Electron Correlational Effects on Plasmon Damping and Ultraviolet Absorption in Metals

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A calculation is presented of the dissipative part of the wave-number-dependent, high-frequency dielectric tensor of a free-electron gas. The calculation, which is exact to lowest order in k^2 and r_s, includes dynamical correlational (screening) effects which lie outside the random-phase approximation (RPA). The results, which can be expressed entirely in terms of integrations involving the RPA dielectric functions, differ significantly from those of previous work. The k^2 coefficient of the plasmon linewidth is predicted to be about an order of magnitude smaller than that measured in high-energy electron scattering experiments. The source of the discrepancy may be due to the failure of the r_s expansion for metals or to phonon or impurity scattering and interband transitions, which have been ignored in previous work. These effects are discussed here, but have not been calculated. The contribution of electron-electron correlations to optical absorption is shown to be smaller than previously calculated by essentially a factor of the Fermi velocity squared divided by c^2. Previous calculations ignored terms arising from the perturbation of the screening electrons by the high-frequency fields; these terms greatly reduce the correlational effects.

I. INTRODUCTION

CORRELATION properties of the metallic valence electron gas have been investigated extensively in the last twenty years by electron-scattering experiments[1] and measurements of optical properties.[2] Gross features of these experiments are consistently described by the random-phase approximation (RPA).[3] Several workers have predicted that electron correlational effects which are not included in the RPA can be distinguished experimentally.[4–6] In this paper, we will treat two closely related phenomena: the dispersion of the plasmon line width due to electron-electron collisions and the ultraviolet absorption into a final state of a plasmon plus an electron-hole pair. We conclude that previous calculations[4–7] of these effects make serious omissions and that the correct results (unfortunately) decrease their experimental significance.

The Landau damping process for plasmons in the RPA vanishes for plasmon wave vector k less than a critical value k_c for a degenerate electron gas.[3] For $k<k_c$, the plasmon damping results from electron-electron collisions not included in the RPA which cause fluctuations of the microscopic electric fields about the average self-consistent field of the RPA. It would be of great interest to observe this damping experimentally since it is one of the few effects in a free electron gas which is caused *entirely* by electron correlations. Nozières and Pines[8] on heuristic grounds estimated that the plasmon damping $\gamma(k)$ due to these effects would be proportional to k^2. The first serious microscopic calculation was carried out by DuBois[9] (this reference is referred to as I), who neglected screening effects and whose results were numerically in error by a factor of 4. The correct unscreened calculation of the k^2 coefficient of $\gamma(k)$ gives a result about an order of magnitude greater than that experimentally observed by Powell and Swanson.[4] Ninham[4] repeated this calculation with the hope of adding the effects of dynamic screening. We will show that his method was inconsistent because several important coherent terms were omitted from the transition amplitude. Using a technique previously applied to the high-temperature electron gas,[10] we have included the missing terms and carried out the calculations in a more direct manner requiring fewer simplifying approximations. The results of our latest calculation given in Sec. III show that screening effects reduce the k^2 coefficient to a value about an order of magnitude *below* the experimental value[4,11,12] for Al.

[1] G. Rutheman, Naturwissenschaften **29**, 648 (1941); **30**, 142 (1942); Ann. Physik **2**, 113 (1948); W. Lang, Optik **3**, 233 (1948); Marton, Leder, and Mandlowitz, in *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic Press Inc., New York, 1955), Vol. 7 (survey up to 1955); H. Watanabe, J. Phys. Soc. Japan **11**, 112 (1956); C. Kunz, Z. Physik **167**, 53 (1962); N. Swanson and C. J. Powell, Phys. Rev. **145**, 195 (1965).

[2] H. Ehrenreich and H. R. Phillip, Phys. Rev. **128**, 1622 (1962); H. R. Phillip and H. Ehrenreich, *ibid.* **129**, 1550 (1963); H. Ehrenreich, H. R. Philipp, and B. Segal, *ibid.* **132**, 1918 (1963).

[3] See for example D. Pines, *Elementary Excitations in Solids* (W. A. Benjamin, Inc., New York, 1964).

[4] B. W. Ninham, C. J. Powell, and N. Swanson, Phys. Rev. **145**, 209 (1966).

[5] (a) N. Tzoar and A. Klein, Phys. Rev. **124**, 1297 (1961); (b) D. F. DuBois, V. Gilinsky, and M. G. Kivelson, *ibid.* **129**, 2376 (1963); (c) M. Matsudaira, J. Phys. Soc. Japan **17**, 1563 (1962).

[6] R. J. Esposito, L. Muldawer, and P. E. Bloomfield, Phys. Rev. **168**, 744 (1968).

[7] R. J. Cohn, Ph.D. thesis, University of Maryland, 1966 (unpublished).

[8] P. Nozières and D. Pines, Phys. Rev. **113**, 1254 (1969).

[9] D. F. DuBois, Ann. Phys. (N. Y.) **8**, 24 (1959), hereafter referred to as I.

[10] D. F. DuBois and V. Gilinsky, Phys. Rev. **135**, A1519 (1964), henceforth referred to as DG. (See also DGK in Ref. 5b.)

[11] C. Kunz, Z. Physik **167**, 53 (1962).

[12] C. V. Festenberg, Z. Physik **207**, 47 (1967).

The plasmon damping is proportional to the imaginary part of the longitudinal component of the local dielectric tensor. The optical absorption is proportional to the transverse part of the tensor. Beginning with the work of Tzoar and Klein,[5] several authors[5] have considered the contribution to the optical absorption from a pair-plasmon final state. Most recently, Esposito, Muldawer, and Bloomfield[6] have considered the detailed experimental significance of these results. The conclusion of the present paper is that all of these previous results are seriously incomplete. We find that the previous results for optical absorption by the pair-plasmon process must be reduced by the square of the Fermi velocity divided by c^2 and is almost certainly not experimentally detectable.

The previous papers[4–6] omitted amplitudes for processes corresponding to the perturbation of the screening electrons by the exciting fields. These amplitudes lead to cancellations causing a large reduction in the strength of these processes.

In Sec. II, the calculation of the dissipative part of the dielectric tensor for a degenerate electron gas is summarized. The calculation is based on a "golden rule" formulation of the problem used in several previous papers.[5b,10,13] It should be emphasized that in this calculation we adopt the popular fiction that the weak coupling expansion in powers of r_s has some physical significance even though $r_s>1.8$ for all metals. This is discussed at the end of the present section. The most important feature of the present calculation is the inclusion of the so-called triangle amplitudes which were neglected in previous calculations.[4–6]

In Sec. III, the specific evaluation of these formulas for $T\equiv 0$ is considered. A separation into pair-pair, pair-plasmon, and plasmon-plasmon processes is made in a unique way. The leading contribution to plasmon damping, of order $(k/p_F)^2$, comes from the nonexchange pair-pair process. The results can be written as frequency and wave-number integrations over an integrand which is expressed essentially in terms of components of the RPA dielectric tensor. This integral is evaluated numerically. The contribution of the pair-plasmon final state to optical (ultraviolet) absorption is vastly reduced by a factor $(v_F/c)^2$ compared to the previous calculations mentioned above.[5]

Since for $k<k_c$ a plasmon (or photon) cannot decay into a single electron-hole pair, the damping arises from the decay into two (and higher) pair states. The plasmon can couple to these states only during electron-electron collisions. Because such collisions are screened, they are actually many-body processes. The plasmon can interact with the screening electrons, as well as with "colliding" electrons. This effect is taken into account by the triangle amplitudes.

In Sec. IV, the additional complications which arise from the periodic ion lattice are listed and briefly examined. The most important of these are interband transitions which contribute to plasmon damping even at $k=0$ and which also contribute terms of order k^2. The effects of umklapp processes and nonparabolic energy bands are also mentioned. We conclude that the experimentally observed dispersion of plasmon damping does not give a direct measure of electron correlation effects. It is first necessary to subtract out the lattice contributions and impurity scattering effects which remain to be calculated.

In the RPA, the expression for the wave vector and frequency-dependent dielectric tensor ϵ_{ij} is well known (in units for which $\hbar=1$)[3]:

$$\epsilon_{i\mathrm{k}}{}^0(k,\omega)=\delta_{ij}\left(1-\frac{\omega_p{}^2}{\omega^2}\right)+\frac{4\pi e^2}{\omega^2 m^2}\sum_{\text{spins}}\int\frac{d^3p}{(2\pi)^3}p_ip_j\frac{f_{\mathrm{p}-\frac{1}{2}\mathrm{k}}-f_{\mathrm{p}+\frac{1}{2}\mathrm{k}}}{\omega-E_{\mathrm{p}+\frac{1}{2}\mathrm{k}}+E_{\mathrm{p}-\frac{1}{2}\mathrm{k}}+i\epsilon},\quad(1.1)$$

where $f_p=(e^{\beta(E_p-\mu)}+1)^{-1}$ in terms of the chemical potential μ, $E_p=p^2/2m$, $\beta=T^{-1}$, where T is the absolute temperature in energy units, e and m are the electron charge and mass, respectively, n is the electron density, and $\omega_p=(4\pi ne^2/m)^{1/2}$ is the plasma frequency. (The superscript 0 is used to distinguish the RPA from the complete dielectric function.) In an isotropic system, $\epsilon_{ij}(k,\omega)$ can be decomposed into longitudinal and transverse components:

$$\epsilon_{ij}(k,\omega)=\epsilon_L(k,\omega)\frac{k_ik_j}{k^2}+\epsilon_T(k,\omega)\left(\delta_{ij}-\frac{k_ik_j}{k^2}\right).\quad(1.2)$$

The plasmon or photon damping is related to the dissipative part of the dielectric tensor by the relations (assuming $\gamma_L/\omega_L\ll 1$, $\gamma_T/\omega_T\ll 1$)

$$\gamma_{L,T}(k)/\omega_{L,T}(k)=Z_{L,T}(k)\ \mathrm{Im}\epsilon_{L,T}(k,\omega_{L,T}(k)).\quad(1.3)$$

The plasmon frequency $\omega_L(k)$ is the root of

$$\mathrm{Re}\epsilon_L(k,\omega_L(k))=0\quad(1.4)$$

and the plasmon normalization factor is

$$Z_L{}^{-1}(k)=\tfrac{1}{2}\omega[\partial\ \mathrm{Re}\epsilon_L(k,\omega)/\partial\omega]|_{\omega=\omega_L(k)}.\quad(1.5)$$

In the RPA, (1.1) gives

$$\omega_L{}^2(k)=\omega_p{}^2+k^2\langle v^2\rangle+O(k^4),\qquad Z_L=\left[1-\frac{k^2}{k_D{}^2}\right],\quad(1.6)$$

where

$$n\langle v^2\rangle=\sum_{\text{spins}}\int\frac{d^3p}{(2\pi)^3}\frac{p^2}{m^2}f_p\quad\text{and}\quad k_D{}^2=\omega_p{}^2/\langle v^2\rangle.$$

[13] D. F. DuBois, in *Lectures in Theoretical Physics*, edited by Brittin, Barut, and Guenin (Interscience Publishers, Inc., New York, 1959), Vol. 9C, p. 469.

The transverse photon frequency is determined by

$$\omega_T^2 \,\mathrm{Re}\epsilon_T(k,\omega_T(k)) = k^2c^2 \tag{1.7}$$

and

$$Z_T^{-1}(k) = \frac{k^2c^2}{\omega_T^2} + \frac{\omega}{2}\frac{\partial\,\mathrm{Re}\epsilon_T(k,\omega)}{\partial\omega}\bigg|_{\omega=\omega_T}. \tag{1.8}$$

In the RPA, (1.1) gives

$$\begin{aligned}\omega_T^2 &= \omega_p^2 + k^2c^2,\\ Z_T(k) &= [1-O(k^2)].\end{aligned} \tag{1.9}$$

In the RPA, the Landau damping given by (1.3) and (1.1) is the familiar[3]

$$\gamma_L^0(k) = \frac{4\pi e^2}{k^2}\sum_{\text{spins}}\int\frac{d^3p}{(2\pi)^3}[f_{\mathbf{p}-\frac{1}{2}\mathbf{k}} - f_{\mathbf{p}+\frac{1}{2}\mathbf{k}}] \times\pi\delta(\omega_L(k) - E_{\mathbf{p}+\frac{1}{2}\mathbf{k}} + E_{\mathbf{p}-\frac{1}{2}\mathbf{k}}). \tag{1.10}$$

It is well known that for a degenerate Fermi gas this damping vanishes for $k<k_c$, where

$$\omega_L(k_c) = k_c p_F/m + k_c^2/2m \tag{1.11}$$

and p_F is the Fermi momentum.

The only method presently available to include corrections to the RPA is the many-body perturbation theory in powers of the dimensionless parameter $r_s = r_0/a$, where r_0 is the interelectron spacing ($4\pi r_0^3/3 = n^{-1}$) and a is the Bohr radius, $\hbar^2/me^2$. The expansion in r_s is essentially an expansion in e^2 and is not a simple power series, but more likely some sort of asymptotic expansion. For metals r_s ranges from 1.8 for Al to 5.6 for Cs. Corrections to the RPA value of the specific-heat indicate that the theory does not converge even for r_s as low as 1.8.[9,14] However, the calculated exchange corrections of order r_s to the plasmon energy are very small even for $r_s=5.6$.[8,9,15] Corrections to the RPA value of the correlation energy are estimated to be about 20%.[3] Therefore, it is *a priori* doubtful that the theory is quantitatively accurate at $r_s=1.8$. The purpose of the exercise presented here is to present the correct perturbation calculation of the electron correlational corrections to $\mathrm{Im}\epsilon_{L,T}(k,\omega)$ and to make clear the relationship of such a free electron gas calculation to the actual experimental situation. If the lattice corrections discussed in Sec. IV can be evaluated, it may eventually be possible to test the validity of the calculation against experiment.

II. CALCULATION OF ABSORPTIVE PART OF DIELECTRIC FUNCTION

The method of calculation used here is a simple extension of that used by DuBois and Gilinsky[10] (DG) to calculate the dissipative part of the conductivity tensor of a classical electron gas.[16] This work in turn is an extension of the original calculation in I for the unscreened degenerate electron gas. A formal derivation of the "golden rule" formula from the principles of nonequilibrium quantum statistical mechanics has been given by DuBois.[13] The result is that the dissipative part of the dielectric tensor can be written in the form

$$\begin{aligned}&4\pi e_i e_j\,\mathrm{Im}\epsilon_{ij}(k,\omega)\\ &= \frac{1}{8\omega^2}\sum_{\text{spins}}\int\frac{d^3p_1}{(2\pi)^3}\int\frac{d^3p_2}{(2\pi)^3}\int\frac{d^3p_3}{(2\pi)^3}\int\frac{d^3p_4}{(2\pi)^3}\\ &\times(1-e^{-\beta\omega})f_{p_1}(1-f_{p_2})f_{p_3}(1-f_{p_4})\,|\mathbf{J}\cdot\mathbf{e}|^2\\ &\times(2\pi)^4\delta^3(\mathbf{k}+\mathbf{p}_1+\mathbf{p}_2-\mathbf{p}_3-\mathbf{p}_4)\\ &\times\delta(\omega+E_1+E_2-E_3-E_4).\end{aligned} \tag{2.1}$$

The polarization unit vectors are $\mathbf{e}^{(0)}=\hat{k}$ for the longitudinal case and $\mathbf{e}^{(1,2)}$ for the transverse case, where

$$\sum_{\alpha=1,2} e_i^{(\alpha)}e_j^{(\alpha)} = \delta_{ij} - k_ik_j/k^2. \tag{2.2}$$

In (2.4), $\mathbf{J}$ is a vector amplitude calculated from the diagrams in Fig. 1 according to the rules given in DG[10]

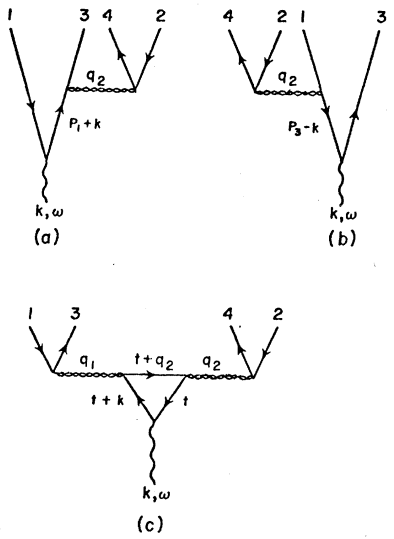

FIG. 1. Three diagrams of order r_s contributing to collisional conductivity. The remaining nine diagrams of this order are obtained from these three by exchange of particles ($3\leftrightarrow4$), exchange of holes ($1\leftrightarrow2$), and exchange of both particles and holes ($1\leftrightarrow2$, $3\leftrightarrow4$). In each case, the intermediate momenta must be relabeled.

[14] B. W. Ninham, Ann. Phys. (N. Y.) **28**, 220 (1964).
[15] R. A. Ferrell, Phys. Rev. **107**, 450 (1957); D. Pines, Physica Suppl. **26**, 103 (1960).

[16] We take this opportunity to list several errata for DG (Ref. 10):
(i) A serious misprint is found in the square bracket on the right-hand side of (3.78) which should read
$$[1;\ (u_1u_2/q^2);\ (u_1^2/q^2)+2].$$
(ii) There are several errors in signs in the equations leading up to (3.22) and in (3.22), (3.25), and (3.35) the variables p_1 and p_2 should be interchanged.
The results (3.29)–(3.33) and (3.37)–(3.38) are unchanged.

or in Ref. 13. This amplitude differs from that for the classical electron gas considered previously only in that electron spin and electron exchange are now taken into account. The result is

$$\begin{aligned}\mathbf{J}\cdot\mathbf{e}=&i4\pi e^2(-i\sqrt{(4\pi e^2)/m})i\\&\times\{(X_3^\dagger X_1)(X_4^\dagger X_2)[V(\mathbf{p}_4-\mathbf{p}_2,\,E_4-E_2)C_k(\mathbf{p}_1,\mathbf{p}_3)\\&+V(\mathbf{p}_3-\mathbf{p}_1,\,E_3-E_1)C_k(\mathbf{p}_2,\mathbf{p}_4)\\&-4\pi e^2V(\mathbf{p}_4-\mathbf{p}_2,\,E_4-E_2)V(\mathbf{p}_3-\mathbf{p}_1,\,E_3-E_1)\\&\times\mathbf{e}\cdot\mathbf{T}(\mathbf{p}_3-\mathbf{p}_1,\,E_3-E_1;\,\mathbf{p}_4-\mathbf{p}_2;\,E_4-E_2)]\\&-(X_4^\dagger X_1)(X_3^\dagger X_2)[V(\mathbf{p}_3-\mathbf{p}_2,\,E_3-E_2)C_k(\mathbf{p}_1,\mathbf{p}_4)\\&+V(\mathbf{p}_4-\mathbf{p}_1,\,E_4-E_1)C_k(\mathbf{p}_2,\mathbf{p}_3)\\&-4\pi e^2V(\mathbf{p}_3-\mathbf{p}_2,\,E_3-E_2)V(\mathbf{p}_4-\mathbf{p}_1,\,E_4-E_1)\\&\times\mathbf{e}\cdot\mathbf{T}(\mathbf{p}_3-\mathbf{p}_2,\,E_3-E_2;\,\mathbf{p}_4-\mathbf{p}_1,\,E_4-E_1)]\},\end{aligned}\tag{2.3}$$

with

$$C_k(\mathbf{p}_i,\mathbf{p}_j)=\frac{(\mathbf{p}_i+\tfrac{1}{2}\mathbf{k})\cdot\mathbf{e}}{\omega-E_{\mathbf{p}_i+\mathbf{k}}+E_i}-\frac{(\mathbf{p}_j-\tfrac{1}{2}\mathbf{k})\cdot\mathbf{e}}{\omega+E_{\mathbf{p}_j-\mathbf{k}}-E_j}\tag{2.4}$$

and X_i the spin-wave function.

The amplitude $\mathbf{T}$ arises from the closed triangular loop in diagram C of Fig. 1 and its exchange counterparts. In terms of the variables,

$$\mathbf{q}_1=\mathbf{p}_3-\mathbf{p}_1,\quad u_1=E_3-E_1,$$
$$\mathbf{q}_2=\mathbf{p}_4-\mathbf{p}_2,\quad u_2=E_4-E_2,$$

we can write[10]

$$T_i(q_1,u_1,q_2,u_2;\,k,\omega)=\tau_i^+(q_1,u_1;\,k,\omega)+\tau_i^+(q_2,u_2;\,k,\omega),\tag{2.5}$$

$$\begin{aligned}\tau_i^+(q,u;\,k,\omega)=&\frac{4\pi e^2}{m}\sum_{\text{spins}}\int\frac{d^3t}{(2\pi)^3}(\mathbf{t}+\tfrac{1}{2}\mathbf{k})_i\frac{1}{\omega+E_\mathbf{t}-E_{\mathbf{t}+\mathbf{k}}}\\&\times\left\{\frac{f_{\mathbf{t}+\mathbf{k}}-f_{\mathbf{t}+\mathbf{q}}}{u-\omega+E_{\mathbf{t}+\mathbf{k}}-E_{\mathbf{t}+\mathbf{q}}+i\epsilon}\right.\\&\left.-\frac{f_\mathbf{t}-f_{\mathbf{t}+\mathbf{q}}}{u+E_\mathbf{t}-E_{\mathbf{t}+\mathbf{q}}+i\epsilon}\right\}.\end{aligned}\tag{2.6}$$

In (2.2), $V(q,\omega)$ is the dynamically screened interaction

$$V(q,\omega)=1/q^2\epsilon_L{}^0(q,\omega),\tag{2.7}$$

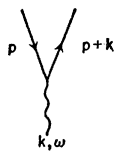

FIG. 2. Diagram for Landau damping.

where $\epsilon_L{}^0(q,\omega)$ is the RPA longitudinal dielectric function

$$\epsilon_L{}^0(q,\omega)=1+\frac{4\pi e^2}{q^2}\sum_{\text{spins}}\int d^3t\frac{f_\mathbf{t}-f_{\mathbf{t}+\mathbf{q}}}{\omega-E_{\mathbf{t}+\mathbf{q}}+E_\mathbf{t}+i\epsilon}.\tag{2.8}$$

This expression follows from (1.1) and the condition of longitudinal current conservation. This expression is valid to lowest order in the potential (i.e., in r_s) provided that $k<k_c$ so that the Landau damping process corresponding to the diagram in Fig. 2 is zero. [For $k>k_c$, the expression is not valid; and problems arise in (2.1)–(2.6) because of vanishing energy denominators. In this case, the Landau damping is so strong that the correlation corrections are not of interest.]

Equation (2.1) is finite for $k<k_c$ (but anomalously large) even when $V(q,\omega)$ is replaced by the unscreened q^{-2}, in which case it is consistent to neglect the diagrams with the triangular loops as was done in I, since these are formally of higher order in e^2 or r_s. However, when the full screened interaction is included, these diagrams must also be included, since they are of the same order in e^2 as the screening corrections (see Fig. 3). Ninham[4] included screening but neglected the triangular amplitude. The reduction of the complicated expressions (2.1)–(2.8) to a useful form follows the same steps as in DG so the details will be omitted here. The amplitudes are expanded in powers of $\mathbf{k}\cdot\mathbf{p}/m\omega$. Within the amplitude, many cancellations occur which result in the vanishing of all terms of order one and leave a leading term of order k. If the triangular amplitudes are not included, the cancellation does *not* occur; and one should incorrectly obtain a leading term of order one. Ninham, who neglects the triangles, in fact obtains an amplitude of order k because of some additional and, in our opinion, ad hoc approximations.

Following the procedure of DG, we can reduce (2.1) to the form

$$\begin{aligned}e_ie_j\,\mathrm{Im}\epsilon_{ij}(k,\omega)=&\frac{1}{8\omega^2}(1-e^{\beta\omega})\sum_{\text{spins}}\int\frac{d^3p_1}{(2\pi)^3}\int\frac{d^3p_2}{(2\pi)^3}\int\frac{d^3q}{(2\pi)^3}\int du_1\int du_2\,f_{p1}(1-f_{\mathbf{p}1+\mathbf{q}})f_{p2}(1-f_{\mathbf{p}2-\mathbf{q}})(2\pi)\delta(u_1+u_2-\omega)\\&\times\delta\left(u_1-\frac{\mathbf{q}\cdot\mathbf{p}_1}{m}-\frac{1}{2m}q^2\right)\delta\left(u_2-\frac{\mathbf{q}\cdot\mathbf{p}_2}{m}-\frac{1}{2m}q^2\right)\frac{(4\pi e^2)^3}{m^2}\frac{k^2}{\omega^2}\{|V(\mathbf{q},u_1)|^2|V(\mathbf{q},u_2)|^2|\mathbf{e}\cdot\mathbf{M}(\mathbf{q},\mathbf{k};\,\mathbf{p}_1,\mathbf{p}_2,u_1,u_2)|^2\\&-\mathrm{Re}[V(\mathbf{q},u_1)V^*(\mathbf{q}',\bar{u}_1)V(\mathbf{q},u_2)V^*(\mathbf{q}',\bar{u}_2)\mathbf{e}\cdot\mathbf{M}(\mathbf{q},\mathbf{k};\,\mathbf{p}_1,\mathbf{p}_2,u_1,u_2)\mathbf{e}\cdot\mathbf{M}^*(\mathbf{q}',\mathbf{k};\,\mathbf{p}_1,\mathbf{p}_2,\bar{u}_1,\bar{u}_2)]\},\end{aligned}\tag{2.9}$$

where

$$M=q^2(\mathbf{k}\cdot\mathbf{e})-4(\mathbf{k}\cdot\mathbf{q})(\mathbf{q}\cdot\mathbf{e})-\frac{V^{-1}(\mathbf{q},u_1)}{\omega}\left[\frac{\mathbf{k}\cdot\mathbf{q}}{m}\mathbf{e}\cdot\mathbf{p}_1+\frac{\mathbf{k}\cdot\mathbf{p}_1}{m}\mathbf{e}\cdot\mathbf{q}-\frac{2\mathbf{e}\cdot\mathbf{q}\mathbf{q}\cdot\mathbf{k}}{q^2}u_1\left(1-\frac{q^2}{2u_1m}\right)\right]$$
$$+\frac{V^{-1}(\mathbf{q},u_2)}{\omega}\left[\frac{\mathbf{k}\cdot\mathbf{q}}{m}\mathbf{e}\cdot\mathbf{p}_2+\frac{\mathbf{k}\cdot\mathbf{p}_2}{m}\mathbf{e}\cdot\mathbf{q}+\frac{2\mathbf{e}\cdot\mathbf{q}\mathbf{q}\cdot\mathbf{k}}{q^2}u_2\left(1-\frac{q^2}{2u_2m}\right)\right]. \quad (2.10)$$

The exchange momentum and energy transfer variables are

$$\mathbf{q}'=\mathbf{q}+\mathbf{p}_1-\mathbf{p}_2\,,\quad \bar{u}_1=u_1+E_4-E_3\,,\quad \bar{u}_2=u_2+E_2-E_1. \quad (2.11)$$

It is interesting to note that the amplitude M is the same as in the classical case [DG Eq. (3.22)] except for the factors of $(1-q^2/2u_1m)$ and $(1-q^2/2u_2m)$ which are unity for the classical case, as can be seen by restoring the $\hbar$ dependence explicitly.

The next step is to average over the directions of $\hat{k}$ since the system is assumed isotropic. We then obtain the analog of DG, Eqs. (3.29) and (3.37). The exchange corrections can be dropped to lowest order as noted in I since they make a contribution one order in r_s smaller than the direct terms. We then have

$$\mathrm{Im}\epsilon_{L,T}(k,\omega)=\frac{1}{60\pi}\frac{(4\pi e^2)}{m^2}\frac{k^2}{\omega^5}\frac{(1-e^{-\beta\omega})}{\omega}\int dq\int du\frac{1}{(e^{-\beta\{\frac{1}{2}\omega+u\}}-1)}\mathrm{Im}\frac{1}{\epsilon_L{}^0(q,\frac{1}{2}\omega+u)}\frac{1}{(e^{-\beta\{\frac{1}{2}\omega-u\}}-1)}\mathrm{Im}\frac{1}{\epsilon_L{}^0(q,\frac{1}{2}\omega-u)}$$
$$\times[\alpha_{L,T}q^2\omega^2+\tfrac{1}{2}\beta_{L,T}v_\perp{}^2(q,\tfrac{1}{2}\omega+u)q^4|\,\epsilon_L{}^0(q,\tfrac{1}{2}\omega+u)|^2+\tfrac{1}{2}\beta_{L,T}v_\perp{}^2(q,\tfrac{1}{2}\omega-u)q^4|\,\epsilon_L{}^0(q,\tfrac{1}{2}\omega-u)|^2]. \quad (2.12)$$

Here

$$\alpha_L=23\,,\quad \beta_L=16\,,\quad \alpha_T=16\,,\quad \beta_T=12. \quad (2.13)$$

In this expression, $\epsilon_L{}^0(q,\omega)$ is the RPA dielectric function given in (2.8). We have defined the quantity $v_\perp{}^2(q,u)$, which is the averaged square of the component of p perpendicular to q consistent with energy conservation as

$$v_\perp{}^2(q,u)=\frac{(1/2m^2)\int d^3p\; f_p(1-f_{p+q})[p^2-(\mathbf{p}\cdot\mathbf{q})^2/q^2]\delta[u-(\mathbf{p}\cdot\mathbf{q})/m-(q^2/2m)]}{\int d^3p\; f_p(1-f_{p+q})\delta(u-(\mathbf{p}\cdot\mathbf{q})/m-q^2/2m)}. \quad (2.14)$$

This quantity can be reduced to an expression involving only the RPA dielectric functions. From Eq. (1.1), it can be shown that

$$v_\perp{}^2(q,u)=\frac{u^2}{2q^2}\frac{\mathrm{Im}\epsilon_T{}^0(\mathbf{q},u)}{\mathrm{Im}\epsilon_L{}^0(\mathbf{q},u)}. \quad (2.15)$$

For classical statistics ($f_p\ll 1$), we find

$$v_\perp{}^2=m/(\beta)=mk_\beta T.$$

Using this result, it is easily seen that (2.12) reduces to Eqs. (3.29) and (3.37) of DG in the limit of classical statistics. It is convenient to express the result in terms of two integrals I and J as done in DG. For Fermi-Dirac statistics, we have

$$\mathrm{Im}\epsilon_{L,T}(k,\omega)=\frac{1}{60\pi^3}\frac{(4\pi e^2)}{m^2}\frac{k^2}{\omega^5}\frac{(1-e^{-\beta\omega})}{\omega}$$
$$\times\{\alpha_{L,T}J(\omega)+\beta_{L,T}I(\omega)\}\,, \quad (2.16)$$

where

$$J(\omega)=\omega^2\int_0^\infty dq\,q^2\int_{-\infty}^\infty du\frac{1}{e^{-\beta(\frac{1}{2}\omega+u)}-1}\frac{1}{e^{-\beta(\frac{1}{2}\omega-u)}-1}$$
$$\times\mathrm{Im}\frac{1}{\epsilon_L{}^0(q,\frac{1}{2}\omega+u)}\mathrm{Im}\frac{1}{\epsilon_L{}^0(q,\frac{1}{2}\omega-u)}, \quad (2.17)$$

$$I(\omega)=-\frac{1}{2}\int_0^\infty dq\,q^2\int_{-\infty}^\infty du(\tfrac{1}{2}\omega+u)^2$$
$$\times\frac{1}{e^{-\beta(\frac{1}{2}\omega+u)}-1}\frac{1}{e^{-\beta(\frac{1}{2}\omega-u)}-1}$$
$$\times\mathrm{Im}\epsilon_T{}^0(q,\tfrac{1}{2}\omega+u)\,\mathrm{Im}\frac{1}{\epsilon_L{}^0(q,\frac{1}{2}\omega-u)}. \quad (2.18)$$

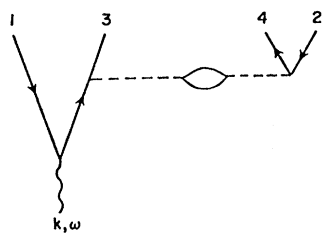

FIG. 3. Diagram of typical contribution to the screened interaction. This diagram which is included in Fig. 1(a) is of the same order in e^2 as the lowest-order contribution from the triangle diagram, Fig. 1(c).

Note that these integrands are now expressed entirely in terms of RPA dielectric functions $\epsilon_{L,T}^0(q, \frac{1}{2}\omega \pm u)$.

III. DEGENERATE ELECTRON GAS

A. Zero-Temperature Limit

In this section, we will work with the $T=0$ $(\beta \to \infty)$ limit of equations (2.16)–(2.18). We also introduce a system of units in which momentum is expressed in units of the Fermi momentum $p_F=\hbar/(\alpha r_0)$ where $\alpha=(4/9\pi)^{1/3}$ and r_0 is the average interelectron radius $[(4\pi/3)r_0^3=n^{-1}]$. Energy is expressed in units of $2E_F=p_F/m$, where E_F is the Fermi energy. In these units, the plasmon energy is

$$\omega_p=(4\alpha r_s/3\pi)^{1/3} \tag{3.1}$$

and $r_s=r_0/a$, where $a=\hbar^2/me^2$, the Bohr radius. In this notation, we write (2.16) in the limit $\beta \to \infty$,

$$\mathrm{Im}\epsilon_{L,T}(k,\omega)=\frac{1}{20\pi}\left(\frac{4\alpha r_s}{3\pi}\right)\frac{k^2}{\omega^6}[\alpha_{L,T}J(\omega)+\beta_{L,T}I(\omega)] \tag{3.2}$$

and (2.17) and (2.18) become

$$J(\omega)=\omega^2\int_0^\infty dq\, q^2\int_{-\frac{1}{2}\omega}^{\frac{1}{2}\omega} du\ \mathrm{Im}\frac{1}{\epsilon_L^0(q,\frac{1}{2}\omega+u)}\ \mathrm{Im}\frac{1}{\epsilon_L^0(q,\frac{1}{2}\omega-u)}, \tag{3.3}$$

$$I(\omega)=-\frac{1}{2}\int_0^\infty dq\, q^2\int_{-\frac{1}{2}\omega}^{\frac{1}{2}\omega} du(\tfrac{1}{2}\omega+u)^2\times\mathrm{Im}\epsilon_T^0(q,\tfrac{1}{2}\omega+u)\ \mathrm{Im}\frac{1}{\epsilon_L^0(q,\frac{1}{2}\omega-u)}. \tag{3.4}$$

From Eq. (1.1) an expression for $\mathrm{Im}\epsilon_T^0(q,u)$ in the RPA is easily obtained for a degenerate Fermi distribution for $u>0$:

$$\begin{aligned}
\mathrm{Im}\epsilon_T^0(q,u)&=\frac{2\alpha r_s}{\pi q}\left(1-\frac{u^2}{q^2}-\frac{q^2}{4}\right), && \text{for } \left(\frac{u+\frac{1}{2}q^2}{q}\right)^2<1\\
&=\frac{\alpha r_s}{2qu^2}\left(1-\left[\frac{u-\frac{1}{2}q^2}{q}\right]^2\right), && \text{for } \left(\frac{u+\frac{1}{2}q^2}{q}\right)^2>1>\left(\frac{u-\frac{1}{2}q^2}{q}\right)^2\\
&=0, && \text{for } \left(\frac{u-\frac{1}{2}q^2}{q}\right)^2>1.
\end{aligned} \tag{3.5}$$

The result of evaluating $\epsilon_L^0(q,\omega)$ [Eq. (2.8)] in this limit is well known,[3] but we give the result for easy reference:

$$\mathrm{Re}\epsilon_L^0(q,u)=1+\frac{2\alpha r_s}{\pi q^2}\left\{1+\left[\frac{(u+\frac{1}{2}q^2)^2-q^2}{2q^3}\right]\ln\left[\frac{u+\frac{1}{2}q^2-q}{u+\frac{1}{2}q^2+q}\right]+\left[\frac{(u-\frac{1}{2}q^2)^2-q^2}{2q^3}\right]\ln\left[\frac{u-\frac{1}{2}q^2+q}{u-\frac{1}{2}q^2-q}\right]\right\}, \tag{3.6}$$

$$\begin{aligned}
\mathrm{Im}\epsilon_L^0(q,u)&=\frac{\alpha r_s}{q^3}2u, && \text{for } \left(\frac{u+\frac{1}{2}q^2}{q}\right)^2<1\\
&=\frac{\alpha r_s}{q^3}\left(1-\frac{(u-\frac{1}{2}q^2)^2}{q^2}\right), && \text{for } \left(\frac{u+\frac{1}{2}q^2}{q}\right)^2>1>\left(\frac{u-\frac{1}{2}q^2}{q}\right)^2\\
&=0, && \text{for } \left(\frac{u-\frac{1}{2}q^2}{q}\right)^2>1.
\end{aligned} \tag{3.7}$$

In the region $\mathrm{Im}\epsilon_L^0(q,\omega)=0$, it is well known that $\mathrm{Re}\epsilon_L^0(q,\omega_L(q))=0$ at

$$\omega_L(q)=\omega_p+3q^2/10\omega_p+\cdots. \tag{3.8}$$

This pole provides the plasmon resonance contribution, and we must write

$$-\mathrm{Im}\frac{1}{\epsilon_L^0(q,\omega)}=\frac{\pi}{2}\omega_L(q)Z_L^0(q)\delta(\omega-\omega_L(q))+\frac{\mathrm{Im}\epsilon_L^0(q,\omega)}{|\epsilon_L^0(q,\omega)|^2},\quad q<k_c \tag{3.9a}$$

$$=+\frac{\mathrm{Im}\epsilon_L{}^0(q,\omega)}{|\epsilon_L{}^0(q,\omega)|^2}, \quad q>k_c \tag{3.9b}$$

where k_c is the plasmon cutoff wave number, determined by the condition that

$$\begin{aligned}\epsilon_L{}^0(q,\omega_L(q))&=0, \quad q<k_c\\ &\neq 0, \quad q>k_c.\end{aligned} \tag{3.10}$$

The residue factor is

$$Z_L{}^{-1}(q)=\frac{\omega_L(q)}{2}\frac{\partial}{\partial\omega}\mathrm{Re}\epsilon_L{}^0(q,\omega)\Big|_{\omega=\omega_L(q)}$$
$$=1+\frac{3}{5}\frac{q^2}{\omega_p{}^2}+\cdots. \tag{3.11}$$

In the region $\mathrm{Im}\epsilon_L{}^0(q,\omega)\neq 0$, the term $\mathrm{Im}\epsilon_L{}^0\cdot|\epsilon_L{}^0|^{-2}$ represents the screened electron-hole pair continuum.

For practical calculations, we must separate out these contributions and, thus, obtain a partition of I and J into pair-pair (pr-pr), plasmon-pair (pl-pr) and plasmon-plasmon (pl-pl) contributions. Then, $\mathrm{Im}\epsilon_{L,T}(k,\omega)$ is a sum of the following contributions:

$$\mathrm{Im}\epsilon_{L,T}(k,\omega)_{\text{pr-pr}}=\frac{1}{20\pi}\left(\frac{4\alpha r_s}{3\pi}\right)\frac{k^2}{\omega^6}\int_0^\infty dq\, q^2\int_{-\frac{1}{2}\omega}^{\frac{1}{2}\omega}du\frac{\mathrm{Im}\epsilon_L{}^0(q,\frac{1}{2}\omega-u)}{|\epsilon_L{}^0(q,\frac{1}{2}\omega-u)|^2}$$
$$\times\left[\beta_{L,T}\tfrac{1}{2}(\tfrac{1}{2}\omega+u)^2\,\mathrm{Im}\epsilon_T{}^0(q,\tfrac{1}{2}\omega+u)+\alpha_{T,L}\omega^2\frac{\mathrm{Im}\epsilon_L{}^0(q,\frac{1}{2}\omega+u)}{|\epsilon_L{}^0(q,\frac{1}{2}\omega+u)|^2}\right], \tag{3.12}$$

$$\mathrm{Im}\epsilon_{L,T}(k,\omega)_{\text{pr-pl}}=\frac{1}{20\pi}\left(\frac{4\alpha r_s}{3\pi}\right)\frac{k^2}{\omega^6}\pi\int_0^{k_c}dq\, q^2Z_L(q)\omega_L(q)\eta(\omega-\omega_L(q))$$
$$\times\left[\beta_{L,T}\tfrac{1}{2}(\omega-\omega_L(q))^2\,\mathrm{Im}\epsilon_T{}^0(q,\omega-\omega_L(q))+2\alpha_{T,L}\omega^2\frac{\mathrm{Im}\epsilon_L{}^0(q,\omega-\omega_L(q))}{|\epsilon_L{}^0(q,\omega-\omega_L(q))|^2}\right], \tag{3.13}$$

$$\mathrm{Im}\epsilon_{L,T}(k,\omega)_{\text{pl-pl}}=\frac{1}{20\pi}\left(\frac{4\alpha r_s}{3\pi}\right)\frac{k^2}{\omega^6}\pi^2\alpha_{T,L}\omega^2\int_0^{k_c}dq\, q^2Z_L{}^2(q)\omega_L{}^2(q)\delta(\omega-2\omega_L(q)). \tag{3.14}$$

In (3.13) η is the step function

$$\eta(x)=\tfrac{1}{2}(1+x/|x|). \tag{3.15}$$

The range of integration in (3.12) and (3.13) is implicitly understood to be over the domain of q and ω for which which $\mathrm{Im}\epsilon_{L,T}{}^0(q,\frac{1}{2}\omega\pm u)$ is nonzero, i.e., over the pair continuum, since the plasmon pole contribution has been explicitly separated out.

B. Plasmon Damping

The pair-plasmon (pr-pl) and (pl-pl) contributions to the damping of a plasmon of frequency ω_p are zero since $\omega=\omega_p$ is at or below threshold for these processes. [If we substitute $\omega=\omega_L(k)$ and consider the full k dependence of $\omega_L(k)$, then it is easily shown from (3.13) that $\mathrm{Im}\epsilon_L{}^0(k,\omega_L(k))_{\text{pr-pl}}$ is of order k^4 which is very small. This process was estimated in I and by Ninham[4] to be of order k^4; but in both calculations, the triangle amplitudes were ignored. We will not consider terms of order k^4 in this paper.] Thus, the largest term is $\mathrm{Im}\epsilon_L(k,\omega_p)_{\text{pr-pr}}$. If we substitute the explicit expressions for $\epsilon_{L,T}{}^0(q,\omega)$ from (1.1) into (3.2)–(3.4), we find

$$\mathrm{Im}\epsilon_L(k,\omega_p)_{\text{pr-pr}}=\frac{\gamma_L(k)_{\text{pr-pr}}}{\omega_p}=\frac{9}{40}\frac{k^2}{\pi}\omega_p{}^2\int\frac{dq}{q^2}\int_{\substack{p_1<1\\ |\mathbf{p}_1+\mathbf{q}|>1}}d^3p_1\int_{\substack{p_2<1\\ |\mathbf{p}_2+\mathbf{q}|>1}}d^3p_2\;\delta(\mathbf{q}\cdot\mathbf{q}'-\omega_p)$$
$$\times\frac{1}{|\epsilon_L{}^0(q,\mathbf{q}\cdot\mathbf{p}_2+\frac{1}{2}q^2)|^2}\left[\frac{q^2}{\omega_p{}^2}\left(p_1{}^2-\frac{(\mathbf{p}_1\cdot\mathbf{q})^2}{q^2}\right)+\frac{23}{8}\frac{1}{|\epsilon_L{}^0(q,\mathbf{p}_1\cdot\mathbf{q}+\frac{1}{2}q^2)|^2}\right], \tag{3.16}$$

where $\mathbf{q}'=\mathbf{p}_1+\mathbf{p}_2+\mathbf{q}$. If we set the $\epsilon_L{}^0$ factors in the denominators of (3.16) equal to unity, we obtain exactly one-fourth of the unscreened result of I. The discrepancy of a factor of 4 resulted from a numerical error in I.

It is most convenient to use the form of Eq. (3.12) to

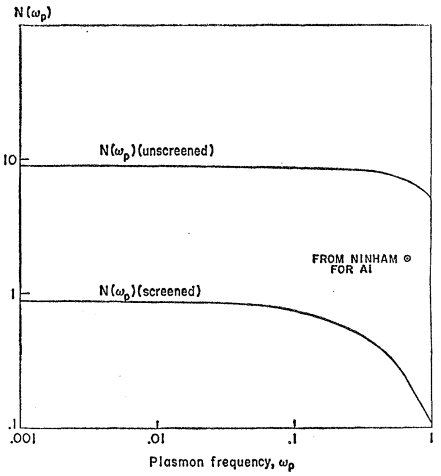

FIG. 4. Plot of $N(\omega_p)$ of Eq. (3.20) versus the plasmon frequency ω_p for both unscreened and dynamically screened interactions. The point calculated by Ninham (Ref. 4) for Al is plotted for comparison.

evaluate the integral. We introduce the variables $x=\omega_p/q$ and $y=u/q$ and define the functions $\tilde{\epsilon}_{L,T}{}^0(x,y)$ by the identity

$$\tilde{\epsilon}_{L,T}{}^0(x,y)=\epsilon_{L,T}{}^0(\omega_p/x,\ \omega_p y/x). \tag{3.17}$$

This substitution into (3.12) leads to

$$\mathrm{Im}\epsilon_L(k)_{\text{pr-pr}}=\frac{4\omega_p{}^3k^2}{5\pi}\int_0^\infty\frac{dx}{x^5}\int_{-x/2}^{x/2}dy \times\frac{\mathrm{Im}\tilde{\epsilon}_L{}^0(x,\,x/2-y)}{|\tilde{\epsilon}_L{}^0(x,\,x/2+y)|^2}\frac{1}{16}\left\{\frac{23\ \mathrm{Im}\tilde{\epsilon}_L{}^0(x,\,x/2+y)}{|\tilde{\epsilon}_L{}^0(x,\,x/2+y)|^2}+\frac{8(x/2+y)^2}{x^2}\mathrm{Im}\tilde{\epsilon}_T{}^0(x,\,x/2+y)\right\}. \tag{3.18}$$

In the limit $\omega_p\ll1$ and $|\tilde{\epsilon}_L{}^0|^2=1$ (i.e., no screening), the integration can be carried out analytically using (3.5) and (3.7), with the result

$$\gamma_{\text{pr-pr}}{}^{\text{uns}}(k,\omega_p)=\frac{3\pi\omega_p{}^3k^2}{80}[34-22\ \ln 2], \tag{3.19}$$

which is smaller by a factor of 4 than the result in I.[17] We had noted this discrepancy already in comparing the form of Eq. (3.16) with I.

The fully screened damping must be evaluated from Eq. (3.18) by numerical integration. The damping is conveniently written as

$$\gamma_{L\,\text{pr-pr}}=\omega_p\ \mathrm{Im}\epsilon_{L\,\text{pr-pr}}=\frac{4k^2\omega_p{}^3}{5\pi}N(\omega_p), \tag{3.20}$$

where $N(\omega_p)$ is the integral in (3.18) and depends on $\omega_p(\omega_p=(4\alpha r_s/3\pi)^{1/2})$ through the definition of $\tilde{\epsilon}_L{}^0(x,y)$. The function $N(\omega_p)$ is plotted in Fig. 4 for both the screened and unscreened cases. In both cases, it has a nonvanishing limit as $\omega_p\to0$. In this limit, screening reduces the damping by a factor of 10. At larger ω_p, for which the theory begins to break down, we can compare our results with previous work.

For Al where $\omega_p=\frac{2}{3}$, we have indicated a value of $N(\omega_p)$ which we obtain from Ninham's calculations (Ninham estimates the effect of screening only for frequencies of the order of $\frac{2}{3}$). We find that the damping is about $\frac{1}{6}$ the damping calculated by Ninham. Explicitly, we have for aluminum, converting to ordinary (cgs) units,

$$\gamma_L(k,\tfrac{2}{3})=\frac{4}{5\pi}(\tfrac{2}{3})^3N(\tfrac{2}{3})\frac{k^2}{p_F{}^2}(2E_F) =(5.6\times10^{-1})\frac{k^2}{p_F{}^2}\ \text{eV}. \tag{3.21}$$

The experiments of Kunz,[11] Powell and Swanson,[4] and Festenberg[12] are in rather good agreement and produce essentially the same γ-versus-k curves *if* restricted to wavelengths smaller than the mean crystallite size (see Festenberg[12]). To compare with the experiments of Powell and Swanson,[4] we write the coefficient B of their observations in terms of γ as

$$B=(\gamma/k^2)(E_{\text{el}}/2E_F), \tag{3.22}$$

where E_{el} is the energy of the incident electrons.

At $\omega_p=\frac{2}{3}$, this gives $B=450$ eV/(rad)^2, whereas, the experimental value is 3.5×10^3 eV/(rad)^2.

C. Optical Absorption into a Plasmon-Pair Final State

Next, we briefly dispose of the electron correlation corrections to optical absorption. We note that $\mathrm{Im}\epsilon_T(k,\omega)$ is proportional to $k^2/p_F{}^2$ in ordinary units. From (1.3) and (1.9), we obtain the optical absorption $\gamma_T(k)$ by setting $\omega^2=k^2c^2+\omega_p{}^2$ in $\mathrm{Im}\epsilon_T{}^0(k,\omega)$. The factor $k^2/p_F{}^2$ can then be written

$$\frac{\omega^2-\omega_p{}^2}{p_F{}^2}\frac{1}{c^2}=\left(\frac{\omega^2}{\omega_p{}^2}-1\right)\frac{k_D{}^2}{p_F{}^2}\frac{\langle v^2\rangle}{c^2},$$

which is less than or of the order $\langle v^2\rangle/c^2$ for $\omega\sim\omega_p$ since $k_D{}^2/p_F{}^2=O(r_s)$. This factor is of the order of 10^{-5} for metals. Because of this small factor, which arises from the relatively long wavelength of light for ω near ω_p, the effects of electron correlations on the transverse absorption in this region are unobservably small.

Several authors[5,6] have proposed that optical absorption into a final state of a pair-plus a plasmon would be an observable effect. We can identify the contribution from this final state as arising from (3.13). Previous calculations did not find this absorption proportional to

[17] Note that there is also a misprint in I such that 2(34–22 ln2) is given as (70–44 ln2). From Ninham's result for the unscreened damping [see his Eq. (10) with the expression in brackets as described in the following sentence which follows his equation], we would obtain $\gamma_{\text{pr-pr}}{}^{\text{uns}}=\mathrm{Im}\epsilon_{\text{pr-pr}}=\frac{1}{2}\Gamma_{(1)}=(3\pi/10)q^2\omega_p{}^2(34-22\ \ln2)$. Therefore, our calculation yields only $\frac{1}{8}$ of Ninham's value in the unscreened case.

the small factor k^2. This is due to the omission in all these calculations of the triangle amplitudes. The inclusion of these terms, as we have pointed out several times, results in additional cancellations which greatly reduce the magnitude of this absorption. We will, therefore, not consider further details of this relatively insignificant process here. Lattice effects discussed in Sec. IV may increase the magnitude of this process, but the required calculations remain to be done.

IV. EFFECTS OF POSITIVE ION LATTICE

The periodic ion lattice adds important modifications to the free electron gas model. In this paper, we will not attempt to include these effects; but it is important to list them, to place the free electron gas calculations into the proper context. Detailed calculations are, of course, needed to determine the size of these modifications.

To include the periodic lattice in the many-body theory, we clearly must replace the plane wave states which we have been using by Bloch waves which can be expressed in the form

$$\varphi_{p,l}(x)=e^{ip\cdot x}\sum_{\mathbf{K}_n} e^{i\mathbf{K}_n\cdot\mathbf{x}}a_{n,p}{}^{l}. \tag{4.1}$$

Here, the lattice momentum p is restricted to the first Brillouin zone and the sum is over all reciprocal lattice vectors $\mathbf{K}_n$. The index l labels the bands which have energies $E_p{}^l$. The expansion coefficients $a_{n,p}{}^l$ completely determine the state.

The general formula for $\epsilon_{ij}{}^0(k,\omega)$ in the RPA or collisionless approximation can be written as

$$\epsilon_{ij}{}^0(k,\omega)=\delta_{ij}\left(1-\frac{4\pi e^2 n}{m\omega^2}\right)+\frac{4\pi e^2}{m^2\Omega\omega^2}\sum_{p,l,l'}\langle\mathbf{p}+\mathbf{k},l'|\left(\mathbf{P}+\frac{\mathbf{k}}{2}\right)_i|\mathbf{p},l\rangle\times\langle\mathbf{p},l|\left(\mathbf{P}+\frac{\mathbf{k}}{2}\right)_j|\mathbf{p}+\mathbf{k},l'\rangle\frac{f(E_{\mathbf{p}}{}^l)-f(E_{\mathbf{p}+\mathbf{k}}{}^{l'})}{\omega-E_{\mathbf{p}+\mathbf{k}}{}^{l'}+E_{\mathbf{p}}{}^l+i\epsilon}, \tag{4.2}$$

where Ω is the total volume and we define the matrix element

$$\langle\mathbf{p}',l'|\left(\mathbf{P}+\frac{\mathbf{k}}{2}\right)_i|\mathbf{p},l\rangle=\int d^3r\, e^{i\mathbf{k}\cdot\mathbf{r}}\lim_{r'\to r}\left[\frac{(\nabla_{r'}-\nabla_r)}{2i}\varphi_{\mathbf{p}'l'}{}^*(r)\varphi_{\mathbf{p},l}(r')\right]. \tag{4.3}$$

This formula is equivalent to that derived by Adler[18] but neglects the local field corrections to the $l\neq l'$ terms arising from umklapp processes which were first calculated by Adler and which are small for Al.

[18] S. Adler, Phys. Rev. **126**, 413 (1963).

For $l=l'$, we have intraband contributions which are the analog of the free electron gas effects. In a crystal with cubic symmetry, we can again separate transverse and longitudinal components.[18] For the longitudinal components, we can write

$$\langle\mathbf{p}+\mathbf{k},l|\left(\mathbf{P}+\frac{\mathbf{k}}{2}\right)\cdot\hat{k}|\mathbf{p},l\rangle=\frac{(E_{\mathbf{p}+\mathbf{k}}{}^l-E_{\mathbf{p}}{}^l)}{k}\int d^3r\, e^{i\mathbf{k}\cdot\mathbf{r}}\varphi_{\mathbf{p}+\mathbf{k},l}{}^*(r)\varphi_{\mathbf{p},l}(r) \tag{4.4}$$

by using current conservation. In the limit $k\to 0$, we have, because of the orthonormality of the φ's,

$$\lim_{k\to 0}\langle\mathbf{p}+\mathbf{k},l|\left(\mathbf{P}+\frac{\mathbf{k}}{2}\right)\cdot\mathbf{k}|\mathbf{p},l\rangle=\frac{\mathbf{k}\cdot\partial E^l/\partial\mathbf{p}}{k}[1+O(k)]. \tag{4.5}$$

This result is the same as that for the free electron gas result in this limit if $E_p{}^l=p^2/2m^*$. The intraband Landau damping can be treated in the same way as the free electron case. For more details on this, we refer to the review of Pines.[3]

This collisionless approximation is not suitable for discussing real materials as is well known. Collisions with impurities, phonons, defects, and perhaps electron-electron umklapp collisions provide absorptive mechanisms even in the case $k=0$. It is usually assumed that the effect of these collisions can be described by replacing ω in (4.2) by $\omega-i/\tau$, where τ is an appropriate collision time. Such a collision significantly modifies only the intraband terms ($l=l'$) since the interband energies are generally much larger than $\hbar/\tau$. Without repeating details, we note that τ for Al has been estimated by Erhenreich, Phillip, and Segal[19] from low-frequency optical data ($k=0$) to be

$$\tau\simeq 5.12\times 10^{-15}\text{ sec}.$$

From this result, it appears from the optical data that the principal loss mechanisms at frequencies near $\omega_p(\hbar\omega_p\sim 16$ eV) are from such collisional effects. The data are probably not accurate enough, however, to preclude a contribution of *similar* magnitude due to interband transitions.

The contribution to $\mathrm{Im}\epsilon_{L,T}(k,\omega)$ for $k\neq 0$ from electron correlations as calculated in Secs. II and III can be viewed as a collisional correction to the intraband Landau damping (which vanishes for $k<k_c$), provided that the conduction band is only partly filled so that a parabolic energy-lattice momentum relationship (with an effective mass) is valid. For this to be valid, the important transitions must not take electrons close to

[19] H. Erhenreich, H. R. Phillip, and B. Segal, Phys. Rev. **132**, 1918 (1963).

the top of the conduction band. In the calculation of Sec. III, we find that the important momentum transfers are in the range (in ordinary units) $q<p_F$. Thus, the maximum lattice momentum involved will be of the order $2p_F$. For a half-filled conduction band, this means that electrons are excited up to the edge of the Brillouin zone so the effects of nonparabolicity may be important.

The vanishing of the electron correlational contribution as $k \to 0$ in a free electron gas is a direct result of the parabolic energy-momentum relation for which velocity and momentum are proportional. In this case, since total current and total momentum are also proportional, the momentum conserving electron-electron collisions cannot alter the current which means that the high-frequency conductivity is unaffected by electron-electron collisions. The presence of a wave of finite k destroys the momentum conservation in the collisions and leads to a correction to the conductivity. For a nonparabolic energy-momentum relation, as in a lattice, current and momentum are *not* proportional; and we expect a contribution from electron collisions to $\mathrm{Im}\epsilon_{L,T}(0,\omega)$. In Al, the conduction band is free-electron-like, so we would expect only a weak contribution at $k=0$; then the coefficient of k^2 [in $\mathrm{Im}\epsilon_{L,T}(k,\omega)$] should be well represented by the free electron gas calculation.

The free electron gas calculation assumed that momentum (now interpreted as lattice momentum) is conserved. In a lattice, lattice momentum is conserved modulo a reciprocal-lattice vector. The nonconserving umklapp processes are usually expected to be small.[20] One effect of umklapp processes will be an additional contribution to $\mathrm{Im}\epsilon_{L,T}(0,\omega)$ for $k=0$ from electron-electron collisions. It would perhaps be of some interest to calculate the effects of nonparabolicity, umklapp collisions, and interband transitions all from a consistent nearly free electron pseudopotential model in order to compare their relative importance. The fact that electron-electron correlation effects can (in a lattice) contribute even for $k=0$ may be of particular interest in the optical absorption. In the free electron case, we saw in Sec. III C that the correlation effects were small because of a factor k^2/p_F^2 which in turn is of order $\langle v^2\rangle/c^2\sim 10^{-5}$. The correlation contributions, resulting from the lattice effects, which do not vanish as k^2 may be larger than this free electron estimate. It would be interesting to see if the absorption into the pr-pl final state is sufficiently enhanced by lattice effects to be experimentally observable.

It is important to note that the intraband collisional effects from impurities, phonons, etc., which contribute to $\mathrm{Im}\epsilon_{L,T}(0,\omega)$ will also contribute for $k\neq 0$ and, in particular, will contribute to the coefficient of k^2. Such corrections cannot be measured by optical means, and they have not been calculated. For low temperatures and for very pure material, of course, the effects of phonon and impurity collisions can be reduced. These parameters do not appear to have been optimized in the actual experiments.[4,11,12]

It must be emphasized that $\mathrm{Im}\epsilon_{L,T}(0,\omega)$ can be measured in optical experiments. The values of $\gamma_L(k=0)$ as measured in forward electron-scattering experiments were shown by Swanson[21] to be consistent with the optical data. The interband transitions arise from the $l\neq l'$ terms. It is well known[3] that interband transitions also contribute to $\mathrm{Im}\epsilon_{L,T}(0,\omega)$. Recently, Pethick[22] has calculated $\mathrm{Im}\epsilon_L(0,\omega)$ for Al (and other materials) using a weak pseudopotential model for the lattice. His calculated $\mathrm{Im}\epsilon_L(0,\omega)$ is too small to account for the optical data. This may indicate that the model fails[23] or that the major contribution to $\mathrm{Im}\epsilon_L(0,\omega)$ is due to effects other than interband transitions, such as impurity scattering. If such a pseudopotential calculation is meaningful, it would be straightforward to include finite k and compute the k^2 coefficient of $\mathrm{Im}\epsilon_L(k,\omega)$ caused by interband transitions. Optical data give no information on the k dependence of $\mathrm{Im}\epsilon_L(k,\omega)$, so we must rely on calculations.

The corrections to the interband transitions from additional scattering mechanisms, such as phonons, impurities, and electron-electron collisions, should be relatively small, of the order $(\Delta E_g\tau)^{-1}\ll 1$, where ΔE_g are the minimum gap energies involved and τ is an appropriate collision time.

V. CONCLUSIONS AND SPECULATION

To terms of order k^2, we can write generally using (1.3), (1.5), and (1.6):

$$\gamma_L(k)=(1-k^2/2k_D^2)\omega_p\,\mathrm{Im}\epsilon_L(0,\omega_p) +\tfrac{1}{2}k^2(\partial^2/\partial k^2)[\omega_0\,\mathrm{Im}\epsilon_L(k,\omega_L(k))]_{k=0}. \quad (5.1)$$

The coefficient of $\mathrm{Im}\epsilon_L(0,\omega_p)$ follows from the expansion of the factor

$$Z_L(k)\omega_L(k)=(1-k^2/k_D^2+\cdots) \times\omega_p[1+\tfrac{1}{2}(k^2/k_D^2)+\cdots] \quad (5.2)$$

which occurs in (1.3) to terms of order k^2/k_D^2. Note that there is a *negative* contribution to the terms of order k^2/k_D^2. This contribution can be calculated by using the optical data for $\mathrm{Im}\epsilon_L(0,\omega_p)$ or equivalently by using the experimental value for $k=0$ ($\theta=0$) electron scattering. This negative contribution can then be written:

$$-\tfrac{1}{2}(k^2/k_D^2)\gamma_L(0)=-(k^2/p_F^2)(\gamma_L(0)/\alpha r_s)(9\pi/40), \quad (5.3)$$

[20] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1960).

[21] N. Swanson, J. Opt. Soc. Am. **54**, 1130 (1964).

[22] C. J. Pethick (private communication).

[23] In Pethick's preliminary calculations (Ref. 22) a local, energy-independent pseudopotential, extracted by Ashcroft from Fermi-surface data, was used. The energy dependence of the pseudopotential may be very important since the plasmon energies are very large. The disagreement between theory and experiment may well be due to the inadequacy of using a local, energy-independent potential. We wish to thank Dr. Pethick for communicating the status of his work to us.

where we have used $k_D^2=(20/9\pi)\alpha r_s p_F^2$, which follows from (1.6). For Al, Powell and Swanson measure $\gamma_L(0)\simeq 1.3$ eV and we know $\omega_p\simeq 15$ eV and $r_s=1.8$. Thus, the negative contribution to $\gamma_L(k)/\omega_p$ for Al is

$$-(k^2/p_F^2)6.3\times 10^{-2}\text{ eV}. \tag{5.4}$$

This is small compared with the positive contribution in (3.21) due to electron-electron collisions. The electron-electron correlational contribution to (5.1) is only part of the complete k^2 term as discussed in Sec. IV. Intraband phonon and impurity scattering and interband transitions will contribute to $\mathrm{Im}\epsilon_L(k,\omega_L(k))$ for finite k and will, therefore, also contribute to the coefficient of k^2. The calculation of these contributions is beyond the scope of the present paper, but such calculations may be feasible using something like a pseudopotential model for the lattice effects.

We may summarize our principal results as follows:

(1) An exact calculation has been given of the contribution to the dissipative part of the high-frequency dielectric tensor caused by electron correlations in a free electron gas to terms of lowest order in r_s. The result differs considerably from that given in previous work. The applicability of this result to real metals such as Al is uncertain.

(2) The contribution to the optical absorption from a pr-pl final state is unobservably small. Our result is smaller roughly by a factor of $(v_F/c)^2$ than previous calculations.

(3) The k^2 coefficient of the plasmon width as here calculated is about an order of magnitude lower than that experimentally observed. The agreement between theory and experiment found by Ninham, Powell, and Swanson[4] resulted from an incorrect calculation. The source of the discrepancy between theory and experiment may be scattering (impurity, phonon) or lattice effects (interband transitions) which have yet to be calculated or may be due to the failure of the weak coupling theory.

Probably, the most difficult to surmount of the uncertainties mentioned above is the small r_s expansion. The Fermi liquid theory which is a very powerful and helpful phenomenological dodge for some calculations is of no help here since we must deal with transitions far from the Fermi surface. Detailed, higher-order perturbation calculations are prohibitively difficult and meaningless for large r_s.

The general results for $\mathrm{Im}\epsilon_{L,T}(\mathbf{k},\omega)$ in (2.16)–(2.18) have a very interesting and elegant form in that the integrals $I(\omega)$ and $J(\omega)$ can be expressed entirely in terms of RPA dielectric functions $\epsilon_{L,T}^0(q,\frac{1}{2}\omega\pm u)$. It is tempting to speculate that these expressions have meaning if we replace the RPA $\epsilon_{L,T}^0$ by *exact* dielectric functions $\epsilon_{L,T}$. We then have a set of nonlinear integral equations for $\epsilon_{L,T}$. It seems clear from some preliminary work that replacing $\epsilon_{L,T}^0$ by $\epsilon_{L,T}$ is equivalent to summing a selected set of higher-perturbation terms to all orders in r_s. Whether this set of terms is more important than terms not included in this sum is not yet clear. Since we have a rather good feeling for the qualitative properties of the complete $\epsilon_{L,T}(\mathbf{k},\omega)$ from experiments, sum rule arguments, etc., it may be possible to use this expression (*if* it is valid) to estimate the intermediate coupling effects.

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