

Range of Excited Electrons in Metals*

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The energy of interaction of a single excited electron with the sea of conduction electrons in a metal has been calculated by a self-energy approach. The imaginary part of the self-energy of the excited electron can be interpreted in terms of a total rate of real collisions with the sea of conduction electrons. By weighting the differential scattering rate by the amount of energy lost in each scattering event, one obtains an expression for the instantaneous rate of energy loss of the excited electron as a function of its initial energy. The extremely strong dependence of this rate on the initial energy is the main result of this paper. For slow electrons, by which we mean those of initial energy smaller than the sum of the Fermi energy and the plasma energy of the electron gas, the rate of energy loss is determined by the small imaginary part of the dielectric constant. For electrons close to the Fermi surface this rate is proportional to $(p-p_0)^3$, where p is the momentum of the excited electron and p_0 the Fermi momentum; therefore in this range the rate of energy loss is very sensitive to the initial energy. For fast electrons, a new contribution to the rate of energy loss arises due to a pole of the inverse dielectric constant. This new process corresponds to the excitation of plasma oscillations by the excited electron, and causes fast electrons to lose energy very rapidly.

I. INTRODUCTION

THE self-energy or quasi-particle approach to interactions in a degenerate electron gas has proven quite useful in the study of the electronic properties of metals.^{1,2} The simplicity of the quasi-particle approach arises from the fact that for many electronic properties one need only know the renormalized quasi-particle energies. It is well known^{1,3} that the renormalized quasi-particle energy is a complex function of momentum. The imaginary part of the quasi-particle energy results from the fact that quasi-particle states are not eigenstates of the system. The excited quasi-particle can transfer energy and momentum to the electron gas. The transition rate for these real collisions is responsible for the imaginary part of the electron self-energy, while the real part is due to virtual collisions. In the present paper we investigate the imaginary part of the self-energy in an attempt to understand the rate of energy loss of excited electrons.⁴

When discussing electronic properties of metals, one normally points out the fact that quasi-particles close to the Fermi surface have very long lifetimes. This is essential to such considerations since it becomes meaningless to describe a system in terms of excitations whose lifetimes are short compared to characteristic times of interest in the system (e.g., cyclotron period). In this work we need only the restriction that the energy uncertainty due to the finite lifetime is small compared to the real part of the energy of the excited particle. Thus, unless the imaginary part is small compared to the real part of the quasi-particle energy, the concept of a quasi-particle becomes meaningless.

II. LIFETIME

Since the wave function for a single excited electron has a time dependence⁵ $e^{-iE(p)t}$, where $E(p) \equiv E_R(p) + iE_I(p)$ is the renormalized quasi-particle energy, the probability of an electron occupying a given excited state decays in time with the factor $e^{-2|E_I|t}$. The factor $2|E_I|$ can be interpreted as a transition rate, or its inverse as a lifetime. In the first reference the following formula for the imaginary part of the self-energy is derived:

$$E_I(p) = \text{Im} \frac{e^2}{2\pi^2} \int \frac{d^3k}{k^2 \epsilon(k, E(p) - E(p-k) + i\delta)}, \quad (1)$$

$$E_0 < E(p-k) < E(p),$$

where Im denotes imaginary part of, $\epsilon(k, \omega)$ is the Lindhard⁶ dielectric constant, δ is a positive infinitesimal, and E_0 is the Fermi energy. The Lindhard dielectric constant is given by the expression

$$\epsilon(k, \omega) = 1 + \frac{3\omega_p^2}{k^2 v_0^2} \left\{ \frac{1}{2} + \frac{1}{8z} [1 - (z - \mu)^2] \ln \left(\frac{z - \mu + 1}{z - \mu - 1} \right) + \frac{1}{8z} [1 - (z + \mu)^2] \ln \left(\frac{z + \mu + 1}{z + \mu - 1} \right) \right\}, \quad (2)$$

where ω_p is the plasma frequency and v_0 the Fermi velocity of the electron gas; z is the ratio of wave number k to twice the Fermi wave number $2k_0$. The parameter μ is equal to ω/kv_0 . The dielectric constant has both a real and an imaginary part; we shall use the notation $\epsilon(k, \omega) = \epsilon_1(k, \omega) + i\epsilon_2(k, \omega)$. The imaginary part of $\epsilon(k, \omega)$ is simply a measure of the number of states

¹ J. J. Quinn and R. A. Ferrell, Phys. Rev. **112**, 812 (1958).

² J. J. Quinn and R. A. Ferrell, J. Nuclear Energy, Pt. C, Plasma Physics **2**, 18 (1961).

³ N. M. Hugenholtz and L. Van Hove, Physica **24**, 363 (1958).

⁴ For a preliminary report of this work see J. J. Quinn, Bull. Am. Phys. Soc. **6**, 10 (1961).

⁵ Throughout this paper we take \hbar , the reduced Planck constant, equal to unity.

⁶ J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **28**, No. 8 (1954).

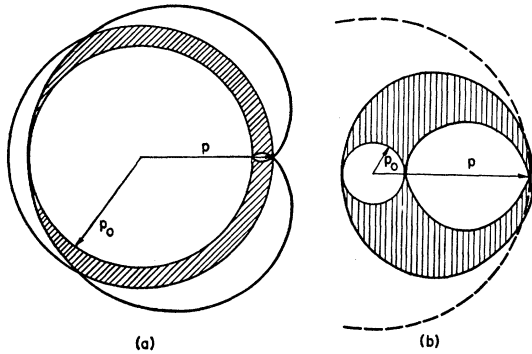


FIG. 1. The region of momentum space contributing to the integral appearing in Eq. (6). The region of integration lies between the concentric spheres of radii p_0 and p . The imaginary part of $\epsilon(k, E(p) - E(\mathbf{p} - \mathbf{k}))$ is nonzero only in the region lying between the two branches of the Pascal limaçon, the curve given by $k = p \cos \theta \pm p_0$ centered about the point \mathbf{p} in momentum space. θ is the angle between $-\mathbf{p}$ and \mathbf{k} . The region giving a finite contribution to the integral is indicated by the striped area. Figure 1(a) is for the case where p is approximately equal to p_0 ; Fig. 1(b) corresponds to p considerably larger than p_0 .

available for real transitions involving a given energy ω and momentum transfer k . The imaginary part of ϵ expressed in the form

$$\begin{aligned} \epsilon_2(k, \omega) &= (3\omega_p^2/k^2 v_0^2) (\pi/2) \mu \text{ for } z + \mu < 1 \\ &= (3\omega_p^2/k^2 v_0^2) (\pi/8z) [1 - (z - \mu)^2] \\ &\quad \text{for } |z - \mu| < 1 < |z + \mu| \\ &= 0, \text{ otherwise.} \end{aligned} \quad (3)$$

To obtain an approximate idea of the behavior of $\epsilon_1(k, \omega)$ one can expand the logarithmic terms in powers of the appropriate small parameters. For example, for $z \ll 1$, $\mu \ll 1$ one can obtain

$$\epsilon_1(k, \omega) \approx 1 + (k_s^2/k^2)(1 - \mu^2), \quad (4)$$

where k_s^2 , the square of the reciprocal Fermi-Thomas screening length, equals $4k_0/\pi a_0$. a_0 is the radius of the first Bohr orbit in hydrogen. For $\mu \gg 1$ one can easily show that

$$\epsilon_1(k, \omega) \approx 1 - \omega_p^2/\omega^2. \quad (5)$$

The dielectric constant has a zero at the plasma frequency corresponding to the natural oscillations of the system. From the behavior of the dielectric constant and the form of Eq. (1), it can be seen that two separate contributions to the imaginary part of the self-energy exist. One contribution results from the pole of $1/\epsilon$ at the plasma frequency and corresponds to the creation of a plasma oscillation by the excited electron. The second contribution arises from integration over that portion of k space for which $\epsilon_2(k, \Delta E)$ is finite. We want to obtain the value of $E_I(p)$ as a function of p for values of p in the range $p_0 \leq p \leq$ several times p_0 . The lifetime of the excited quasi-particle state is then $\tau = [2E_I(p)]^{-1}$. We also wish to demonstrate that the

uncertainty in the energy of the quasi-particle due to its finite lifetime is small compared to its excitation energy. Because the dielectric constant appearing in Eq. (1) is quite a complicated function of \mathbf{k} , it is not possible to obtain a simple closed expression for $E_I(p)$. By making some simplifying assumptions, one can obtain reasonably accurate approximations for different contribution to $E_I(p)$ in certain limiting cases. By resorting to numerical integration in the range where it is impossible to obtain an accurate analytical expression, one can obtain a fairly good picture of $E_I(p)$ vs p .

(a) Low-Energy Excitations

For an excited electron whose initial energy is very close to the Fermi energy $E(p) \approx E_0$, the frequency $[E(p) - E(\mathbf{p} - \mathbf{k})]$ entering the dielectric constant which appears in Eq. (1) is always small. For such frequencies one is always safely removed from the pole of $\epsilon^{-1}(k, \omega)$, and Eq. (1) can be written

$$E_I(p) = -\frac{e^2}{2\pi^2} \int \frac{d^3k}{k^2} \frac{\epsilon_2(k, \Delta E(\mathbf{k}))}{|\epsilon(k, \Delta E(\mathbf{k}))|^2}, \quad (6)$$

$$E_0 < E(\mathbf{p} - \mathbf{k}) < E(p),$$

where $\Delta E(\mathbf{k}) = E(p) - E(\mathbf{p} - \mathbf{k})$. Since we are considering the case where ΔE is always very small $[E(p) \approx E_0]$, $|\epsilon(k, \Delta E)|^2$ in the denominator can be replaced by $|\epsilon(k, 0)|^2$ to a reasonably good approximation. The region of momentum space over which $\epsilon_2(k, \Delta E(\mathbf{k}))$ is finite lies between the surfaces $k = p \cos \theta \pm p_0$ centered about the point \mathbf{p} in momentum space. Here we restrict θ , the angle between $-\mathbf{p}$ and \mathbf{k} , to the range zero to $\pi/2$. The entire surface $k = p \cos \theta \pm p_0$ is a surface of revolution of a two-dimensional curve known as the Pascal limaçon; we shall refer to the plus and minus sections of this surface as the two branches of the surface. The entire contribution to the integral appearing in Eq. (6) comes from that region of momentum space between the two branches of the Pascal limaçon which also lies between two concentric spheres of radii p_0 and p centered about the origin. Examples of the region contributing to the integral are shown in Fig. 1 for two different values of p/p_0 . By substituting from Eqs. (3) and (4) into Eq. (6) one obtains for $p \approx p_0$

$$\begin{aligned} E_I(p) \approx & -\frac{e^2}{2a_0} \frac{\pi^{\frac{1}{2}}}{32(\alpha r_s)^{\frac{1}{2}}} \\ & \times \left[\tan^{-1} \left(\frac{\pi}{\alpha r_s} \right)^{\frac{1}{2}} + \frac{(\alpha r_s/\pi)^{\frac{1}{2}}}{1 + \alpha r_s/\pi} \right] \frac{(x^2 - 1)^2}{x}, \end{aligned} \quad (7)$$

where $\alpha \equiv (4/9\pi)^{\frac{1}{2}}$, r_s is the radius (measured in units of the Bohr radius a_0) of a sphere equal in volume to the volume per electron, and x is p/p_0 .

(b) High-Energy Excitations—Plasmon Creation

For high frequencies $\omega > \omega_p$ the dielectric constant is real and can be approximated by Eq. (5). The ω_p appearing in Eq. (5) actually depends on k , but this dispersion is fairly small and, for the present, we shall neglect it. With this approximation, the integral appearing in Eq. (1) has a simple pole at $E(p) - E(\mathbf{p}-\mathbf{k}) = \omega_p$. The $i\delta$ appearing in the dielectric constant gives the prescription for integration around the pole. We perform the integration as follows: first integrate over the component of \mathbf{k} parallel to \mathbf{p} . The only contribution to the imaginary part is πi times the residue at the pole. Then integrate over angle, restricting k to the interior of the Pascal limaçon, where the imaginary part of $\epsilon(k, \omega)$ is zero. The result for $E(p) > E_0 + \omega_p$ is:

$$E_I(p) = -\frac{\omega_p}{2a_0 p} \ln \left(\frac{(p_0^2 + 2m\omega_p)^{1/2} - p_0}{p - (p^2 - 2m\omega_p)^{1/2}} \right), \quad (8)$$

The upper factor in the logarithm is simply k_c , the cut-off momentum for plasma oscillations:

$$k_c/k_0 = [(1 + \omega_p/E_0)^{1/2} - 1]. \quad (9)$$

In the limit of high densities ($r_s \rightarrow 0$), this reduces to $k_c \approx 0.47 r_s^{1/2} k_0$, by now a well-known expression.^{7,8}

Actually near the threshold for creation of plasma oscillations we should correct Eq. (8) for the dispersion of the plasma oscillations as a function of k . Near the threshold, $E(p) \approx E_0 + \omega_p$, the momentum change of the excited electron is always close to k_c . Since the plasmon created must take up this momentum as well

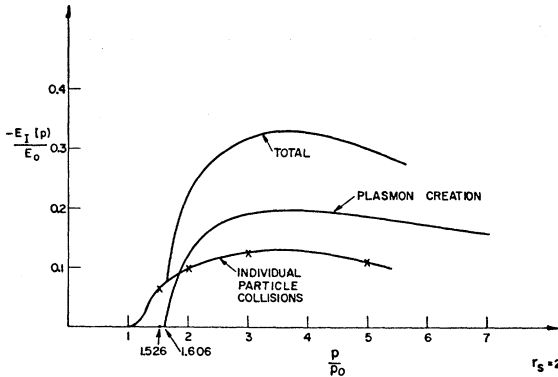


FIG. 2. Graph of $-E_I(p)$, measured in units of the Fermi energy, as a function of p , measured in units of the Fermi momentum for the case $r_s=2$. The curve labeled "individual particle collisions" is the contribution to $E_I(p)$ obtained from Eq. (6). Near $p/p_0=1$, $E_I(p)$ is proportional to $(p/p_0-1)^2$. For p considerably larger than p_0 , the "individual particle collision" contribution to $E_I(p)$ was obtained by numerical integration. The points obtained numerically are indicated by crosses. The curve labeled "plasmon creation" is the contribution from Eq. (8). The correction given by Eq. (11) shifts the threshold for plasmon creation from 1.526 to 1.606. These points are indicated on the graph. The curve labeled total is the sum of the other two.

⁷ J. J. Quinn and R. A. Ferrell, Bull. Am. Phys. Soc. 1, 44 (1956).

⁸ K. Sawada, K. A. Brueckner, N. Fukuda, and R. Brout, Phys. Rev. 108, 507 (1957).

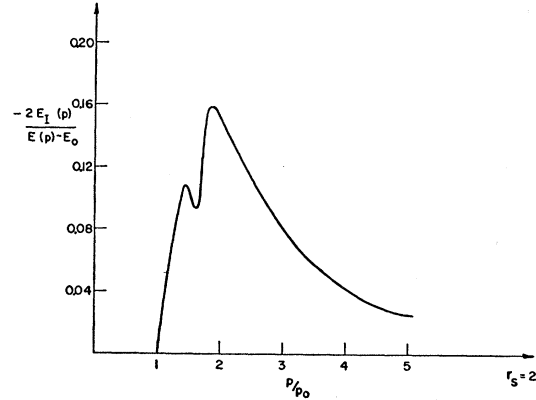


FIG. 3. The ratio of the uncertainty in the quasi-particle energy to its excitation energy as a function of p/p_0 .

as the energy, the energy of the plasmon is somewhat higher than $\omega_p(0)$:

$$\omega_p(k) = \omega_p(0) \left[1 + \frac{3}{10} k^2 v_0^2 / \omega_p^2 \right]. \quad (10)$$

Substituting k_c for k , one obtains for the threshold excitation energy

$$\omega_p(k_c) = \omega_p(0) \left\{ 1 + \frac{6}{5} \frac{E_0^2}{\omega_p^2(0)} \left[\left(1 + \frac{\omega_p}{E_0} \right)^{1/2} - 1 \right]^2 \right\}. \quad (11)$$

This correction factor can amount to about a 15% to 20% shift in the excitation energy required for plasmon creation.

(c) Intermediate and High-Energy Excitations—Individual Particle Collision

It is extremely difficult to obtain any accurate expression for the integral appearing in Eq. (6) when $E(p)$ is more than 20% to 30% bigger than E_0 . For initial energies in this range one must resort to numerical integration. The double integral, over angle θ and magnitude of k , has been evaluated for the case $r_s=2$ (roughly corresponding to aluminum) for several values of p/p_0 . The results are included on the graph labeled "individual particle collisions" of Fig. 2. Figure 2 is a plot of $-E_I(p)/E_0$ as a function of p/p_0 for $r_s=2$. The curve labeled "individual particle collisions" is the contribution to $E_I(p)$ resulting from the finiteness of $\epsilon_2(k, \Delta E(\mathbf{k}))$. The points on this curve marked by a cross were obtained by numerical integration of Eq. (6). The values for $p/p_0 < 1.30$ were obtained by using Eq. (7). The curve labeled plasmon creation is obtained from Eq. (8), making the correction for the threshold described by Eq. (11). This correction shifts the threshold for plasmon creation from $p/p_0=1.526$ to 1.606. The curve labeled "total" is simply the sum of the other two. All the curves are rather roughly sketched through a series of points. This is in accord with the accuracy of the calculation which shall be discussed later. Figure 3 is a plot of the uncertainty in the

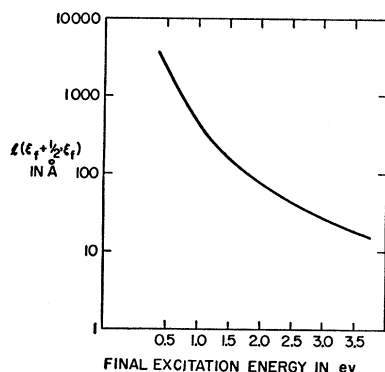


FIG. 4. The mean distance traveled by an excited electron along its own path in losing one half an electron volt of energy, plotted as a function of its final excitation energy. The ordinate is measured in angstrom units, the abscissa in electron volts.

quasi-particle energy divided by its excitation energy as a function of p/p_0 . For the quasi-particle concept to be meaningful the uncertainty in energy must always be small compared to the excitation energy. For this case ($r_s=2$) the uncertainty is never more than 16% of the excitation energy, so that the quasi-particle concept is not too bad.

III. RANGE OF EXCITED ELECTRONS

(a) Low-Energy Excitations

Perhaps a more interesting parameter than the lifetime τ for the low lying excitations is the instantaneous rate of energy loss. Since we have interpreted $2|E_I(p)|$ as the total transition rate for real scattering events, we can obtain the rate of energy loss by weighting the differential scattering rate by the energy lost in each collision. For low-lying excitation [$E(p) \approx E_0$], one obtains

$$\frac{dE(p)}{dt} = -\frac{e^2}{\pi^2} \int \frac{d^3k}{k^2} \times \frac{[E(p) - E(p-k)]\epsilon_2(k, E(p) - E(p-k))}{|\epsilon_1(k, 0)|^2},$$

$$E_0 < E(p-k) < E(p). \quad (12)$$

Dividing both sides of the above equation by the velocity v gives the energy loss per unit length, along the path of the excited electron. Carrying out the integration with the same approximations used in obtaining Eq. (7) gives

$$\frac{dE(p)}{dl} = -\frac{e^2}{2a_0} \frac{2m^2}{3\pi k_s^3 E(p)} \left[\tan^{-1} \frac{2k_0}{k_s} + \frac{2k_0 k_s}{4k_0^2 + k_s^2} \right] \times [E(p) - E_0]^3, \quad (13)$$

where l measures the position of the electron along its path. By straightforward integration one can obtain the following equation for the parameter l , which is a rough measure of the distance along its path that an excited electron travels in going from an initial excita-

tion energy ξ_i [$E_i(p) = E_0 + \xi_i$] to a final excitation energy ξ_f .

$$l(\xi_i, \xi_f) \approx \frac{24(\alpha r_s/\pi)^{1/2} [E_0^2(1/\xi_f^2 - 1/\xi_i^2) + 2E_0(1/\xi_f - 1/\xi_i)] a_0}{[\tan^{-1}(\pi/\alpha r_s)^{1/2} + (\alpha r_s/\pi)^{1/2}/(1 + \alpha r_s/\pi)]} \quad (14)$$

For the case of aluminum ($r_s \approx 2$, $E_0 \approx 12$ ev) the equation reduces to

$$l(\xi_i, \xi_f) \approx 710 \left[\left(\frac{1}{\xi_f^2} - \frac{1}{\xi_i^2} \right) + \frac{1}{6} \left(\frac{1}{\xi_f} - \frac{1}{\xi_i} \right) \right] \text{Å}, \quad (15)$$

where ξ_i and ξ_f are measured in electron volts and l is measured in angstrom units. Figure 4 is a graph of the average distance traveled by an excited electron before it loses one half of an electron volt energy, plotted as a function of its final excitation energy. This distance is obviously extremely sensitive to the final energy. The mean distance traveled in going from 1 to $\frac{1}{2}$ ev, about 2200 Å in aluminum, is not inconsistent with the long mean free paths observed by Spratt *et al.*⁹ It should be emphasized that this mean distance is measured along the path of the excited electron, and we have not considered how the excited electron random walks through the crystal. It should also be pointed out that the mean energy lost by an excited electron which does suffer a collision is not small compared to its initial excitation energy. In fact, for the present case of low-energy excitation one can easily show that an excited

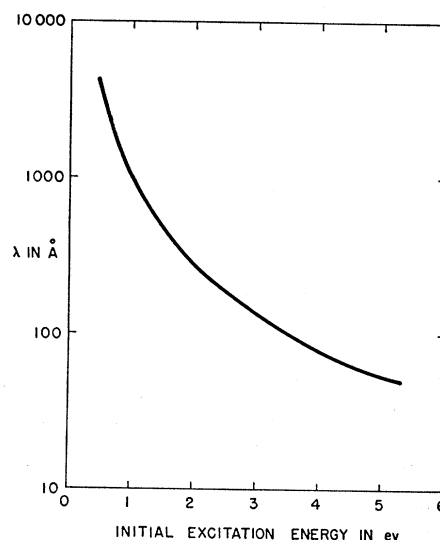


FIG. 5. The "mean free path" for electron-electron collision as a function of the initial excitation energy. The "mean free path" is defined as the product of the velocity and the lifetime of the excited particle, and is measured in angstrom units. The initial excitation energy is measured in electron volts.

⁹ J. Spratt, R. Schwarz, and W. Kane, Phys. Rev. Letters **6**, 341 (1961).

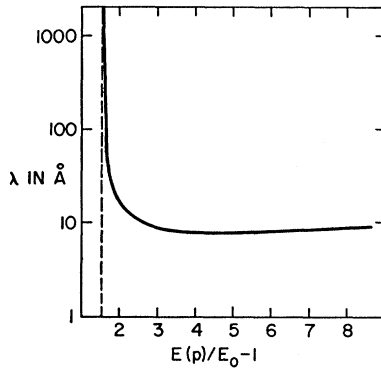


FIG. 6. The mean free path for plasmon emission plotted as a function of the initial energy of the excited electron. The ordinate is measured in angstrom units, the abscissa in units of the Fermi energy.

electron loses $\frac{2}{3}$ of its excitation energy in a typical collision. This implies that the integration of Eq. (13) to obtain Eq. (14) is not really altogether appropriate.

Perhaps a more significant number, as far as the hot electron tunneling experiment is concerned, is the "mean free path" for any electron-electron collision. We define this quantity as the product of the lifetime of the excited particle and its velocity. If no other mechanism (phonons or impurities) are of importance, this mean free path is equal to the distance at which $1/e$ of the excited electrons have not suffered any collision. For an excitation energy of 1 ev in aluminum, this mean free path is approximately 1000 Å. A graph of this "mean free path" as a function of the initial energy is given in Fig. 5.

(b) Mean Free Path for Plasmon Creation

The lifetime for plasmon emission is equal to $[2E_I(p)_{\text{plasmon}}]^{-1}$, where $E_I(p)_{\text{plasmon}}$ is the contribution to $E_I(p)$ resulting from the pole in $\epsilon^{-1}(k, \omega)$. The product of τ and v , the velocity of the excited electron, is equal

to λ , the mean free path for plasmon emission:

$$\lambda = \frac{2a_0 E(p)}{\omega_p} \left[\ln \left(\frac{(p_0^2 + 2m\omega_p)^{\frac{1}{2}} - p_0}{p - (p^2 - 2m\omega_p)^{\frac{1}{2}}} \right) \right]^{-1}. \quad (16)$$

For $E(p) \gg E_0$, as for example in the characteristic energy loss experiments, this expression reduces to the result of Ferrell.¹⁰ Figure 6 contains a graph of the mean free path for plasmon emission as a function of the initial energy for the case of aluminum ($r_s \approx 2$).

IV. SUMMARY

The results of this paper are based upon the dielectric constant approach to the electron-electron interaction. This treatment is strictly valid only for r_s small compared to unity. Since we have applied the results to aluminum ($r_s = 2$), we cannot hope for very good quantitative results, but certainly the qualitative result that the rate of energy loss is strongly dependent on the excitation energy should be true. The calculations of DuBois¹¹ indicate that the dielectric constant approach may be reasonably good even in the range $1 < r_s < 2$. It should be possible to study the dependence of the rate of energy loss on the excitation energy both by tunneling experiments in which different barrier heights are used at the collector, and by photoelectric emission from wedge-shaped specimens whose surfaces have been treated so as to give a range of different work functions.

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¹⁰ R. A. Ferrell, Phys. Rev. **101**, 554 (1956).

¹¹ D. F. DuBois, Ann. Phys. (New York) **7**, 174 (1959).