components of $\mathbf{E}$ at the boundary in these two cases are

$$E_z=-k\varphi_0\cos(\mathbf{k}\cdot\mathbf{x}+\delta)(e^{k\tau/2}\mp e^{-k\tau/2}),$$

and

$$E_z=k\varphi_0\cos(\mathbf{k}\cdot\mathbf{x}+\delta)(e^{k\tau/2}\pm e^{-k\tau/2}).$$

Using Eq. (9) and equating values of D_z gives

$$\epsilon(\omega)=-\left(\frac{e^{k\tau/2}\pm e^{-k\tau/2}}{e^{k\tau/2}\mp e^{-k\tau/2}}\right), \tag{12}$$

the dispersion relation between the wave number k and the natural frequency of vibration ω. Substitution from Eq. (8) and solving for ω gives the relation more explicitly as

$$\omega=\omega_p\left(\frac{1\mp e^{-k\tau}}{2}\right)^{\frac{1}{2}}. \tag{13}$$

This equation, first derived by Ritchie[13] in a different way, has two interesting limiting cases. For short wavelengths, $k\tau\gg1$, $e^{-k\tau}\ll1$, and the surface waves become decoupled and do not interfere with one another. Each surface sustains independent oscillations at the reduced frequency of $\omega_p/\sqrt{2}$, characteristic of a semi-infinite electron gas with a single plane boundary. The opposite limit of very long wavelengths interests us here, in which case we get "tangential" oscillations, at the highly relaxed frequency $\omega\approx\omega_p(k\tau/2)^{\frac{1}{2}}$, and the electron motion is essentially parallel to the surfaces of the foil. In addition we have "normal" oscillations, at

$$\omega\approx\omega_p(1-k\tau/4), \tag{14}$$

or essentially the full bulk frequency. In this vibrational mode the electrons flow back and forth across the foil from one surface to the other. As will be seen in the next section, it is only for the normal oscillations that there seems to be any prospect of radiation.

III. RETARDATION

For a foil to radiate it is necessary for the phase velocity of the plasma oscillation to surpass that of light, or

$$\omega/k>c. \tag{15}$$

(This is equivalent to the conservation of momentum condition of Sec. I.) It is therefore essential to know how ω varies in the limit $k\to0$. In this long wavelength limit the plasma oscillation involves the transport of electrical charge over considerable distances (i.e., roughly $\lambda/2$) and the resulting current sets up magnetic fields. These in turn, according to Maxwell's equations,

[12] The optical properties of nonuniform metallic films of this type have been studied by E. David [Z. Physik **114**, 389 (1939); **115**, 514 (1940)].

[13] R. H. Ritchie, Phys. Rev. **106**, 874 (1957).

produce additional electric fields and exert additional forces on the electrons in the gas, thereby shifting the frequency. The additional fields can be considered as the corrections in the instantaneous Coulomb interactions which are necessary to take into account the time needed for the propagation of the true retarded interaction from one point in the wave to another. Investigation of the effect of retardation is still under way. Stern[14] has shown that the correct dispersion relation for a semi-infinite gas is

$$\omega=\omega_p\left[1+\frac{1}{2q^2}+\left(1+\frac{1}{4q^4}\right)^{\frac{1}{2}}\right]^{-\frac{1}{2}}, \qquad (16)$$

where $q=k/k_p$. For short wavelengths ($q\gg 1$) we have Ritchie's[13] simple result $\omega=\omega_p/\sqrt{2}$, found above in Sec. II without regard for retardation. Equation (16) gives considerable relaxation at the longer wavelengths, however. As a result, the crucial quantity, the phase velocity

$$\omega/k=c[\tfrac{1}{2}+q^2+(\tfrac{1}{4}+q^4)^{\frac{1}{2}}]^{-\frac{1}{2}} \qquad (17)$$

never attains the necessary value c but only approaches it asymptotically in the limit $q\rightarrow 0$. Thus there can be no radiation from the surface oscillations of a semi-infinite gas.

The effect of retardation in the actual case of thin foils has not yet been worked out, but it is fairly clear that the tangential oscillations, already greatly relaxed, will be further relaxed by retardation and can be dropped from consideration. The normal oscillations, on the other hand, must have a much different behavior. In the limit of infinite wavelength one has simply a plane parallel condenser discharging across itself. The displacement current cancels out the convective current, so that no magnetic field is set up and no retardation correction is necessary. This can also be seen from the fact that the restoring force acting upon any given electron originates from charge piled up at

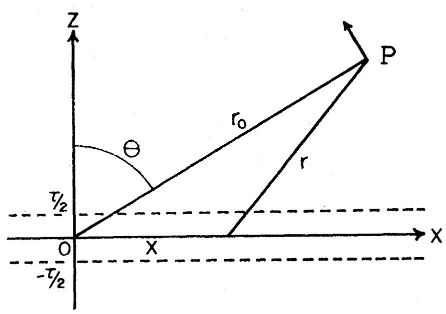

FIG. 1. Radiation field outside a plasma oscillation in a thin metal film. The metal film fills the region between the planes $z=\pm\tau/2$ and extends indefinitely in the directions of the x and y axes. (The y axis is into the paper.) The radiation field at P is the coherent sum of radiation originating at all parts of the film as a result of the oscillatory motion of the electrons in the z direction.

[14] E. A. Stern (private communication).

relatively nearby points on the surfaces of the foil. Therefore Eq. (14) is valid in this limit ($k\rightarrow 0$, $\omega\rightarrow\omega_p$), as well as for the relatively shorter wavelengths. In lieu of a definitive retardation calculation we will for the present interpolate and assume that Eq. (14) also holds for the intermediate wavelengths, or at least, that it is not seriously altered by retardation. The phase velocity then becomes, approximately,

$$\frac{\omega_p}{k}-\frac{\omega_p\tau}{4}=c\left(\frac{\lambda}{\lambda_p}-\frac{\pi}{2}\frac{\tau}{\lambda_p}\right), \qquad (18)$$

and the inequality (15) is satisfied for

$$\lambda\gtrsim\lambda_p+(\pi/2)\tau. \qquad (19)$$

IV. RADIATION

The polarization current which generates the radiation is

$$\mathbf{J}=\frac{\partial\mathbf{P}}{\partial t}=\left(\frac{\epsilon-1}{4\pi}\right)\frac{\partial\mathbf{E}}{\partial t}$$
$$=\left(\frac{1-\epsilon}{4\pi}\right)\text{grad}\frac{\partial\varphi}{\partial t}. \qquad (20)$$

It is expedient to change to a more convenient notation than that of Sec. II and adopt the convention that the physically real field variables are only the real parts of the complex expressions which we now write for them. The potential inside the foil then becomes, for a normal oscillation,

$$\varphi=2\varphi_0 e^{i(\mathbf{k}\cdot\mathbf{x}-\omega t)}\sinh kz, \qquad (21)$$

where we have included explicitly the time dependence for a running wave. (The phase δ is absorbed in the definition of the origin of time.) For a thin foil only the z component of current is appreciable and, from Eqs. (12), (20), and (21), is approximately

$$J_z\approx -i\left(\frac{\omega k}{2\pi}\right)\varphi_0 e^{i(\mathbf{k}\cdot\mathbf{x}-\omega t)}, \qquad (22)$$

independent of z (since $\mathbf{k}$ is a two-dimensional vector). To calculate the radiation at a point P, many wavelengths from the foil, it is convenient to choose the orientation of the coordinate axes so that the direction of propagation of the plasmon falls along the x axis. Then $\mathbf{k}\cdot\mathbf{x}$ reduces to kx. It is further useful to choose the origin of the y and x coordinates so that P lies in the $x-z$ plane and so that the radius vector to P makes the angle θ with the z axis (foil normal), respectively. θ gives the direction of radiation and is defined by Eq. (1) of Sec. I. If r_0 is the distance of P from the origin then, as seen from Fig. 1, the distance of P from any other point in the $x-y$ plane is

$$r=[(r_0\cos\theta)^2+(r_0\sin\theta-x)^2+y^2]^{\frac{1}{2}}$$
$$\approx r_0-x\sin\theta+(x^2\cos^2\theta+y^2)/2r_0. \qquad (23)$$

As is also evident from Fig. 1, the transverse vector potential $A(P,t)$ at P makes the angle $\pi/2-\theta$ with the z axis. Therefore the retarded potential solution gives, with the substitution of Eqs. (22) and (23),

$$A(P,t)=c^{-1}\sin\theta\int\int\int J_z(x,\, t-r/c)r^{-1}dxdydz$$

$$\approx -i\left(\frac{\omega k\tau}{2\pi c r_0}\right)\varphi_0\sin\theta e^{-i\omega t}\int\int e^{i(kx+\omega r/c)}dxdy$$

$$\approx -i\left(\frac{\omega k\tau}{2\pi c r_0}\right)\varphi_0\sin\theta e^{-i\omega(t-r_0/c)}$$

$$\times\int\int\exp\left[i\left(\frac{\omega}{2cr_0}\right)(x^2\cos^2\theta+y^2)\right]dxdy. \quad (24)$$

Because of the thinness of the foil the variation of phase in the z direction has been neglected. The integrals are of the familiar form

$$\int_{-\infty}^{+\infty}\exp(iAu^2)du=(\pi i/A)^{\frac{1}{2}},$$

so that Eq. (24) reduces to

$$A(P,t)=k\tau\varphi_0\tan\theta e^{-i\omega(t-r_0/c)}. \quad (25)$$

From this expression the normal component of Poynting's vector averaged over time is easily found to be

$$S=\left(\frac{\omega^2k^2\tau^2}{8\pi c}\right)\varphi_0^2\sin\theta\tan\theta. \quad (26)$$

Since radiation is propagated from both sides of the foil, Eq. (26) gives one-half of the rate at which energy is lost, per unit area. To find the lifetime it is now only necessary to find the amount of energy actually stored in the oscillation described by Eq. (21).

Differentiating Eq. (21) and taking the real part gives an electric field of approximately $2k\varphi_0\cos(kx-\omega t)$, or an average energy per unit area stored in the electric scalar field of

$$U=\tau k^2\varphi_0^2/4\pi. \quad (27)$$

Because of the equality of potential and kinetic energy for a harmonic oscillator, U is just one-half of the total energy of oscillation per unit area. The radiative mean life τ_r is therefore given by

$$\tau_r^{-1}=S/U=(\omega^2\tau/2c)\sin\theta\tan\theta\approx\omega_p\left(\frac{\pi\tau}{\lambda_p}\right)\frac{\sin^2\theta}{\cos\theta}. \quad (28)$$

As an example, for the case of radiation at 30° ($\theta=\pi/6$) and τ equal to one-tenth λ_p, this formula yields $\tau_r^{-1}=0.09\omega_p$, a surprisingly fast decay rate. The mean life itself is $\tau_r=1.8(2\pi/\omega_p)$, or only about two periods of oscillation. Electronic transition rates in atoms are generally of the order of magnitude of 10^8 sec^{-1}, while the plasmon decay rate found here is of the order of magnitude of 0.1 ω_p, or 10^{15} sec^{-1}. This astonishingly large enhancement factor of 10^7 is caused by the extremely high degree of collectivity in the long wavelength plasmons. Because of the lack of dependence of the amplitude of the vector potential $A(P,t)$ on r_0 we can think of P approaching to within only a few wavelengths from the foil. It then becomes clear that a rough picture of the process is that all the electrons within a square of the foil one wavelength on a side contribute coherently to the radiation field. From the classical formula for the plasma frequency we have $\lambda_p=2\pi c/\omega_p=(\pi mc^2/ne^2)^{\frac{1}{2}}$, where n is the density of electrons in the metal, and e and m are the electron charge and mass. The number of electrons contained in the square is

$$\tau\lambda_p^2n=\pi(\tau/a_0)(h^2c^2/e^4)=\pi(137)^2(\tau/a_0)\approx 2\times 10^7, \quad (29)$$

for $\tau\approx 200$ A$\approx 400a_0$, (where a_0 is the Bohr radius of the hydrogen atom). This enormous number of electrons radiating coherently accounts for the collective enhancement, since the transition rate of a many-particle system is in general proportional to the number of particles participating in the transition. The situation in the metal foils is analogous to, but much more extreme than, the well-known collective enhancement of Coulomb excitation and gamma ray emission in the heavy nuclei.[15]

As θ is allowed to increase toward $\pi/2$ the decay rate given by Eq. (28) rises to excessive values, corresponding to a drastic broadening of the plasmon energy. On the other hand, as small values of θ are approached the plasmon energy sharpens up. The radiative decay rate decreases and eventually becomes comparable to the damping rate τ_d^{-1} due to interband transitions. When this happens the probability of the emission of a photon by any given plasmon is reduced from unity by the factor $\tau_r^{-1}/(\tau_r^{-1}+\tau_d^{-1})=(1+\tau_r/\tau_d)^{-1}$ because of the branching. Consequently the factor $\cos\theta$ in Eq. (3) of Sec. I, which gives the angular distribution of the radiation, must be replaced by the intensity function

$$I(\theta)=\cos\theta\left(1+\frac{1}{\tau_d\omega_p}\frac{\lambda_p}{\pi\tau}\frac{\cos\theta}{\sin^2\theta}\right)^{-1}. \quad (30)$$

The damping lifetime can be calculated from the optical constants of the metal along lines indicated by Fröhlich and Pelzer.[16] The complex index of refraction $n+ik$ is related to the dielectric constant by the

[15] Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. **28**, 432 (1956). A similar collective enhancement also occurs in the superradiant states of a gas [R. H. Dicke, Phys. Rev. **93**, 99 (1954) and A. Gamba, Phys. Rev. **110**, 601 (1958)].

[16] H. Fröhlich and H. Pelzer, Proc. Phys. Soc. (London) **A68**, 525 (1955).

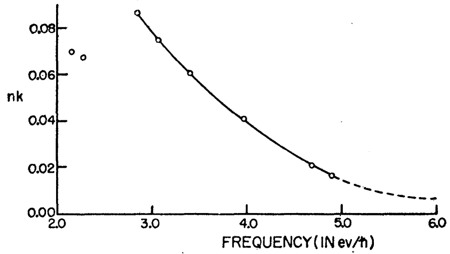

FIG. 2. Optical constant product nk for sodium as a function of frequency (in units of ev/$\hbar$). nk is one-half the imaginary part of the complex dielectric constant. The circles give the measured values of Ives and Briggs.[18] The dashed curve shows the extrapolation which yields $nk=0.006$ at the plasma frequency of $\omega_p=6$ ev/$\hbar$. The intrinsic mean life of a plasmon against interband damping is consequently 0.9×10^{-14} sec, or about thirteen periods of oscillation. Because of the arbitrary nature of the extrapolation this should be regarded only as a order-of-magnitude estimate. For photon emission angles greater than about 10° the radiative mean life is shorter than this estimate, so that the majority of the plasmons of the corresponding wavelengths decay by emitting a photon.

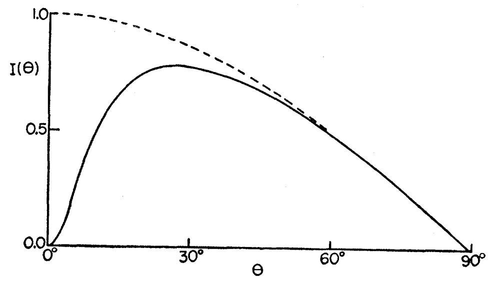

FIG. 3. Predicted photon intensity as a function of the angle θ from the foil normal, for a sodium film. Because of the competition of the interband damping with the radiative decay, the photon intensity falls below the ideal $\cos\theta$ distribution (dashed curve) at the smaller values of θ. In the limit of very small angles the radiative mean life varies as the inverse square of the angle, so that the corresponding very long wavelength plasmons decay almost entirely by intrinsic interband damping. The angle (here 10°) at which the intensity drops by a factor of two provides a determination of the intrinsic damping rate. Although the figure has been drawn for sodium, it should be regarded only as schematic, because of the uncertainty in the optical constants for sodium. (See Fig. 2.)

equation

$$\epsilon=(n+ik)^2=n^2-k^2+2ink. \tag{31}$$

For relatively ideal metals the imaginary part of ϵ, $2nk$, will be small, while the real part will be reproduced sufficiently accurately by the ideal expression appearing in Eq. (8). For $nk\neq0$ the frequency at which $\epsilon=0$ is shifted from the real axis to the value $\omega-i/2\tau_d$. Substituting from Eq. (8) into Eq. (31) gives

$$\epsilon=1-\omega_p^2(\omega-i/2\tau_d)^{-2}+2ink$$
$$\approx1-\frac{\omega_p^2}{\omega^2}-i\left(\frac{\omega_p^2}{\omega^3\tau_d}-2nk\right). \tag{32}$$

Thus the real part of the natural frequency of vibration of the system remains unshifted (to first order in nk) from $\omega=\omega_p$, while the imaginary part is fixed by

$$\tau_d^{-1}=2\omega_p nk. \tag{33}$$

This formula for the rate of damping by interband transitions has already been derived by Wolff[17] and is also discussed by Pines.[3] Both these authors seem, however, to have made an error of a factor of two. The expression they give is too small by this amount.

Unfortunately, the optical constants have not yet been measured in the necessary frequency range for most metals. Ives and Briggs[18] have, however, investigated the alkali metals and their data for sodium, which we shall now consider as a special case, are shown in Fig. 2. The value of nk is plotted *vs* the frequency times Planck's constant, measured in electron volts. Although Ives and Briggs covered a considerable frequency range, their highest frequency unfortunately still falls short of ω_p, making necessary the extrapolation shown in Fig. 2 by the dashed line. Since the plasmon energy amounts to 5.95 ev (Table I of reference 1), we find $nk=0.006$, $\tau_d^{-1}=0.012\omega_p$, and

$$\tau_d\approx0.9\times10^{-14}\text{ sec}. \tag{34}$$

This value should only be regarded as an order of magnitude estimate of the mean life with respect to interband damping. Because of the highly arbitrary nature of the extrapolation in Fig. 2, Eq. (34) could easily be in error by a factor of two in either direction. If, however, with this risk of error clearly in mind, we proceed to substitute τ_d from Eq. (34) into Eq. (30) we find that the critical quantity, the coefficient of $\cos\theta/\sin^2\theta$, takes on the value

$$\frac{1}{\tau_d\omega_p}\left(\frac{\lambda_p}{\pi\tau}\right)=\frac{2nk\lambda}{\pi\tau}=0.04. \tag{35}$$

Because of the smallness of this quantity it has an effect only at small angles, where we replace $\cos\theta$ by unity and $\sin\theta$ by θ. Thus, Eq. (30) becomes

$$I(\theta)\approx\frac{\cos\theta}{1+(\theta_0/\theta)^2}, \tag{36}$$

where $\theta_0\approx0.2$ radian$\approx10°$. This function is plotted in Fig. 3, which shows how the expected intensity distribution should follow a $\cos\theta$ law (shown as the dashed curve), except at the small angles where it drops below and passes to zero in the direction normal to the foil. θ_0 is characterized as the angle at which the intensity drops below the cosine curve by fifty percent. Hence experimental determination of θ_0, and consequently the value of nk at the plasmon frequency should be quite feasible.

An independent experimental test of the present theory would be provided by measuring the broadening

[17] P. Wolff, Phys. Rev. **92**, 18 (1953).
[18] H. E. Ives and H. B. Briggs, J. Opt. Soc. Am. **27**, 181 (1937).

of the radiation at various angles. The net mean life τ_t resulting from the total damping is determined by

$$\tau_t^{-1}=\tau_r^{-1}+\tau_d^{-1}. \quad (37)$$

As a consequence of the finiteness of τ_t the radiation at any angle θ will be spread over a range of frequency according to the extended distribution function

$$I(\theta;\omega)=I(\theta)[1+4\tau_t^2(\omega-\omega_p')^2]^{-1}. \quad (38)$$

The prime on ω_p indicates the slight shift determined in Sec. II for the normal oscillations. The half-width of the curve of intensity *vs* ω is just τ_t^{-1}. Measurement of the angle-dependent part of τ_t^{-1} should give, according to Eq. (37), an independent verification of Eq. (28). The irreducible constant part of the half-width, on the other hand, is just the interband damping breadth τ_d^{-1} and provides a check on the angular distribution method of determining this quantity. As shown above, these breadths are of the order of a few percent at the small angles. There should be no difficulty in accurately measuring them, even with a spectrometer of very modest resolution.

V. SUMMARY

Experimental verification of the radiation predicted in this paper would be of considerable interest, since it would constitute conclusive proof of the existence of plasma oscillations in metals. The required outlay in apparatus is very modest, consisting only of an electron gun, a photon detector, and a means of preparing the target film. Because the electron energy is not measured the equipment needs to be very much less elaborate than that required by the conventional energy loss experiment. Preparation of the target is the most critical part of the radiation experiment, since both the thickness and the structure of the film must be carefully controlled.

The characteristic angular dependence of both the photon intensity and the line breadth should make the identification of the radiation quite unique. It is possible from each of these measurements separately to derive the intrinsic interband damping rate of the metal. The experiment consequently contains an internal check. Although it may be argued that this damping rate, as well as the frequency of plasma oscillation, can be obtained from the optical constants, it should be borne in mind that experimental values of the latter in the necessary frequency range are virtually nonexistent at the present time. This situation is no doubt due to the technical difficulty of obtaining a satisfactory source of short-wavelength radiation. The experiment proposed here does not contain this difficulty, since the target is itself the source of the radiation. Thus the possibility arises that the radiation from oscillations in metal foils will not only give information on the optical constants of the target, but might also provide a practical source of radiation in the ultraviolet for more conventional optical experiments. Of course, difficult intensity problems might well be encountered here. In this connection it should be emphasized that the radiation should be expected on both sides of the foil. Such background radiation as that from bremsstrahlung might be minimized by placing the detector on the same side as the incident electron beam.

In conclusion, although there is as yet no experimental evidence for radiation from plasma oscillations in metal foils it is clear that under proper conditions this radiation must actually exist. The quantitative predictions made in this paper are the result of straightforward calculations based on well established physical principles. Although the calculations have been carried out classically, they can easily be put into quantum-mechanical garb with very minor modifications. This is because of the very simple nature of the harmonic oscillator. In particular, Eq. (28) for the radiative mean life remains completely unchanged.

APPENDIX I

In this Appendix a brief derivation will be given of the formula for the scattering coefficient of a thin metal film. This formula has already been exhibited in Sec. I as Eq. (6a). Although the problem can be handled completely quantum mechanically as in reference 1 the equivalent but simpler semiclassical approach of reference 9 will be used. Let us suppose that a normal plasma oscillation of amplitude sufficiently large to be treated classically is present in a square of the film of area A and thickness τ. Suppose in addition that the incident electrons are quantized in a rectangular parallelpiped of the same cross-sectional area but of the much greater thickness τ'. Thus the volume of quantization is $V'=A\tau'$, while the volume of the square of foil is $V=A\tau$. From the real part of Eq. (21) the energy of interaction of an incident electron [coordinates z and $\mathbf{x}=(x,y)$] with the plasma oscillation is

$$-e\varphi_0(e^{i\mathbf{k}\cdot\mathbf{x}-i\omega t}+e^{-i\mathbf{k}\cdot\mathbf{x}+i\omega t}) \times\begin{cases}\sinh kz, & |z|<\tau/2\\ \pm e^{-k|z|}e^{k\tau/2}\sinh(k\tau/2), & |z|>\tau/2,\end{cases} \quad (39)$$

where $\pm$ is the sign of z. Only the second term in the parentheses can cause the incident electron to lose energy. It is therefore convenient to abbreviate the coefficient of $e^{i\omega t}$ in Eq. (39) by H', which is thus the perturbing term in the Hamiltonian for the incident electron. We can now employ first order time-dependent perturbation theory to determine the transition rate caused by H'. According to Fig. 1 of reference 1 the incident electron makes a quantum jump from momentum state $\mathbf{p}$ to $\mathbf{p}-\hbar\mathbf{k}^{(3)}$. $\mathbf{k}^{(3)}$ is a three-dimensional vector whose x and y components, by conservation of momentum parallel to the foil, equal those of $\mathbf{k}$. The component of $\mathbf{k}^{(3)}$ on the other hand has its values concentrated about $\Delta k=\omega_p/v$, because of conservation of energy. The matrix element of H' between the initial and final states is therefore

$$\langle H'\rangle = V'^{-1}\int dzd^2\mathbf{x}\, e^{i(\Delta kz+\mathbf{k}\cdot\mathbf{x})}H'. \tag{40}$$

At this point it should be noted that it is of vital importance to include in Eq. (40) the contributions to the integral which come from the field outside the foil, as well as inside. Gabor[19] has studied the excitation of plasma oscillations in thin foils but failed to take into account the energy of interaction outside the foil. As a result he reached the erroneous conclusion that the probability of excitation is extremely small for very thin films. As we shall see below, the dependence on thickness is merely linear and not according to the higher power found by Gabor.

The right-hand member of Eq. (40) is most easily evaluated by considering the integral to be the energy of interaction of an effective incident electron density of $-e(V')^{-1}e^{i(\Delta kz+\mathbf{k}\cdot\mathbf{x})}$ with the electric scalar potential set up by the plasma oscillation. But by reciprocity of the Coulomb potential, this energy is the same as that of the charge density in the plasma oscillation integrated over the effective potential

$$\varphi_{\text{eff}} = -4\pi eV'^{-1}[(\Delta k)^2+k^2]^{-1}e^{i(\Delta kz+\mathbf{k}\cdot\mathbf{x})} \tag{41}$$

set up by the incident electron. The coefficient of $e^{i\omega t}$ in the charge density for a normal oscillation is simply a surface density at $z=\pm\tau/2$ of

$$\pm\frac{1}{4\pi}\left(1-\frac{1}{\epsilon}\right)D_z = \pm\frac{k\varphi_0}{4\pi}\left(1-\frac{1}{\epsilon}\right)\sinh(k\tau/2)e^{-i\mathbf{k}\cdot\mathbf{x}}$$
$$= \pm\left(\frac{k\varphi_0}{4\pi}\right)e^{k\tau/2}e^{-i\mathbf{k}\cdot\mathbf{x}} \approx \pm\left(\frac{k\varphi_0}{4\pi}\right)e^{-i\mathbf{k}\cdot\mathbf{x}},$$

where use has been made of Eq. (12). The charge density is therefore approximately

$$\rho = \left(\frac{k\varphi_0}{4\pi}\right)e^{-i\mathbf{k}\cdot\mathbf{x}}[\delta(z-\tau/2)-\delta(z+\tau/2)], \tag{42}$$

and Eq. (40) becomes

$$\langle H'\rangle = \int \varphi_{\text{eff}}\,\rho\, dzd^2\mathbf{x} = -\frac{2iek\varphi_0}{\tau'[(\Delta k)^2+k^2]}\sin(\Delta k\tau/2). \tag{43}$$

It is now necessary to find the degree of quantum excitation to which our classical oscillation of amplitude φ_0 corresponds. If the plasma oscillator is in its nth excited state the excitation energy is $n\hbar\omega_p$. Equation (27) gives one-half the energy per unit area calculated classically. Imposing the *correspondence principle* yields

$$n = Vk^2\varphi_0^2/(2\pi\hbar\omega_p). \tag{44}$$

Because of the special nature of the harmonic oscillator, the matrix element for creating a plasmon from the ground state, which we designate by $H_{\mathbf{k}}'$ is $(n+1)^{-\frac{1}{2}}$ times $\langle H'\rangle$. Since we have assumed $n\gg 1$, we obtain

$$|H_{\mathbf{k}}'|^2 = \left(\frac{8\pi e^2\hbar\omega_p}{\tau\tau' V'[(\Delta k)^2+k^2]^2}\right)\sin^2(\Delta k\tau/2). \tag{45}$$

It remains merely to multiply Eq. (45) by $2\pi/\hbar$ times the density of states

$$\rho(E) = V'p^2 d\Omega/vh^3. \tag{46}$$

According to first-order perturbation theory, this gives the rate of scattering into the infinitesimal solid angle $d\Omega$. Dividing by $d\Omega$ times the rate at which electrons impinge on the film, v/τ', yields the scattering coefficient

$$\mu(\alpha;\tau) = \frac{\tau}{2\pi a_0}\left(\frac{\hbar\omega_p}{mv^2}\right)\left(\frac{\theta_E}{\alpha^2+\theta_E{}^2}\right)^2\left(\frac{\sin\pi\tau/\lambda_{\text{ex}}}{\pi\tau/\lambda_{\text{ex}}}\right)^2 \tag{47}$$

where Δk has been put in terms of λ_{ex}. An equivalent form of Eq. (47) appears in Eq. (6a) and has been used in Sec. I to compute the photon yield.

[19] D. Gabor, Phil. Mag. 1, 1 (1956). The present author feels that Gabor's attempt to give a complete account of all the types of plasma oscillations excited in thin films is premature, because of the considerable uncertainty which exists concerning the electron distribution in these special modes of oscillation. It is by no means correctly represented by his simple cosine dependence as is clear from the oscillations studied here, which involve only surface charges. In any case it should be noted that the spurious cross-shaped scattering pattern he finds arises from the arbitrary omission of certain modes, and is not absent for the reason he gives. Also, there seems to be a numerical error in his discussion of the function he defines in his Eq. (24). The sum can be evaluated in closed form as $F(x)=\frac{1}{2}[1-(\pi x/2)^{-2}\sin^2(\pi x/2)]$. Consequently the asymptotic value of $F(x)$ is $\frac{1}{2}$ and not 0.481. Finally, it must be remarked that Gabor's use of time-dependent perturbation theory has been criticized by many authors (see, for example, reference 13). We likewise disagree with Gabor's statements concerning coherence, and claim that the theory of the scattering of plane-wave incident electrons gives essentially a complete description of the phenomenon. The scattering coefficient for less ideal experimental situations is found by making incoherent summations over the ideal plane-wave cases. (In this connection, the author wishes to acknowledge valuable discussion with R. H. Dicke.)