Optical Emission from Irradiated Foils. I

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The present work is concerned with the emission of optical radiation from foils irradiated with charged particles. A generalization of the Ginsburg-Frank treatment is presented in which the wave properties of normally incident charged particles are accounted for in the Born approximation. The foil is assumed to be of finite thickness and to possess a dielectric constant of the form $\epsilon(\omega)=\epsilon_1(\omega)+i\epsilon_2(\omega)$. Detailed prediction of the energy and angle distribution of emitted photons is made. The present result is shown to reduce to that obtained by Ferrell if $\epsilon(\omega)$ appropriate to a free electron gas is assumed and if one takes $v_i/c \ll 1$, where v_i is the speed of the incident particle. Numerical results have been obtained for foils of Ag and Al.

I. INTRODUCTION

THE dynamic many-particle interaction between electrons in solids has been the subject of much interesting work in the past few years. In particular, the concept of plasma oscillation in metals has been developed and extended by Pines and Bohm and subsequent workers[1] to the point where many aspects of the behavior of conduction-band electrons have been made clear. Characteristic losses by charged particles in solids have been explored extensively by many experimenters, and it is apparent that the plasma concept has contributed greatly to the understanding of these losses.

It now seems clear that plasmons exist as well-defined quasiparticle excitations in the electronic systems of solids. Evidence supporting this fact comes primarily from characteristic loss experiments in metals in which the value of this energy loss is found to correlate well with the plasmon energy calculated from the valence band electron density. In addition, the dependence of the characteristic energy loss upon the angular deviation of the charged particle undergoing this loss is predicted well from the plasma theory of Pines and Bohm. Pines has also interpreted energy loss spectra in a wide variety of solids as well as in metals in terms of plasmon excitation.

Following the early interpretation of this sort by Pines[2] on the basis of the Pines-Bohm theory, one of the present authors[3] postulated that certain low-lying losses which had been observed in the characteristic loss spectrum of several metals were due to plasma oscillations occurring near the foil surfaces. In this case one expects a depolarizing effect which, if the surface is planar, tends to cause the eigenfrequency of surface oscillations to be less than that of oscillations occurring in the volume of the metal by a factor of $1/\sqrt{2}$. Characteristic losses of electrons in creating this sort of excitation are said to be due to the creation of "surface plasmons" as opposed to the creation of "volume plasmons" at the full characteristic energy loss value. Recent theoretical work extending this approach to multilayered systems has been carried out by Stern and Ferrell.[4] This prediction has been borne out in a series of elegant experiments carried out by Powell and Swan and co-workers[5] who observed inelastic losses experienced by electrons reflected from newly evaporated layers of Al and Mg. They found the volume-plasmon loss at the expected value of $\hbar\omega_p$ where ω_p is the free-electron plasma frequency appropriate to each metal, and a surface-plasmon loss at the predicted lowered value of $\hbar\omega_p/\sqrt{2}$. They found after an operating time of the order of minutes that the surface-plasmon loss at $\hbar\omega_p/\sqrt{2}$ disappears and is replaced by a loss at an even lower energy. Stern and Ferrell[4] have explained this phenomenon in both Al and Mg by postulating that a thin oxide layer, with dielectric constant greater than unity, forms on the surface of each metal and that the additional depolarizing effect of this layer is great enough to account for the new loss line. They obtain quantitative agreement between theory and experiment in that the dielectric constant of the oxide layer necessary to account for the lowered loss value is quite consistent with measured values of this constant for the oxides. They also are able to show that a layer of only 20 Å thickness may cause the observed effect in the two metals considered. Pines[1] has reviewed the available experimental data on surface-plasmon losses and suggests that from them one may determine whether there is an appreciable admixture of relatively large energy interband transitions in the loss line, and that one may determine the energies and oscillator strengths associated with these interband transitions. The existence of these surface-plasmon losses in a given metal serves to establish firmly the collective nature of electron dynamics in that metal.

Still another method for unravelling the details of electron excitation in solids has been proposed by

* Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

[1] For the most recent review of theoretical and experimental developments in this field, see D. Pines, *Proceedings of the International Conference on Many-Body Problems*, Utrecht, Supplement to Physica, **26**, December, 1960.

[2] D. Pines, Revs. Modern Phys. **28**, 184 (1956).

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

[4] E. A. Stern and R. A. Ferrell, Phys. Rev. **120**, 130 (1960).

[5] C. J. Powell, J. L. Robin, and J. B. Swan, Phys. Rev. **110**, 657 (1958); C. J. Powell and J. B. Swan, *ibid.* **115**, 869 (1959); **116**, 81 (1959); **118**, 640 (1960); C. J. Powell, Australian J. Phys. **13**, 145 (1960); C. J. Powell, Proc. Phys. Soc. (London) **76**, 593 (1960).

Ferrell.[6] He has predicted that a plasmon generated in a metal foil, e.g., by charged-particle irradiation, may decay by the emission of a transverse photon which could be detected experimentally. He has drawn an analogy between this process and nuclear Coulomb excitation by incident-charged particles followed by the detection of the fluorescent gamma ray. For the case of a metallic plasma, he makes a detailed prediction of the angular distribution of Coulomb-stimulated photons emitted at the plasmon energy, as well as the dependence of the number of photons upon the foil thickness and energy of the incident electron. The physics of this process is discussed in a very illuminating manner. The results are of great value and have stimulated much experimental and theoretical work in this field even though it is pointed out in his paper that retardation effects are not included in a consistent way in his theory.

A theoretical treatment of the process by which "transition radiation" is emitted when a classical point-charged particle crosses a plane interface between two media of differing dielectric constants has been given by Frank and Ginsburg[7] and subsequently extended by many workers.[8,9] Goldsmith and Jelley[10] have observed this radiation at wavelengths in the visible range from protons incident on Al, Ag, and Au surfaces. The observations were carried out near the tangent to the foil, and the protons were incident nearly normal to the surface. The observers found good agreement between the number of photons in the polarized component of the emitted radiation, and that predicted by the theory of Frank and Ginsburg, if one assumes that the metals have infinite conductivity.

At the suggestion of Ferrell, there has been a considerable amount of work by several different experimental groups on the optical emission from Ag, where one expects a peaking at $\sim$3400 Å. Steinmann[11] found a maximum intensity at about the expected wavelength and also found that the magnitude of this maximum tends to oscillate as the foil thickness is increased, much as predicted by Ferrell.[6] Steinmann interprets his results as conclusive evidence for the existence of plasmons of 3.75 ev in Ag. Brown, Wessel, and Trounson[12] have also presented evidence in favor of the plasmon decay interpretation showing an angular dependence of the radiation at 3400 Å which is in general agreement with Ferrell's predictions.

More recently a paper by Boersch, Radeloff, and Sauerbrey[13] has appeared in which the spectra from thick foils of a number of different metals are reported. Comparisons are made between their experimental data and the Frank-Ginsburg theory for thick foils. Observations of polarization, spectra, angular distribution, and dependence of photon emission on beam energy have recently been reported by Arakawa *et al.*[14] and are reported in detail in a companion paper.[15]

It is clear that the considerations of Ferrell, and of Frank and Ginsburg are directed at different aspects of the same problem and that there is an intimate relation between them. The present work is concerned with a generalization of the Frank-Ginsburg treatment to exhibit explicitly the wave nature of an incident charged particle in the Born approximation. The particle is assumed to be normally incident on a foil of thickness a, characterized by a general dielectric constant $\epsilon(\omega)$. Detailed predictions of the joint energy-angle distribution of photons emitted in the process are given explicitly for certain idealized forms of $\epsilon(\omega)$. The connection between the treatment of Ferrell and that of the present authors is discussed in some detail. It is shown that one expects photon emission from a solid not only at energies in the neighborhood of the plasmon energy (in the case of collective excitations in the solid), but also around the interband transition energy when one-particle excitations occur. The effect of a thin oxide film on the photon spectrum is considered briefly. A coupling between the most probable energy in the distribution of emitted photons and the angle of observation is predicted for thick foils. A digital computer code has been written to evaluate the emitted-photon distribution function for the thin foil case. Numerical results have been obtained for two different metals for which optical measurements of $\epsilon(\omega)$ are available.

It should be mentioned that considerable attention has been given to various special cases of the transition radiation phenomenon, especially in the Russian literature. In particular, Eq. (14) of the present paper, giving the photon distribution from a thin foil, has been derived[16] using an approach somewhat different from that of the present paper. However, the present approach is more general since the wave properties of

[6] R. A. Ferrell, Phys. Rev. **111**, 1214 (1958).

[7] I. M. Frank and V. I. Ginsburg, J. Phys. (U.S.S.R.) **9**, 353 (1945).

[8] The Russian literature contains many papers on this subject, e.g., see G. M. Garibian, J. Exptl. Theoret. Phys. (U.S.S.R.) **33**, 1403 (1957) [translation: Soviet Phys.—JETP **6**, 1079 (1958)]; J. Exptl. Theoret. Phys. (U.S.S.R.) **35**, 1435 (1958) [translation: Soviet Phys.—JETP **8**, 1003 (1959)]; J. Exptl. Theoret. Phys. (U.S.S.R.) **37**, 527 (1959) [translation: Soviet Phys.—JETP **10**, 372 (1960)]; G. M. Garibian and G. A. Chalikyan, J. Exptl. Theoret. Phys. (U.S.S.R.) **35**, 1282 (1958) [translation: Soviet Phys.—JETP **8**, 894 (1959)]; V. E. Pafomov, J. Exptl. Theoret. Phys. (U.S.S.R.) **33**, 1074 (1957) [translation: Soviet Phys.—JETP **6**, 829 (1958)]; J. Exptl. Theoret. Phys. (U.S.S.R.) **39**, 134 (1960) [translation: Soviet Phys.—JETP **12**, 97 (1961)].

[9] R. H. Ritchie and H. B. Eldridge, Bull. Am. Phys. Soc. 4, 384 (1959), Oak Ridge National Laboratory, Health Physics Division Annual Progress Report ORNL-2806, 1959 (unpublished), p. 133.

[10] P. Goldsmith and J. V. Jelley, Phil. Mag. **4**, 836 (1959).

[11] W. Steinmann, Phys. Rev. Letters **5**, 470 (1960); Z. Physik. **163**, 92 (1961).

[12] R. W. Brown, P. Wessel, and E. P. Trounson, Phys. Rev. Letters **5**, 472 (1960).

[13] H. Boersch, C. Radeloff, and G. Sauerbrey, Phys. Rev. Letters **7**, 52 (1961).

[14] E. T. Arakawa, A. L. Frank, R. D. Birkhoff, and R. H. Ritchie, Bull. Am. Phys. Soc. **6**, 266 (1961).

[15] A. L. Frank, E. T. Arakawa, and R. D. Birkhoff, following paper [Phys. Rev. **126**, 1947 (1962)].

[16] V. E. Pafomov, J. Exptl. Theoret. Phys. (U.S.S.R.) **39**, 134 (1960) [translation: Soviet Phys.—JETP **12**, 97 (1961)].

the incident-charged particles are considered explicitly. In addition, detailed examination of the photon-distribution function for various forms of the dielectric constant is made and numerical results are presented for some interesting cases.

II. DIELECTRIC THEORY OF THE FOIL RESPONSE

It is well known that an energetic charged particle is equivalent for many purposes to a nearly "white" source of photons. Hence one might consider the present problem to be that of investigating the reflection and transmission by a foil of photons having a spectral distribution characteristic of a swiftly-moving electric charge. In this view, the advantage of charged particles in the present connection lies in the fact that they are easily obtainable sources of high-energy photons of known spectral composition.

The dynamical response of the electronic system of the solid will be codified in a dielectric constant which depends upon the applied frequency, but not upon the wave vectors of the fields. This is known to give accurate results for long wavelength excitations in the solid.[2] For short wavelength disturbances, involving large-momentum transfers to the solid, collective effects become relatively unimportant and in the limit of short wavelengths only individual interactions between the incident particle and electrons in the foil need be considered. In the present connection, collective effects are of prime importance.

In this section we consider the electromagnetic field generated throughout space when a charged particle makes a transition from a given plane wave state of kinetic energy E_i to one of lower energy. The fluctuating charge density due to this transition induces currents in a foil. The system of total currents gives rise to transverse-electromagnetic waves. The flux of the Poynting vector in the far zone is computed, and from this the distribution of photons in angle and frequency is obtained from the correspondence principle. Such a semiclassical description of the process is valid as long as one confines his attention to photon energies $\hbar\omega \ll mc^2$ and as long as $\hbar\omega \ll E_i$. The wave properties of the incident particle are considered in order to bring out clearly the momentum and energy conditions which must be satisfied in the system.

The foil is assumed to lie in the region $0 \leq z \leq a$ and to extend from $-L/2$ to $+L/2$ in both the x and y directions. The normalization volume of the incident particle wave function is assumed to be bounded by the foil edges in the x and y directions and to extend from $-v_iT/2$ to $+v_iT/2$ along the z axis. The initial velocity of the incident particle, $\mathbf{v}_i$, has magnitude v_i. Both L and v_iT are considered to be $\gg a$. The time interval T is taken to be much greater than all electronic periods to be considered. This unconventional normalization is chosen in order that one particle shall strike the surface of the foil in time interval T.

The wave function, ψ_i, of the incident particle may be written as a positive-energy solution of the Dirac equation for a free particle, i.e.,

$$\psi_i = u(\mathbf{p}_i)\exp[i(\mathbf{p}_i\cdot\mathbf{r} - \omega_i t)]/L(v_iT)^{\frac{1}{2}},$$

where, as usual,

$$\mathbf{p}_i = m\mathbf{v}_i/\hbar[1-(v_i/c)^2]^{\frac{1}{2}},$$
$$\omega_i = [m^2c^4/\hbar^2 + p_i^2c^2]^{\frac{1}{2}},$$

and $u(\mathbf{p}_i)$ is the Dirac spinor. The current density generated by a particle of charge Ze in a transition to a final state of positive energy characterized by a wave vector $\mathbf{p}_f$ may be written

$$\mathbf{j} = Zec\psi_f^* \boldsymbol{\alpha}\psi_i, \tag{1}$$

where $\boldsymbol{\alpha}$ is the well-known Dirac matrix, and it will be assumed that the particle recoil $|\mathbf{p}| = |\mathbf{p}_i - \mathbf{p}_f| \ll |\mathbf{p}_i|$. We neglect spin-flip in the transition and only the current component perpendicular to the foil surface will be considered here.

Electromagnetic fields arising from the transition charge density represented by Eq. (1) have now to be computed. It will be convenient to write Maxwell's equations in terms of the Hertz vector $\mathbf{\Pi}(\mathbf{r},t)$.[17] If the x, y, and t dependence of both $\mathbf{\Pi}$ and the current density vector $\mathbf{j}$ are written in terms of Fourier series with period equal to the dimensions of the normalization volume,

$$\left\{\begin{matrix}\mathbf{\Pi}(x,y,z,t)\\ \mathbf{j}(x,y,z,t)\end{matrix}\right\} = \frac{1}{L^2T}\sum_{n_x}\sum_{n_y}\sum_{n_t} e^{i(xk_x+yk_y-\omega t)} \times \left\{\begin{matrix}\mathbf{\Pi}_{k_x,k_y,\omega}(z)\\ \mathbf{j}_{k_x,k_y,\omega}(z)\end{matrix}\right\}, \tag{2}$$

where

$$k_x = (2\pi/L)n_x, \quad k_y = (2\pi/L)n_y, \quad \omega = (2\pi/T)n_t.$$

The wave equation relating $\mathbf{\Pi}$ and $\mathbf{j}$ reads

$$\frac{d^2}{dz^2}\mathbf{\Pi}_{\mathbf{K},\omega}(z) + \{\epsilon\omega^2/c^2 - K^2\}\mathbf{\Pi}_{\mathbf{K},\omega}(z) = -\frac{4\pi i}{\omega\epsilon(\omega)}\mathbf{j}_{\mathbf{K},\omega}(z), \tag{3}$$

when $0 < z < a$. The quantity $K = (k_x^2 + k_y^2)^{\frac{1}{2}}$. When z lies outside this region, one has only to set $\epsilon = 1$ in Eq. (3) to correspond to the case where vacuum bounds the foil on both sides. The fields are related to $\mathbf{\Pi}$ by the equations

$$\mathbf{E} = \nabla(\nabla\cdot\mathbf{\Pi}) + (\epsilon\omega^2/c^2)\mathbf{\Pi} \quad \text{and} \quad \mathbf{H} = (i\omega\epsilon/c)\nabla\times\mathbf{\Pi}.$$

It is understood that Eq. (3) refers to the z components of $\mathbf{\Pi}$ and $\mathbf{j}$, which are the only nonvanishing ones in this approximation.

The Fourier transform of Eq. (1) yields the following expression for the magnitude of $\mathbf{j}_{\mathbf{K},\omega}(z)$,

$$j_{\mathbf{K},\omega}(z) = Ze\delta_{p_x,k_x}\delta_{p_y,k_y}\delta_{\omega_{if},\omega}e^{izp_z}, \tag{4}$$

[17] J. S. Stratton, *Electromagnetic Theory* (McGraw-Hill Book Company, New York, 1941), p. 573.

where p_x, p_y, and p_z are the components of $\mathbf{p}=\mathbf{p}_i-\mathbf{p}_f$ and $\omega_{if}=\omega_i-\omega_f$.

The boundary conditions of continuity of the tangential components of $\mathbf{E}$ and $\mathbf{H}$ leads to the following relations between the z components of $\mathbf{\Pi}$ on either side of a boundary:

$$\epsilon(\omega)[\mathbf{\Pi}_{\mathbf{K},\omega}(z)]_{zi}=[\mathbf{\Pi}_{\mathbf{K},\omega}(z)]_{zo}, \tag{5a}$$

$$\left[\frac{d}{dz}\mathbf{\Pi}_{\mathbf{K},\omega}(z)\right]_{zi}=\left[\frac{d}{dz}\mathbf{\Pi}_{\mathbf{K},\omega}(z)\right]_{zo} \tag{5b}$$

where the zi (zo) subscript indicates the z component of $\mathbf{\Pi}_{\mathbf{K},\omega}(z)$ or its derivative as z approaches the boundary from inside (outside) the foil.

The solution of Eq. (3) and the corresponding equation valid outside the foil may be written:

$$\Pi_{K,\omega}(z)=Ae^{\nu z}+\Lambda e^{izp_z}, \qquad -\infty<z<0 \tag{6a}$$

$$=Be^{\nu' z}+Ce^{-\nu' z}+\Lambda' e^{izp_z}, \quad 0<z<a \tag{6b}$$

$$=De^{-\nu z}+\Lambda e^{izp_z}, \qquad a<z<\infty \tag{6c}$$

where

$$\Lambda=(4\pi Ze/i\omega)\Gamma/(\nu^2+p_z^2)=\Gamma\lambda,$$

$$\Lambda'=[4\pi Ze/i\omega\epsilon(\omega)]\Gamma/(\nu'^2+p_z^2)=\Gamma\lambda'$$

$$\Gamma=\delta_{p_x,k_x}\delta_{p_y,k_y}\delta_{\omega_{if},\omega},$$

and $\nu=(K^2-\omega^2/c^2)^{\frac{1}{2}}$, $\nu'=[K^2-\epsilon(\omega)\omega^2/c^2]^{\frac{1}{2}}$. The quantities A, B, C, and D are to be evaluated by imposing the conditions of Eq. (5) at each boundary. Both ν and ν' are understood to have a positive real part in all subsequent manipulations in order that $\mathbf{\Pi}$ be bounded as $z\rightarrow\pm\infty$. Since the main interest resides in fields at points remote from the foil, i.e., at distances large compared with the wavelength of any photons to be considered, it is necessary only to quote results for the quantities A and D. The terms in Eq. (6) containing Λ and Λ' explicitly are clearly to be interpreted as forcing terms, representing that part of the field uninfluenced by boundaries. One finds

$$\begin{aligned}D(\mathbf{K},\omega,\mathbf{p})=(\Gamma/\Delta)e^{\nu a+iap_z}\\ \times\{[(\nu'-i\epsilon p_z)\lambda-\epsilon(\nu'-ip_z)\lambda'](\nu\epsilon+\nu')e^{\nu' a}\\ +[(\nu'+i\epsilon p_z)\lambda-\epsilon(\nu'+ip_z)\lambda'](\nu\epsilon-\nu')e^{-\nu' a}\\ -2\nu'\epsilon[(\nu-ip_z)\lambda-(\nu\epsilon-ip_z)\lambda']e^{-iap_z}\},\end{aligned} \tag{8a}$$

and

$$A(\mathbf{K},\omega,\mathbf{p})=e^{-\nu a+iap_z}[D(\mathbf{K},\omega,\mathbf{p})]_{p_z\rightarrow -p_z}, \tag{8b}$$

where

$$\Delta=(\nu\epsilon-\nu')^2e^{-\nu' a}-(\nu\epsilon+\nu')^2e^{\nu' a}.$$

Consider the form of the Hertz vector in the region $a\ll z<\infty$. Letting $D=e^{\nu a}D'$, one may write:

$$\Pi_\omega(x,y,z)\equiv\frac{1}{L^2}\sum_{k_x,k_y}e^{i(xk_x+yk_y)-\nu(z-a)}D'(\mathbf{K},\omega,\mathbf{p})$$

$$=\frac{1}{(2\pi)^2}\int_0^{2\pi}d\varphi\int_0^\infty KdK\, e^{iK\rho\cos(\varphi-\varphi_0)-\nu(z-a)}D'(\mathbf{K},\omega,\mathbf{p}),$$

where

$$\rho=(x^2+y^2)^{\frac{1}{2}}.$$

In the last step it is understood that the limits $L\rightarrow\infty$, $T\rightarrow\infty$, have been approached so that the sums become integrals and the Kronecker δ functions in Γ are to be interpreted as Dirac δ functions, i.e.,

$$\Gamma\rightarrow\frac{(2\pi)^3}{L^2T}\delta(p_x-k_x)\delta(p_y-k_y)\delta(\omega_{if}-\omega).$$

Making the substitution $K=(\omega/c)\sin\theta$ in Eq. (8), one has

$$\nu=(K^2-\omega^2/c^2)^{\frac{1}{2}}=-(i\omega/c)\cos\theta,$$

where the signs of K and ν are chosen in order that the asymptotic form of Π shall correspond to a diverging spherical wave. Then one arrives at the well-known form[17]

$$\Pi_\omega=\frac{\omega^2}{4\pi^2c^2}\int_0^{2\pi}d\varphi\int_0^{\frac{1}{2}\pi-i\infty}\cos\theta\sin\theta d\theta \times\exp\left(i\frac{R\omega}{c}\cos\Theta\right)D', \tag{9}$$

where

$$\cos\Theta=\cos\theta\cos\theta_0+\sin\theta\sin\theta_0\cos(\varphi-\varphi_0),$$

$$R\cos\theta_0=z,\quad R\sin\theta_0\cos\phi_0=x\quad\text{and}\quad R\sin\theta_0\sin\varphi_0=y.$$

One now considers that $R\omega/c\gg 1$, so that the points φ and φ_0 and $\theta=\theta_0$ become points of stationary phase in the integration of Eq. (9). The exponential term oscillates rapidly as θ and φ move away from these points, so that to an approximation which improves the larger R becomes one may take other factors out of the integral after evaluating them at the points of stationary phase. Then carrying out the integrals over θ and φ, in the case $(\sin^2\theta_0)(\omega R/c)\gg 1$,

$$\Pi_\omega=(\omega/2\pi ic)(\cos\theta_0 D')(e^{iR\omega/c}/R). \tag{10}$$

$\mathbf{E}$ and $\mathbf{H}$ are related to π by the expressions

$$\mathbf{E}=\nabla\left(\frac{\partial\Pi}{\partial z}\right)+\frac{\omega^2}{c^2}\hat{k}\Pi,$$

$$\mathbf{H}=-(i\omega/c)\nabla\times(\hat{k}\Pi),$$

where $\hat{k}$ is a unit vector in the direction of z. In order to calculate S_R, the Poynting flux of energy at large distances from the foil, one sets

$$S_R=\frac{c}{4\pi}\int_{-\infty}^{\infty}E_{\theta_0}H_{\varphi_0}dt, \tag{11}$$

where the subscripts on θ_0 and φ_0 are dropped henceforth. Calculating the indicated components of $\mathbf{E}$ and $\mathbf{H}$ in the wave zone, one finds

$$E_\theta=H_\varphi=-(\omega^2/c^2)\sin\theta\Pi_\omega,$$

and after carrying out the integral over time indicated in Eq. (11), one finds for the total flux per unit area in the direction of R

$$S_R=\frac{1}{R^2}\int_0^\infty d\omega\,\frac{\omega^6}{16\pi^4c^5}\sin^2\theta\cos^2\theta\tilde{D}(\omega)\tilde{D}(-\omega)$$
$$\equiv\frac{1}{R^2}\int_0^\infty d\omega s_R(\omega), \quad (12)$$

where

$$\tilde{D}(\omega)\equiv[D'(\mathbf{K},\omega,\mathbf{p})]_{K=(\omega/c)\sin\theta}.$$

Since the area dA intercepted on a sphere of radius R by the element of solid angle $d\Omega$ is $R^2d\Omega$, one may set $s_RdA=s_RR^2d\Omega$. Then, recalling that the energy flux is given by the product $\hbar\omega$, and the number flux at the frequency ω, one writes

$$\frac{d^2N}{d\omega d\Omega}=\frac{1}{\hbar\omega}s_R\frac{dA}{d\Omega}=\frac{\omega^5}{16\pi^4\hbar c^5}\sin^2\theta\cos^2\theta\tilde{D}(\omega)\tilde{D}(-\omega),$$

where $d^2N/d\omega d\Omega$ is the number of photons emitted per unit frequency interval at frequency ω and per unit solid angle in the direction θ, with respect to the normal to the foil. There remains only the step of summing over final states of the charged particle in order to obtain the distribution of photons for all allowed transitions. In order to carry out this step, one notes that $d^2N/d\omega d\Omega$ corresponds to the absolute square of a matrix element between initial and final states and thus should be written as

$$\frac{d^2N}{d\omega d\Omega}=\frac{\omega^5}{16\pi^4\hbar c^5}\sin^2\theta\cos^2\theta|\tilde{D}(\omega)|^2\Gamma^2. \quad (13)$$

The sum over final states is given by

$$\sum_{\mathbf{p}_f}\rightarrow\frac{L^2Tv_i}{(2\pi)^3}\int d\mathbf{p}_f,$$

in the limit as $T,\,L\rightarrow\infty$. In this same limit, one may write

$$\Gamma^2\rightarrow\frac{(2\pi)^3}{L^2T}\delta(p_x-k_x)\delta(p_y-k_y)\delta(\omega_{if}-\omega).$$

Carrying out the integrals over $\mathbf{p}_f$ and using the fact that $\omega_{if}\simeq v_ip_z$ if $|\mathbf{p}|\ll|\mathbf{p}_i|$, one finds for n, the number of photons emitted per unit solid angle in the direction specified by the angle θ and per unit frequency interval at the frequency ω per incident particle,

$$n(\mu,\omega,a,\beta)\equiv\sum_{\mathbf{p}_f}\frac{d^2N}{d\omega d\Omega}=\frac{Z^2\alpha\beta^2}{\pi^2\omega}\mu^2(1-\mu^2)\left|\frac{\gamma}{\Delta}\right|^2, \quad (14)$$

where

$$\gamma=\left\{\left[\frac{\beta\sigma+\epsilon}{1-\mu^2\beta^2}-\frac{1}{1-\beta\sigma}\right](\mu\epsilon+\sigma)e^{-it\sigma}\right.$$
$$+\left[\frac{\beta\sigma-\epsilon}{1-\beta^2\mu^2}+\frac{1}{1+\beta\sigma}\right](\mu\epsilon-\sigma)e^{it\sigma}$$
$$\left.-2\sigma\left[\frac{\epsilon}{1-\beta\mu}-\frac{1+\beta\epsilon\mu}{1-\beta^2\sigma^2}\right]e^{-it/\beta}\right\}, \quad (15)$$

and $\sigma=(\epsilon-1+\mu^2)^{\frac{1}{2}}$, $\beta=v_i/c$, $\mu=\cos\theta$, $t=a\omega/c$, $\Delta=(\mu\epsilon-\sigma)^2e^{it\sigma}-(\mu\epsilon+\sigma)^2e^{-it\sigma}$ and $\alpha=e^2/\hbar c$ is the fine structure constant. One may show that the equation for the radiation into the half-space $z<0$ is nearly identical with that given by Eq. (14); it is only necessary to replace μ by $|\cos(\theta)|$, and to put $\beta\rightarrow-\beta$ in Eqs. (14) and (15). In the general case $\epsilon=\epsilon_1+i\epsilon_2$, and one must put $\sigma=R(\cos\Phi+i\sin\Phi)$ with $R=[(\epsilon_1-1+\mu^2)^2+\epsilon_2{}^2]^{\frac{1}{4}}$ and $\Phi=\frac{1}{2}\tan^{-1}[\epsilon_2/(\epsilon_1-1+\mu^2)]$.

The momentum and energy conserving factors in the expression for Γ show clearly that in a given transition from an initial state with momentum perpendicular to the foil surface, the momentum lost by the incident-charged particle in a direction parallel to the foil face must be exactly equal to the component of momentum carried off by the photon in that direction, i.e., $(\omega/c)\cos\theta$. However, the z component of the photon momentum $(\omega/c)\sin\theta$, is not in general equal to the momentum lost by the charged particle in that direction. Clearly, the foil surfaces participate in the process and take up the momentum difference. Then the magnitude of the z momentum of the photon must be less than ω/v_i, the z momentum loss of the charged particle. Thus $(\omega/v_i)\geqslant(\omega/c)|\cos\theta|$, or $\beta|\cos\theta|\leqslant1$, which is always satisfied.

Thus, the present phenomenon is roughly the inverse of the surface photoemission process discussed by Mitchell[18] in which foil surfaces may participate in the photoabsorption and electron-emission process. Momentum and energy may thereby be conserved simultaneously, and the ejection of an electron from the metal may occur when a photon is absorbed. In the photoemission process, however, the electron makes a transition from the conduction band to an energetic state outside of the metal; while in the case considered here, the incident electron goes from one continuum state to another in the process of coupling with the electromagnetic field and with electrons in the solid.

Equation (13), as it stands, would predict the distribution in angle of photons of energy $\hbar\omega$ omitted when a charged particle makes a transition to a particular final state, i.e., is deflected through a given angle with energy loss $\hbar\omega$. On the other hand, if all possible angular deflections and energy losses of the incident particle are permitted, ω and θ may be regarded as independent

[18] K. Mitchell, Proc. Roy. Soc. (London) **A146**, 442 (1934).

variables, the joint distribution of which is predicted by Eq. (14).

The incident charged particle undergoes angular deflection and energy loss in the process of generating photons and, as well, in exciting electronic transitions in the solid which decay without coupling with the electromagnetic field. Although such energy losses are not of primary concern here, they are interesting in connection with the recent experimental and theoretical work on surface-plasmon excitation summarized briefly in the Introduction and described more completely in reference 1. For this reason, the distribution of energy loss and momentum change by the charged particle as it interacts with a thick foil and the electromagnetic field is presented and discussed briefly in the Appendix.

The polarization of the photons described by Eq. (14) is entirely in the plane containing the foil normal and the propagation vector of the photon. This is distinctly different from the polarization of bremsstrahlung emitted during nuclear encounters in the foil. It has been shown[19] that bremsstrahlung photons at optical frequencies are nearly unpolarized for angles $\theta \ll 1$, and are polarized primarily in a direction perpendicular to the plane containing the foil normal and the propagation vector when $\theta \sim \pi/2$. Most treatments of bremsstrahlung in the literature assume that the nuclei upon which the incident particle scatters are isolated. It is of some interest to consider these nuclei as embedded in a foil of a given dielectric constant and to derive the distribution of bremsstrahlung photons in angle and frequency for specific foil thicknesses. It is clear that photons of this sort in the range of optical frequencies may be strongly modified both in direction and number by the dielectric response of the foil.

III. COMPARISON WITH THE FERRELL THEORY

To compare the present results with those of Ferrell, which were obtained on the basis of general physical arguments rather than by a detailed consideration of the electrodynamic equations, one must recall that Ferrell considers a thin foil containing a free-electron plasma in the nonrelativistic case and assumes that the emitted radiation pattern is peaked strongly about the plasma resonance frequency ω_p. In this case one may make an expansion of Eq. (14), assuming that the quantity t is small but that t/β is unrestricted. In addition, one must take the dielectric constant ϵ to be that appropriate to a classical free-electron gas and assume that ω is close to the plasma frequency ω_p.

If one combines Ferrell's equations (3), (6a), (28), (30), (33), (37), and (38),[20] one finds after some simplification and using the notation of the present paper,

$$[n(\mu,a,\omega)]_{\text{Ferrell}}=\frac{\alpha\beta^2}{4\pi^2}\frac{\theta^2\omega_p\sin^2(t/2\beta)}{[\frac{1}{4}\omega_p^2(\epsilon^2+\frac{1}{2}t\theta^2)^2+(\omega-\omega_p)^2]},$$

where $\sin\theta$ has been set equal to θ and $\cos\theta \simeq 1$ in Ferrell's formulas. The present results may be compared with Ferrell's by writing the nonrelativistic limit of Eq. (14) for the case $Z=1$:

$$n^{\mathrm{NR}}(\mu,\omega,a)=\frac{\alpha\beta^2}{\pi^2\omega}\mu^2(1-\mu^2)|\epsilon-1|^2 \times\left|\frac{(\sigma+\mu\epsilon)e^{-it\sigma}+(\sigma-\mu\epsilon)e^{it\sigma}-2\sigma e^{-it/\beta}}{(\mu\epsilon-\sigma)^2e^{it\sigma}-(\mu\epsilon+\sigma)^2e^{-it\sigma}}\right|^2.$$

Assuming that t and θ are both $\ll 1$ but leaving t/β unspecified, one has

$$n^{\mathrm{NR}}(\mu,\omega,a)=\frac{\alpha\beta^2}{\pi^2\omega}\mu^2(1-\mu^2)|\epsilon-1|^2\left|\frac{1-e^{-it/\beta}}{-2\mu\epsilon+it(\mu^2\epsilon^2+\sigma^2)}\right|^2.$$

Since Ferrell considered only the case of photon emission from plasma, one sets $\epsilon=\epsilon_1+i\epsilon_2$, $\epsilon_1=1-(\omega_p/\omega)^2 \simeq(2/\omega_p)(\omega-\omega_p)$ in the neighborhood of the plasma resonance. Then $\sigma^2\simeq\epsilon_1+i\epsilon_2-\theta^2$ and one finds, setting $\epsilon_1=0$, everywhere except in the term giving rise to the characteristic resonance denominator and assuming $\epsilon_2\ll 1$,

$$n^{\mathrm{NR}}(\mu,\omega,a) =\frac{\alpha\beta^2}{4\pi^2}\frac{\theta^2\omega_p\sin^2(t/2\beta)}{[(\omega-\omega_p)+\frac{1}{4}t\omega_p\epsilon_2]^2+\frac{1}{4}\omega_p^2[\epsilon_2+\frac{1}{2}t(\theta^2+\epsilon_2^2)]},$$

which differs from Ferrell only in terms containing ϵ_2. This result confirms the essential correctness of the ingenious physical arguments which Ferrell has employed in deriving his distribution function.

If one attempts to employ this combination of Ferrell's formulas in the region of angles $\theta>1$, a minor discrepancy appears, i.e.,

$$[n(\mu,a,\omega)]_{\text{Ferrell}} =\frac{\alpha\beta^2}{4\pi^2}\frac{\omega_p\sin^2\theta\cos^3\theta\sin^2(t/2\beta)}{\cos^2\theta(\omega-\omega_p)^2+\frac{1}{4}\omega_p^2(\epsilon_2\cos\theta+\frac{1}{2}t\sin^2\theta)^2},$$

while the present approach yields the result

$$n^{\mathrm{NR}}(\mu,a,\omega)\simeq\frac{\alpha\beta^2}{4\pi^2}\frac{\omega_p\sin^2\theta\cos^2\theta\sin^2(t/2\beta)}{[\cos\theta(\omega-\omega_p)+\frac{1}{4}t\epsilon_2]^2+\frac{1}{4}\omega_p^2[\epsilon_2\cos\theta+\frac{1}{2}t(\sin^2\theta+\epsilon_2^2)]^2},$$

[19] See for example, R. L. Gluckstern, M. H. Hull, Jr., and G. Breit, Phys. Rev. **90**, 1026 (1953); R. L. Gluckstern and M. H. Hull, Jr., *ibid.* **90**, 1030 (1953).

[20] Ferrell's equation (38), which gives a frequency distribution multiplying an angular distribution $I(\theta)$, must be multiplied by a factor $\pi/2t_d$ in order to maintain a proper normalization.

indicating that this way of writing his result is most accurate in the region of small angles, as one might expect from a consideration of the approximations made.

IV. THE THICK FOIL

In order to bring out the physical meaning of the rather cumbersome general expression for n (Eq. 14) above, consider the limiting case $a \to \infty$. One finds in this limit,

$$n_\infty(\mu,\omega,\beta)=\frac{Z^2\alpha\beta^2}{\pi^2\omega}\frac{\mu^2(1-\mu^2)}{(1-\beta^2\mu^2)^2} \times\left|\frac{(\epsilon-1)(1-\beta^3\sigma-\beta^2[\epsilon-\mu^2])}{(\sigma+\mu\epsilon)(1-\beta^2\sigma^2)}\right|^2.$$

Specializing further to the nonrelativistic case, setting

$$I_\infty{}^{\mathrm{NR}}(\mu,\omega)\equiv\frac{\pi^2\omega}{Z^2\alpha\beta^2}n^{\mathrm{NR}}(\theta,\omega), \tag{16}$$

one may write

$$I_\infty{}^{\mathrm{NR}}(\mu,\omega)=\frac{\mu^2(1-\mu^2)|\epsilon-1|^2}{|\mu\epsilon+(\epsilon-1+\mu^2)^{\frac{1}{2}}|^2}. \tag{17}$$

Note that I is proportional to the distribution of photon intensity. If one sets $\epsilon(\omega)=\epsilon_1(\omega)+i\epsilon_2(\omega)$, then

$$I_\infty{}^{\mathrm{NR}}(\mu,\omega)=\frac{\mu^2(1-\mu^2)[(\epsilon_1-1)^2+\epsilon_2{}^2]}{(\mu\epsilon_1+R^{\frac{1}{2}}\cos\Phi)^2+(\mu\epsilon_2+R^{\frac{1}{2}}\sin\Phi)^2}, \tag{18}$$

where

$$R=[(\epsilon_1-1+\mu^2)^2+\epsilon_2{}^2]^{\frac{1}{2}},$$

and

$$\Phi=\tfrac{1}{2}\tan^{-1}\left(\frac{\epsilon_2}{\epsilon_1-1+\mu^2}\right).$$

Take $\epsilon_1=1-\omega_p{}^2/\omega^2$ corresponding to the case of the classical free-electron gas. If $|\epsilon_2|\ll|\epsilon_1|$ and if μx is not too close to unity,

$$I_\infty{}^{\mathrm{NR}}(\mu,\omega)=\frac{\mu^2(1-\mu^2)}{\mu^2(1-x^2)^2+x^2(1-\mu^2x^2)}, \quad (\mu x<1), \tag{19a}$$

$$=\frac{\mu^2(1-\mu^2)}{\{\mu(x^2-1)+x(\mu^2x^2-1)^{\frac{1}{2}}\}^2}, \quad (\mu x>1), \tag{19b}$$

where $x=(\omega/\omega_p)$. Consider now some limiting cases: If $x\ll1$, one has $I_\infty{}^{\mathrm{NR}}(\mu,0)=\sin^2\theta$.

This is characteristic of photon emission from a dipole oriented in the direction of the z axis. In classical terms, it represents low-frequency radiation which is emitted when the field of the incident-charged particle and its image charge is completely annihilated upon entry of the particle into the foil.[7] In case the particle emerges from the foil, its effect is not felt in vacuum until it is actually outside the foil, whereupon radiation is emitted due to the sudden "creation" of the particle at the surface together with its image charge in the metal. For frequencies small compared with ω_p, the free-electron gas may be considered to have infinite conductivity and this picture is valid. For frequencies comparable with ω_p, the metal plasma does not shield the electromagnetic field of the particle completely while it is inside the foil, and the radiation pattern is significantly different from the dipole form. In general, the whole system of incident charge plus induced polarization currents in the plasma, coupled through the electromagnetic field, is capable of radiating into vacuum. When $\omega=\omega_p/\sqrt{2}$, one has

$$I_\infty{}^{\mathrm{NR}}(\mu,\omega_p/\sqrt{2})=2\mu^2(1-\mu^2), \tag{20}$$

which is just the angular distribution of radiation emitted by an axial quadrupole with axis oriented in the z direction. When $\omega\sim\omega_p$, the distribution has a more complicated form, depending more critically upon ϵ_2, the imaginary part of the dielectric constant.

If $\epsilon_2\ll1$, one may show that at the photon frequency $\omega=\omega_p$

$$I_\infty{}^{\mathrm{NR}}\sim\mu^2(1-\mu^2)/(\theta^4+\epsilon_2{}^2)^{\frac{1}{2}}, \tag{21}$$

if $\theta\ll1$. Figure 1 shows a plot of the angular distribution of the radiation intensity $I_\infty{}^{\mathrm{NR}}$ emitted from a thick foil having a dielectric constant characteristic of a classical free-electron gas, $\epsilon_1=1-(\omega_p/\omega)^2$, for various photon frequencies. The curve appropriate to $\omega=\omega_p$ was calculated under the assumption that $\epsilon_2=0.01$ while the other curves were calculated assuming $\epsilon_2=0$.

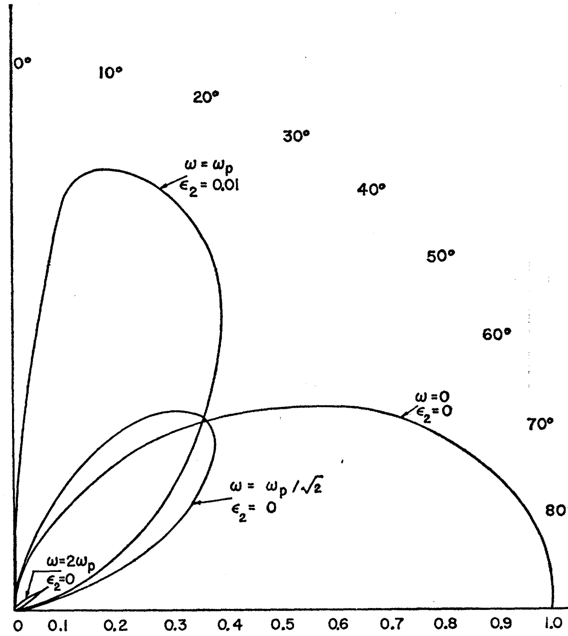

FIG. 1. Angular plot of photon intensity I vs θ at various frequencies for a free-electron dielectric constant.

Figure 2 shows a plot of photon intensity $I_\infty{}^{\mathrm{NR}}$ at various angles with respect to the foil normal for the

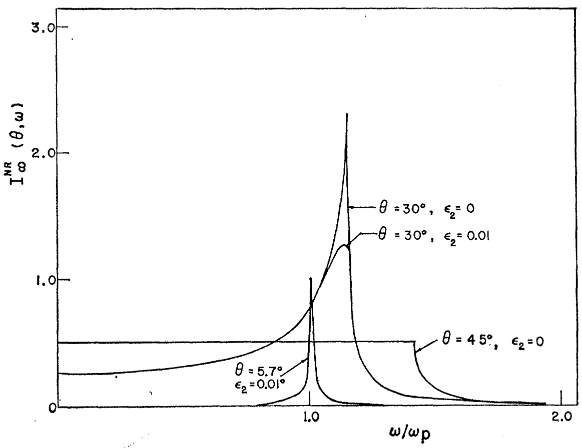

FIG. 2. Predicted variation of photon intensity with frequency at various angles of observation for a thick foil in the nonrelativistic case.

same ϵ_1. The form and magnitude of the distribution is very sensitive to ϵ_2 at small angles and less so for larger angles. The most noteworthy feature of the distribution in frequency is the fact that there is a dependence of the most probable frequency upon the angle of observation. For values of $\theta \ll 1$, the most probable frequency ω_m is only slightly above ω_p, while ω_m increases as θ grows larger. At $\theta = \pi/4$, the maximum disappears, and for $\theta > \pi/4$ the distribution becomes a monotonic decreasing function of ω. This shift may be understood in terms of refraction of light at the surface of the dielectric. From Eq. (18) one sees that the most probable frequency for the intensity distribution occurs near the point at which $\epsilon_1 - 1 + \mu^2 = 0$, if $\epsilon_2 \ll 1$. Setting $\epsilon_1 = 1 - \omega_p^2/\omega^2$, one finds

$$\omega_m = \omega_p/\mu. \tag{22}$$

To understand this effect qualitatively, consider a photon originating in the foil interior and striking the surface at an angle θ' with respect to the foil normal. The foil is transparent to photons of frequency $>\omega_p$ and acts to absorb photons with $\omega < \omega_p$ so that one expects relatively small emission unless $\omega \gtrsim \omega_p$. Since the phase velocity of photons with $\omega > \omega_p$ is $>c$ in the foil, photons in this range undergo refraction at the interface and are bent toward the foil normal. The exit angle θ is related to θ' by Snell's law:

$$\epsilon^{\frac{1}{2}} \sin\theta' = \sin\theta,$$

or $\mu^2 = 1 - \epsilon \sin^2\theta'$. If one sets $\theta' = \pi/2$ and $\epsilon = 1 - \omega_p^2/\omega^2$, one finds $\omega = \omega_p/\mu$, which agrees exactly with Eq. (22), the equation for the most probable frequency. This shows that at a given frequency $\omega > \omega_p$ there is a value of θ beyond which photons originating inside the foil will not be observed. Conversely, if the angle of observation θ is fixed and $\omega_p < \omega < \omega_p/\mu$, only photons which may be rather strongly absorbed are able to emerge from the foil. For $\omega \geqslant \omega_p/\mu$ the foil is transparent, but the response of the electronic system to external perturbation becomes weaker as ω increases from this value. Hence one expects qualitatively that there should be a maximum in the photon intensity distribution at the frequency $\omega = \omega_p/\mu$, with a fairly rapid decrease on either side of this value.

One may examine the photon distribution from a thick foil which is characterized by a more general dielectric function of the form

$$\epsilon(\omega) = 1 + \frac{4\pi e^2}{m}\left[\sum_n \frac{f_{0n}}{\omega_{0n}{}^2 - \omega^2} + i\pi \sum_n \frac{f_{0n}}{\omega}\delta(\omega - \omega_{0n})\right],$$

where, as usual, the f_{0n} are the oscillator strengths per unit volume for interband or intraband transitions between the Bloch one-electron states in the solid, and $\hbar\omega_{0n}$ are the corresponding energies. It is easy to show that the form of the photon-distribution function from such a solid in the neighborhood of the resonance energies is very similar to that from a free-electron gas in the neighborhood of the plasma frequency.

This similarity in the emitted-photon spectra shows that the existence of Coulomb-stimulated photon emission from a given solid will not prove that electronic excitations in that solid are necessarily collective in nature.

V. THE THIN FOIL

Examining further special cases of Eq. (14), one rewrites the general equation in terms of the intensity distribution I as follows:

$$\begin{aligned} I(\mu,\omega) = {} & \frac{\mu^2(1-\mu^2)|\epsilon - 1|^2}{|\Delta|^2(1-\mu^2\beta^2)^2|1-\beta^2\sigma^2|^2} \\ & \times |(1-\beta\sigma-\beta^2)(1+\beta\sigma)(\sigma+\mu\epsilon)e^{-it\sigma} \\ & + (1+\beta\sigma-\beta^2)(1-\beta\sigma)(\sigma-\mu\epsilon)e^{it\sigma} \\ & - 2\sigma(1-\beta\mu-\epsilon\beta^2)(1+\beta\mu)e^{-it/\beta}|^2. \end{aligned} \tag{23}$$

Now if one expands the exponentials in numerator and denominator in power series ($\omega a/v_i \ll 1$ and $|\epsilon|^{\frac{1}{2}}\omega a/c \ll 1$), and retains only those terms of lowest order in t, one finds that the distribution varies quadratically with foil thickness in this range, viz.

$$\begin{aligned} n(\mu,\omega) = {} & \frac{Z^2\alpha\omega\mu^2(1-\mu^2)}{4\pi^2c^2} a^2 \frac{|1-\epsilon\mu\beta-\beta^2|^2}{(1-\beta^2\mu^2)^2} \\ & \times \frac{|\epsilon-1|^2}{|\mu\epsilon - i(\mu^2\epsilon^2+\sigma^2)t/2|^2}, \end{aligned} \tag{24}$$

showing the interesting fact that in the small-thickness case, the number distribution is no longer proportional to β^2, but is more nearly independent of particle velocity, especially in the nonrelativistic limit. If one computes the ratio of n for the thin and thick foil cases at $\epsilon_1 \cong 0$, $\epsilon_2 \neq 0$, $\theta \ll \epsilon_2^{\frac{1}{2}} \ll 1$ for the case of plasma, one finds

$$\frac{[n^{\mathrm{NR}}(\mu,\omega)]_{\mathrm{thin}}}{[n^{\mathrm{NR}}(\mu,\omega)]_{\mathrm{thick}}} = \frac{\omega^2 a^2}{4c^2} \frac{1}{\beta^2[\epsilon_2(\omega_p)]}, \tag{25}$$

showing the strong enhancement which occurs at points close to natural resonances of the system in the thin foil case. Equation (24) shows that photon emission from a thin foil will be strongest at frequencies close to the natural excitation frequencies of the system ($\epsilon \simeq 0$) whether these excitations are collective in nature or are of inter- or intraband type.

Another interesting special case of Eq. (14) may be examined. If one supposes that ϵ_2, the damping constant of excited states, is vanishingly small, and if one confines his attention to photon frequencies such that $\epsilon = 1-\mu^2 (\sigma=0)$ and takes $\beta^2 \ll 1$, Eq. (23) reduces to

$$I^{\mathrm{NR}}|_{\sigma\to 0} = \left\{ \frac{\sin^2(t/2\beta) - t\sin(t/\beta)\mu(1-\mu^2)/2 + t^2\mu^2(1-\mu^2)^2/4}{1+t^2\mu^2(1-\mu^2)^2/4} \right\}, \tag{26}$$

if $(1-\mu^2) \neq 0$. The condition $\sigma=0$ corresponds to a viewing angle $\theta = \cos^{-1}\mu$ and a frequency equal to the most probable frequency of the I distribution for that angle in the case of the infinite foil. If $t\mu^2(1-\mu^2) \ll 1$, $I = [\mu^4/(1-\mu^2)] \sin^2(t/2\beta)$ showing an oscillatory dependence of the photon intensity on the quantity $t/2\beta = a\omega_\sigma/2v_i$, where ω_σ is the frequency at which $\sigma=0$. These oscillations damp out as t grows large and $I \to [4\mu^2/(1-\mu^2)]$ as $t \to \infty$, which agrees with Eq. (19) for the special case $\sigma=0$.

The physical significance of these oscillations in photon intensity with foil thickness may be understood on the basis of the following qualitative considerations. For a transition of the incident electron from an incident plane wave state to one of energy lower by the amount $\Delta E = \hbar\omega$, the transition charge density varies approximately as $e^{+i(\omega z/v_i)}$ if the angular deflection experienced by the electron in this transition is small. This oscillating charge density has equivalent wavelength $2\pi v_i/\omega$ normal to the foil surfaces. One expects coherent addition of field amplitudes generated by such a charge distribution with a maximum radiation of energy when $a = \pi v_i/\omega$. The radiated energy should then decrease with increasing thickness to a minimum at $a = 2\pi v_i/\omega$. This pattern would be expected to repeat with maxima (minima) at odd (even) multiples of $\pi v_i/\omega$. Equation (26) shows that this behavior is predicted by the present work thicknesses which are not too large.

VI. THE OXIDE FILM

The effect on the emitted-photon distribution of a thin impurity layer on the surface of an irradiated foil is considered briefly. Stern and Ferrell[4] have shown that a thin layer (~20 Å) of oxide formed on the surface of Al or Mg foils results in an appreciable change in the characteristic frequency of surface plasmons. It is of some interest to see how the emitted-photon distribution is affected by such an oxide layer.

The algebraic complexity of the general case involving a four or five layered medium is great. The present section will deal only with the simple case of a thin layer deposited on a thick foil in the nonrelativistic limit. One may proceed as before to obtain the far-zone Poynting vector and the emitted-photon distribution function in vacuum from a thick foil with dielectric constant ϵ, covered by a thin layer having thickness a_0, and dielectric constant ϵ_0. One finds for the intensity function I_∞^{NR}

$$I_\infty^{\mathrm{NR}} = \mu^2(1-\mu^2) \times \left| \frac{1-\epsilon-(\epsilon_0-1)(e^{it_0/\beta}-1)\epsilon/\epsilon_0}{\mu\epsilon+\sigma+it_0\{\sigma[\sigma_0\epsilon_0-\mu]+(\epsilon_0-1)(1-\mu^2)\epsilon/\epsilon_0\}} \right|^2,$$

where $t_0 = a_0\omega/c$, and $\sigma_0 = (\epsilon_0 - 1 + \mu^2)^{\frac{1}{2}}$. If one takes $a_0 = 20$ Å, $\epsilon_0 \simeq 2$,[4] and considers the case $\epsilon_1 = 1-(\omega_p/\omega)^2$, $\epsilon_2 = 0$ again, one finds that the term linear in t_0 in the denominator is unimportant compared with $\mu\epsilon+\sigma$ even at the maximum of I, i.e., when $\mu = \omega_p/\omega$. However, the magnitude of this maximum is an oscillatory function of $1/\beta$ in this case. Putting $\hbar\omega_p = 15$ ev, $\mu = 0.866$ and $\omega/\omega_p = 1.155$, one finds

$$I_\infty^{\mathrm{NR}}|_{\max} = 4.25 - 2\cos(0.1487/\beta).$$

In the absence of the oxide film ($\epsilon_0 = 1$ or $t_0 = 0$), one would have for the same conditions $I_\infty^{\mathrm{NR}}|_{\max} = 2.25$. Thus, in this simple case, the position of the maximum in the photon-intensity distribution is insensitive to the presence of the oxide layer, but the magnitude of the maximum may be affected strongly. However, the use of a more realistic dielectric function for the foil, including damping ($\epsilon_2 \neq 0$), in this expression will show less sensitivity to the presence of the oxide layer. Similarly, one expects the effect to be comparatively smaller when the foil itself is thin.

The authors will consider the case of the oxide film in more detail in a later publication. Detailed calculations in the next section have been made neglecting the presence of impurity or oxide films on the foils.

VII. USE OF OPTICAL DATA TO PREDICT $n(\theta,\omega,a,\beta)$ FOR Ag AND Al

Careful measurements of the optical constants of Ag in the region of photon energies from 0.5 to 12 ev have recently been made by Taft and Philipp.[21] The pronounced structure in their curves for n and k vs photon energy in the neighborhood of 3.8 ev has been associated with the existence of interband transitions. Suffczynski[22] has been able to obtain general agreement

[21] E. A. Taft and H. R. Philipp, Phys. Rev. **121**, 1100 (1961).
[22] M. Suffczynski, Phys. Rev. **117**, 663 (1960).

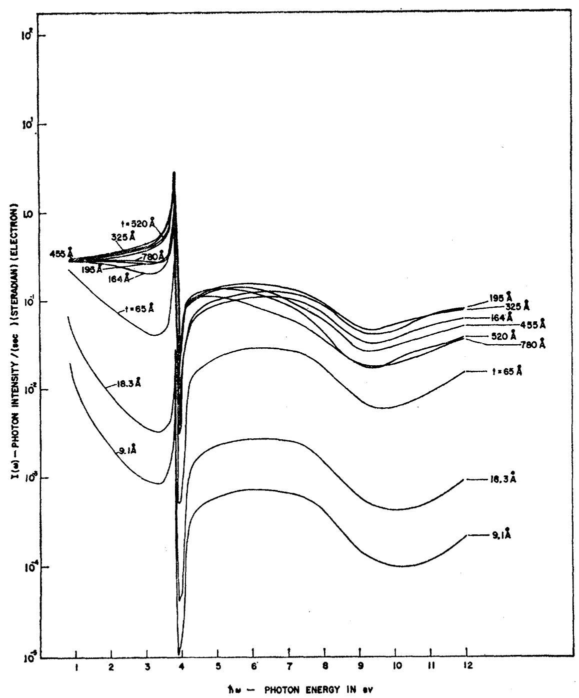

FIG. 3. Variation of photon intensity I from Ag with photon frequency for various foil thicknesses, $\theta=30°$, $\beta=0.316$.

with earlier measurements of n and k in Ag using a theoretical model in which interband transitions from the conduction band to the first unoccupied band are considered. It is clearly not necessary to invoke the mechanism of collective oscillations to explain the pronounced structure of n and k in this range, nor, as noted above, in order to explain a peak in the curve of emitted photon number vs photon energy around 3.8 ev in the light from irradiated foils of Ag.

A digital computer code for the IBM-7090 has been written to evaluate Eq. (14) for a general dielectric constant having a real and an imaginary part. The

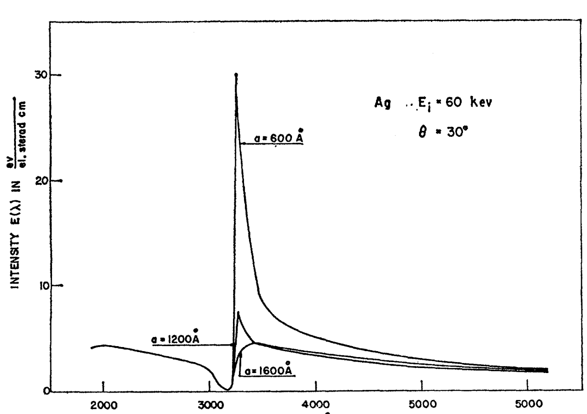

FIG. 4. Photon intensity from Ag vs photon wavelength for various values of a. $E_i=60$ kev, $\theta=30°$.

optical data of Taft and Philipp have been used as input and a number of different spectral distributions have been generated for various values of a, β, and θ for electrons incident on foils of Ag. Figure 3 shows a plot of the intensity $I=\pi^2\omega/\alpha\beta^2 n$ vs photon energy for various foil thicknesses and for $\theta=30°$, $\beta^2=0.1$. One sees the rapid increase in intensity with increasing a when a is small, and for photon energies $\sim$4 ev and larger, with saturation in the neighborhood of 500 Å. This increase is less marked at lower photon energies, as one expects.

Figure 4 shows a plot of

$$\hbar\omega n(\lambda,\theta,a,\beta)d\omega/d\lambda\equiv E(\lambda,\theta,a,\beta),$$

the photon-intensity distribution per unit wavelength λ per incident electron per steradian as a function of λ for three different values of a, and for an incident electron energy $E_i=60$ kev and $\theta=30°$. This illustrates that photon emission at the excitation energy of the

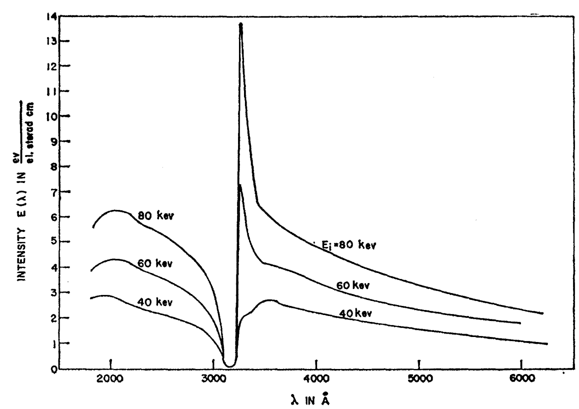

FIG. 5. Photon intensity E vs λ from Ag for various energies of the incident electron. $a=1200$ Å and $\theta=30°$.

electronic system is much sharper for thin foils than for thick ones, as was discussed above for the case of the classical plasma.

Figure 5 shows how the photon intensity from a 1200 Å foil varies with λ for different energies of the incident electron at a viewing angle of $\theta=30°$. The depressed maximum at $\sim$3300 Å with an incident energy of 50 kev compared with the strong peaking at 80 kev at the same wavelength is due in part to destructive interference between currents induced in different portions of the foil by the incident electron (see Sec. IV). Note that the quantity $2\pi v/\omega$ is very close to 1200 Å, the foil thickness, when $E_i=40$ kev, but is about 1600 Å when $E_i=80$ kev.

This same phenomenon is illustrated more clearly in Fig. 6 which shows the variation in photon intensity with foil thickness at various energies: One considers here a silver foil with $\theta=30°$ and $\lambda=3265$ Å.

Figure 7 illustrates the variation of photon intensity from Ag with λ at various viewing angles θ for a 1200 Å foil. One sees how the maximum disappears for $\theta \sim \pi/2$, while the total emission is low when $\theta \ll 1$.

The variation in the number of photons per unit wavelength as a function of θ is shown in Fig. 8 at different wavelengths for the case of a foil of Ag, 800 Å thick, bombarded by 40-kev electrons.

Optical data on[23] Al have been used to predict photon distributions from irradiated foils of this metal. The results are rather unspectacular in the region of visible wavelengths in which the data of Frank *et al.*[15] are available. Figure 9 shows predictions of the number of photons emitted per unit wavelength per electron per steradian as a function of the electron energy E_i, and for two different values of λ and several values of a. In this case θ was taken as 30°. One sees that at the longer wavelength of 3408 Å, the number of photons

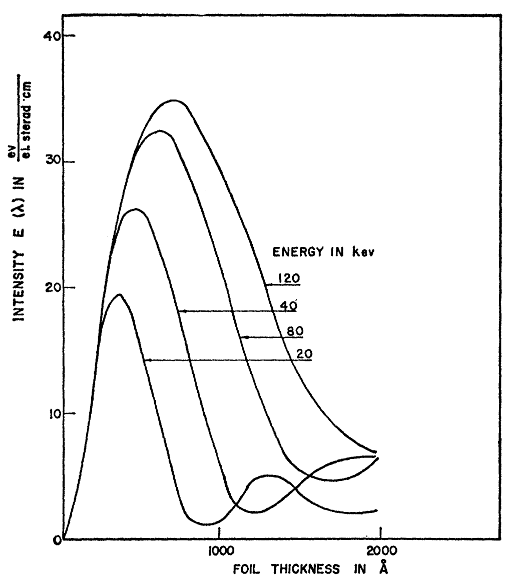

FIG. 6. Photon intensity E vs λ from Ag as a function of foil thickness for various incident energies. $\theta = 30°$, $\lambda = 3265$ Å.

emitted is very insensitive to the foil thickness, indicating that photons are generated in a surface layer of the material of thickness $\lesssim 100$ Å.

Further data computed from Eq. (14) for both Ag and Al together with experimental data are given in the accompanying paper by Frank, Arakawa, and Birkhoff.[15] These comparisons indicate that most of the photons which they observe may be explained on the basis of the present extension of the Frank-Ginsburg theory.

One may note here that although the interpretation of photon-distribution measurements from irradiated foils by the present approach is rather indirect because of the algebraic complication of Eq. (14), it is possible

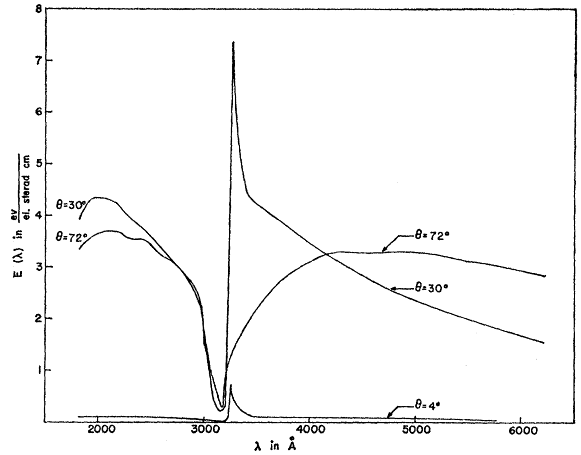

FIG. 7. Photon intensity E vs λ from Ag at various angles θ. $a = 1200$ Å, $E_i = 60$ kev.

to simplify considerably by confining one's attention to limiting cases, e.g., thick or thin foils. It might even be possible to infer $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ from measurements of Coulomb-stimulated photon emission in such simple cases.

VIII. SUMMARY

A generalization of the Frank-Ginsburg approach has resulted in a general equation for the distribution of optical photons generated when a charged particle of velocity v_i bombards a foil characterized by a dielectric constant $\epsilon(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega)$, and having

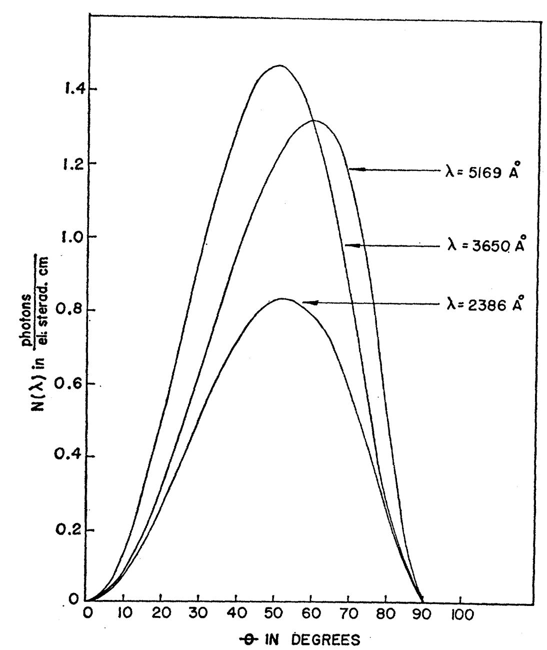

FIG. 8. Number of photons per unit wavelength vs θ from Ag bombarded with 40-kev electrons at various wavelengths. $a = 800$ Å.

[23] P. H. Berning, G. Haas, and R. P. Madden, J. Opt. Soc. Am. **50**, 586 (1960); G. Haas and J. E. Waylonis, J. Opt. Soc. Am. **51**, 719 (1961).

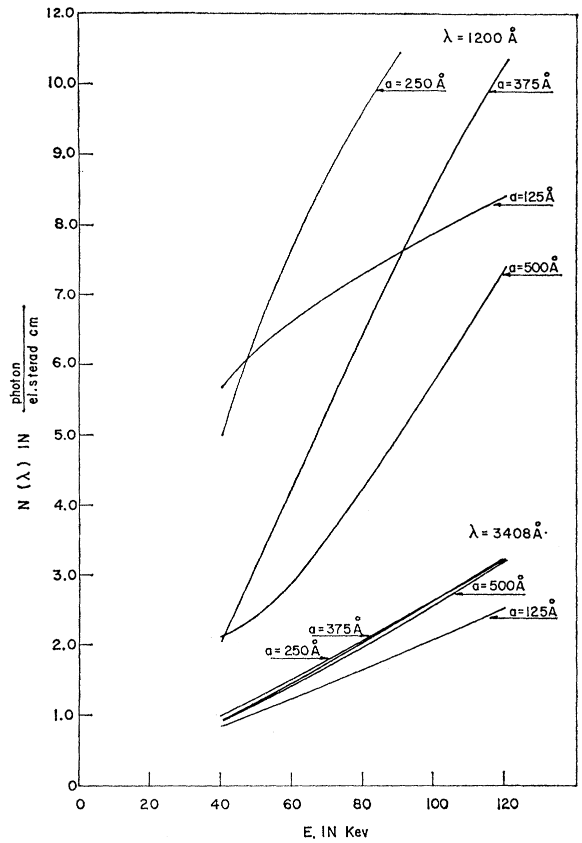

FIG. 9. Photons emitted from Al per unit wavelength vs E_i for various values of a and λ. $\theta=30°$.

thickness a. The treatment is valid when $(2mE_r)^{\frac{1}{2}} \ll v_i \lesssim c$, where m is the particle mass and E_r is the energy corresponding to the most energetic collective or interband transition in the solid which needs to be considered. The expression is shown to reduce to that obtained by Ferrell if one assumes that the charged particle is nonrelativistic and that the $\epsilon(\omega)$ is that of a classical free-electron gas. The general result is examined in the limiting cases of very thick and very thin foils. It is shown that a continuum of frequencies is expected in the general case and that one expects the photon distribution to be strongly peaked at frequencies corresponding to energetic transitions in the electronic system of the solid, irrespective of whether the transitions are collective in nature, corresponding to the creation of plasmons, or whether they are of the interband type. This peaking may be quite pronounced in this thin foil case; and when the damping of excited states is small, the photon yield may be appreciably greater than from a thick foil of the same composition. A connection is found between the most probable frequency in the intensity distribution and the angle of observation in the case of thick foils, and is to be understood in terms of the refraction of photons in crossing the foil-vacuum interface.

Numerical results have been obtained for foils of Ag using the optical data on $\epsilon(\omega)$ taken from the work of Taft and Philipp and for foils of Al using the data of Haas *et al.* These results are compared with experimental data of Frank, Arakawa, and Birkhoff in the accompanying paper.[15]

ACKNOWLEDGMENTS

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APPENDIX: TRANSITION PROBABILITY OF THE INCIDENT CHARGED PARTICLE

Interaction between the particle and the medium plus electromagnetic field may be divided into (a) interactions resulting in electronic transitions in the volume of the solid and, (b) those leading to surface excitations. Processes of type (b) may be further classified according to whether they correspond to radiative or nonradiative excitations.

Interactions of type (a) have been dealt with extensively in the literature. Charged-particle energy-angle distributions corresponding to volume-plasmon excitation were first given in the semiclassical approximation by J. Hubbard [Proc. Phys. Soc. (London) **A68**, 976 (1955)]. Losses of type (b) have been treated mainly in the nonrelativistic approximation.[3,6] To give a relativistic treatment of surface losses requires only a simple extension of the dielectric treatment given above.

One finds that the probability of a loss-act involving energy change $\hbar\omega$, and momentum change $\hbar K$ in a direction perpendicular to $\mathbf{v}_i$ is given by $4Z^2e^2\,\mathrm{Im}w(\omega,K)/\pi\hbar$. The quantity $w(\omega,K)$ is a long algebraic expression which will be quoted only for the thick foil case ($a\rightarrow\infty$). It is given by

$$w(\omega,K)=K^3[B^2/v_i^2\epsilon-\nu\nu' A^2/\omega^2]/(\nu\epsilon+\nu'), \quad \text{(A1)}$$

where

$$A=(\nu^2+\omega^2/v_i^2)^{-1}-(\nu'^2+\omega^2/v_i^2)^{-1},$$
$$B=\epsilon(\nu^2+\omega^2/v_i^2)^{-1}-(\nu'^2+\omega^2/v_i^2)^{-1},$$

and the notation of Sec. II is used. Neglecting contributions to $\mathrm{Im}w$ from the values of ω at which $\epsilon(\omega)$ has its minima, which corresponds merely to boundary corrections to volume excitations, there are two regions in which $w(K,\omega)$ may become large and imaginary. These regions correspond to radiative and nonradiative surface excitations.

Nonradiative excitations correspond to points in $\omega-K$ space where the denominator $(\nu\epsilon+\nu')$ becomes small and imaginary. Setting $\mathrm{Re}(\nu\epsilon+\nu')=0$ and putting $\epsilon(\omega)=1-(\omega_p/\omega)^2+i\epsilon_2(\omega)$ and assuming $\epsilon_2(\omega_p)\ll 1$, one finds the following condition for surface-plasmon creation:

$$\omega^2=K^2c^2+\omega_p^2/2-[K^4c^4+\omega_p^4/4]^{\frac{1}{2}}.$$

This dispersion relation has been given by Stern.[6] For nonrelativistic incident particle energies, the average decrease of the surface plasmon frequency below the value $\omega_p/2$ is small. Stern and Ferrell[6] show that the fractional decrease is $\sim(v_i/2c)^2$ in this region. At relativistic energies, however, the effect should be noticeable.

For the case $a\neq\infty$, the dispersion relation for surface-plasmon creation is given by the solution of

$$(\nu\epsilon+\nu')=(\nu\epsilon-\nu')e^{-\nu' a}.$$

The nonrelativistic form of this equation has been considered previously.[3,6]

Radiative surface excitation occur when either ν or ν' in Eq. (A1) are imaginary. This happens whenever $\omega>Kc$ for plasma, and corresponds to charged particle transitions accompanied by photon emission into the far zone. One may derive the expression for the photon energy-angle distribution given in Sec. II by considering the general expression for losses to the finite foil.

Energy loss by photon emission is quite small compared with loss in the creation of surface plasmons at nonrelativistic energies. However, in the relativistic range, they are of comparable magnitude, and it is conceivable that a characteristic loss experiment could be performed which would show the existence of both of these excitation modes.

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Optical Emission from Irradiated Foils. II

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The spectra from silver foils irradiated by electron beams from an accelerator consist of weak maxima at 3500 Å and broad continua at longer wavelengths. The intensity of the maxima do not exhibit as strong a dependence on foil thickness as predicted by Ferrell and reported by Steinmann. The intensities of both maxima and the continua were found to be directly proportional to beam energy over the range from 40 kev to 115 kev in agreement with the experiments of Goldsmith and Jelley, and of Boersch *et al.*, and the theory of Ginsburg and Frank for transition radiation. The intensity of similar continua found for Al, Au, and Mg also increased linearly with beam energy. Light from Ag and Al foils was found to be polarized in the plane containing the foil normal and the photon direction as predicted theoretically. The intensity of the light from silver was found to be small near the foil normal and at angles approaching 90°, and to achieve a maximum at an intermediate angle in agreement with the theories of Ferrell, and of Ritchie and Eldridge. The absolute light yield from foils of Al and Ag revealed substantial agreement with the predictions of Ritchie and Eldridge from the transition theory of Frank and Ginsburg.

I. INTRODUCTION

THE possibility that studies of the emission of light from thin metal films may prove or disprove the existence of a conduction electron plasma in certain metals has recently led several groups of experimenters to examine the emission spectra of foils irradiated by electron beams. While the electron plasma is thought to play its most significant role in the absorption of energy from high-energy electrons in foils of Al, Mg, Be, and the alkali metals, the rapid oxidation of these metals even as films evaporated and maintained in vacua of the order of 10^{-6} mm Hg has increased the experimental difficulties already severe because the most interesting spectral region lies in the vacuum ultraviolet. Thus, at the suggestion of R. A. Ferrell, considerable work has been initiated and reported recently on the emission of light from Ag where the spectrum has been known for some time to achieve a maximum at about 3400 Å, and oxidation problems are much reduced. Using 25-kev electrons from an accelerator and a quartz spectrograph which looked at the foil on the side of beam incidence, Steinmann[1] found a maximum in the emission spectrum at 3300 Å for a foil 450 Å thick. A thicker foil (850 Å) showed no such maximum; but after a further increase to 1500 Å thickness, the maximum was again apparent. Such a strong dependence on foil thickness was one of the predictions of the Ferrell[2] theory which is based on a simple model of plasma decay involving emission of a single spectral component at a wavelength corresponding to the plasma frequency. Accordingly, Steinmann interpreted his observations as being strong evidence for the existence of an electron plasma in Ag having an energy of 3.75 ev. Additional correlation with the Ferrell theory was obtained by Brown, Wessel, and Trounson[3] who showed that the intensity at the peak was low near the normal and tangent to a 500-Å foil but became quite

* Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

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[2] R. A. Ferrell, Phys. Rev. **111**, 1214 (1958).
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