Plasmon Damping in Metals*†

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A report is given of a theoretical and experimental investigation into the degree of plasmon damping in metals as a function of momentum transfer. A previous theoretical result by DuBois is corrected and extended by taking into account polarization effects. Measurements are reported of the change in half-width $\Delta E_{1/2}$ of the dispersed $\approx$15-eV aluminum plasmon energy-loss peak, excited by 20-keV electrons, as a function of electron scattering angle θ. The results can be expressed in the form $\Delta E_{1/2}=A+B\theta^2+C\theta^4$, and there is good agreement between the values of B and C obtained from the revised theory and those found experimentally.

1. INTRODUCTION

SINCE Bohm and Pines[1] first proposed that metal valence electrons could be excited collectively, there has been discussion in the literature as to the conditions under which such plasmon excitations can exist as well-defined modes. Pines[2,3] and Ferrell[4] have shown that plasmons may be excited by a fast electron passing through a metal for plasmon wave vectors k less than some cutoff wave vector k_c. The existence of such a cutoff was first experimentally shown by Watanabe[5] and a new measurement of k_c in aluminum has been recently reported by two of us.[6]

It is the purpose of this paper to discuss how the plasmon lifetime or plasmon level width varies as a function of momentum transfer q. Using the notation of DuBois,[7] momenta are expressed in units of the Fermi momentum $q_0=\hbar k_0$, where k_0 is the wave vector of an electron at the Fermi surface (assumed spherical). The level width is determined in the range $q_{\min}\leq q\leq q_c$, where $q_{\min}=\Omega_p/P$, $q_c=k_c/k_0$, and P is the momentum of the primary electron. The plasma frequency Ω_p is equal to $(4\alpha r_s/3\pi)^{1/2}$, where $\alpha=(4/9\pi)^{1/3}$, $r_s=(3/4\pi a_0^3 n)^{1/3}$, n is the free-electron density, and $a_0=\hbar^2/me^2$.

Using a free-electron approximation, Nozières and Pines[3] have estimated $\Delta E_{1/2}/\Delta E$, the ratio of the half-width of the plasmon loss peak $\Delta E_{1/2}$ to the plasmon energy ΔE, to be

$$\Delta E_{1/2}/\Delta E=\Omega_p q^2. \tag{1}$$

DuBois[7] has made a more detailed calculation of the transition probabilities for two modes of plasmon decay and has found the plasmon level width $\Gamma(q)$ to be

$$\Gamma(q)=9.30\Omega_p^2(1+0.785\Omega_p)q^2+2.83\Omega_p q^4+\cdots. \tag{2}$$

Higher order decay processes will increase the level width still further, but in the absence of a known relationship between an observed half-width and the level width predicted by Eq. (2), we will make the assumption that $\Delta E_{1/2}$ is given by $\frac{1}{2}\Gamma(q)$. Thus,

$$\Delta E_{1/2}/\Delta E=4.65\Omega_p(1+0.785\Omega_p)q^2+1.415q^4+\cdots. \tag{3}$$

The coefficient of q^2 in Eq. (3), however, is unreasonably high, as the contribution to $\Delta E_{1/2}/\Delta E$ from the q^2 term alone is about 2.6 in aluminum for $q=q_c$, where q_c has been evaluated for 20-keV electrons using the result of Ferrell.[4]

DuBois' calculations leading to Eq. (2) have been repeated, and a number of errors corrected. We have no criticism of DuBois' method of evaluating $\Gamma_1(q)$ or of his excellent techniques for handling the complicated integrals involved. The corrected result [Eq. (10) below] still, however, predicts an unrealistically large coefficient of q^2. It was realized that this large coefficient was due to the omission of polarization effects in DuBois' final result. A revised expression for $\Gamma(q)$ has now been obtained which contains both a realistic coefficient of q^2 and a corrected coefficient of q^4. These calculations are described in Sec. 2.

Watanabe[5] observed that the dispersed $\approx$15-eV electron energy loss in aluminum faded away into the background as $q\rightarrow q_c$. Meyer[8] has published aluminum loss spectra at several different electron-scattering angles but only Kunz[9] seems to have measured previously the loss peak broadening as a function of q (or scattering angle θ). Kunz's result, however, is not a good fit to either Eq. (1) or to the corrected and extended version of Eq. (2). A new measurement of the broadening of the plasmon loss in Al as a function of θ has been made and is presented in Sec. 3.

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† The theoretical calculations were performed by one of us (B. W. N.) while the experimental measurements were performed at the National Bureau of Standards.
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Finally, the revised theory is compared with the present experiment in Sec. 4. The agreement between theory and experiment is found to be satisfactory.

2. THEORY

DuBois' method and notation will be followed, and this section should be read in conjunction with DuBois' papers,[7,10] in particular Sec. II B of Ref. 7. In order to avoid unnecessarily cumbersome equations, the quantity $\Gamma_1(q)$ is first recalculated ignoring polarization effects as these can be considered at the end of the main calculation.

The transition probability per unit time $\Gamma_1(q)$ for plasmon decay by production of two electron-hole pairs is

$$\Gamma_1(q)=(1/2\pi)\int_{p_1<1}d^3p_1\int_{p_2>1}d^3p_2\int_{p_3<1}d^3p_3\int_{p_4>1}d^3p_4\sum_{\text{spins}}|\langle 2\text{ pairs}|M|1\text{ plasmon}\rangle|^2$$
$$\times\delta(\mathbf{p}_2+\mathbf{p}_4-\mathbf{p}_1-\mathbf{p}_3-\mathbf{q})\delta(\tfrac{1}{2}[p_2^2+p_4^2-p_1^2-p_3^2]-\Omega_0(q)),\quad(4)$$

where

$$\langle 2\text{ pairs}|M|1\text{ plasmon}\rangle=(2\pi)^{-7/2}g_p(q)4\pi\alpha r_s i/(2\Omega_0(q))^{1/2}\left[\frac{(X_2^\dagger X_1)(X_4^\dagger X_3)C(\mathbf{p}_1,\mathbf{p}_2)}{(\mathbf{p}_4-\mathbf{p}_3)^2}-\frac{(X_2^\dagger X_3)(X_4^\dagger X_1)C(\mathbf{p}_3,\mathbf{p}_2)}{(\mathbf{p}_4-\mathbf{p}_1)^2}\right.$$
$$\left.-\frac{(X_4^\dagger X_1)(X_2^\dagger X_3)C(\mathbf{p}_1,\mathbf{p}_4)}{(\mathbf{p}_2-\mathbf{p}_3)^2}+\frac{(X_4^\dagger X_3)(X_2^\dagger X_1)C(\mathbf{p}_3,\mathbf{p}_4)}{(\mathbf{p}_2-\mathbf{p}_1)^2}\right],\quad(5)$$

with X_i a Pauli spinor for the ith particle, $X_i^\dagger$ its Hermitian conjugate, and

$$C(\mathbf{p}_1,\mathbf{p}_2)=S_F(\mathbf{p}_1+\mathbf{q},\omega(p_1)+\Omega_0(q))+S_F(\mathbf{p}_2-\mathbf{q},\omega(p_2)-\Omega_0(q))$$
$$=\frac{\mathbf{q}\cdot(\mathbf{p}_1-\mathbf{p}_2)}{\Omega_p^2}+\frac{q^2}{\Omega_p^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_1)^2-(\mathbf{q}\cdot\mathbf{p}_2)^2}{\Omega_p^3}+O(q^3).\quad(6)$$

We integrate out the momentum-conserving delta functions of Eq. (4) and make the transformations $\mathbf{p}_2=\mathbf{p}_1+\mathbf{k}$, $\mathbf{p}_4=\mathbf{p}_3-\mathbf{k}+\mathbf{q}$, replace $\mathbf{p}_3$ by $-\mathbf{p}_3$ and find

$$\Gamma_1(q)=\frac{27\Omega_p^7}{2^9q^2\pi^2}\int d^3k\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}}d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}-\mathbf{q}|>1}}d^3p_3\delta(\tfrac{1}{2}k^2+\tfrac{1}{2}(\mathbf{k}-\mathbf{q})^2+\mathbf{k}\cdot\mathbf{p}_1+(\mathbf{k}-\mathbf{q})\cdot\mathbf{p}_3-\Omega_0(q))$$
$$\times\sum_{\text{spins}}\left|\frac{(X_2^\dagger X_1)(X_4^\dagger X_3)}{(\mathbf{k}-\mathbf{q})^2}\left(\frac{q^2}{\Omega_p^2}-\frac{\mathbf{q}\cdot\mathbf{k}}{\Omega_p^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_1)^2-(\mathbf{q}\cdot(\mathbf{p}_1+\mathbf{k}))^2}{\Omega_p^3}\right)-\frac{(X_2^\dagger X_3)(X_4^\dagger X_1)}{(\bar{\mathbf{k}}-\mathbf{q})^2}\right.$$
$$\times\left(\frac{q^2}{\Omega_p^2}-\frac{(\mathbf{q}\cdot\bar{\mathbf{k}})}{\Omega_p^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_3)^2-(\mathbf{q}\cdot(\mathbf{p}_1+\mathbf{k}))^2}{\Omega_p^3}\right)-\frac{(X_4^\dagger X_1)(X_2^\dagger X_3)}{\bar{k}^2}\left(\frac{\mathbf{q}\cdot\bar{\mathbf{k}}}{\Omega_p^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_1)^2-(\mathbf{q}\cdot(\mathbf{p}_3+\mathbf{k}-\mathbf{q}))^2}{\Omega_p^3}\right)$$
$$\left.+\frac{(X_2^\dagger X_1)(X_4^\dagger X_3)}{k^2}\left(\mathbf{q}\cdot\mathbf{k}+\frac{(\mathbf{q}\cdot\mathbf{p}_3)^2-(\mathbf{q}\cdot(\mathbf{p}_3+\mathbf{k}-\mathbf{q}))^2}{\Omega_p^3}\right)\right|^2,\quad(7)$$

where $\bar{\mathbf{k}}=(\mathbf{k}+\mathbf{p}_1+\mathbf{p}_3)$. DuBois expands the denominators $(\mathbf{k}-\mathbf{q})^2$, $(\bar{\mathbf{k}}-\mathbf{q})^2$ in powers of q, and retains only the leading terms to obtain his equation (2.16), which contains several obvious misprints. However, when polarization effects are included, the contribution to the integral from the corresponding q-dependent denominators is vastly reduced, and while not completely negligible, can for our purposes be ignored. We therefore put $\mathbf{q}=0$ in the denominators, in the step function which restricts the region of the $\mathbf{p}_3$ integration, and in the delta function, to obtain

$$\Gamma_1(q)=\frac{27\Omega_p^3q^2}{2^9\pi^2}\int d^3k\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}}d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}}d^3p_3\delta(\mathbf{k}\cdot\bar{\mathbf{k}}-\Omega_p)$$
$$\times\sum_{\text{spins}}\left[\left\{\frac{(X_2^\dagger X_1)(X_4^\dagger X_3)}{k^2}-\frac{(X_2^\dagger X_3)(X_4^\dagger X_1)}{(\bar{k})^2}\right\}\left(1-\frac{2(\mathbf{q}\cdot\mathbf{k})(\mathbf{q}\cdot\bar{\mathbf{k}})}{\Omega_pq^2}\right)^2\right].$$

[10] D. F. DuBois, Ann. Phys. (N. Y.) 7, 174 (1959).

If we square the brackets, carry out the spin summations, and make use of the equivalence of $\mathbf{k}$ and $\bar{\mathbf{k}}$ to combine terms, the integral becomes

$$\Gamma_1(q)=\frac{27\Omega_p{}^3q^2}{2^9\pi^2}\int d^3k\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}} d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}} d^3p_3\delta(\mathbf{k}\cdot\bar{\mathbf{k}}-\Omega_p)\left(\frac{32}{k^4}-\frac{4}{k^2\bar{k}^2}\right)\left(1-\frac{2(\mathbf{q}\cdot\mathbf{k})(\mathbf{q}\cdot\bar{\mathbf{k}})}{q^2\Omega_p}\right)^2.$$

The integration over the angles of $\mathbf{k}$ can be carried out at once. The result is

$$\Gamma_1(q)=\frac{9\Omega_p{}^3q^2}{5\times2^5\pi}\int_0^\infty dk\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}} d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}} d^3p_3\delta(\mathbf{k}\cdot\bar{\mathbf{k}}-\Omega_p)\left[\frac{24}{k^2}+\frac{32\bar{k}^2}{\Omega^2}-\frac{4k^2}{\Omega^2}-\frac{3}{\bar{k}^2}\right]. \tag{8}$$

The last term of the integrand in square brackets is of order Ω^2 and can be neglected. To evaluate the remaining integral we write the integrand in square brackets, in view of the delta function, and the symmetry with respect to $\mathbf{p}_1$ and $\mathbf{p}_3$, as

$$\left[\frac{24}{k^2}+\frac{32\bar{k}^2}{\Omega^2}-\frac{4k^2}{\Omega^2}\right]=\left[\frac{24}{k^2}+\frac{64}{\Omega}-\frac{36k^2}{\Omega^2}+\frac{64}{\Omega^2}(p_1{}^2+\mathbf{p}_1\cdot\mathbf{p}_3)\right],$$

so that Eq. (8) can be written

$$\Gamma_1(q)=9\Omega_p{}^3q^2(I_1+I_2)/5\times2^5\pi,$$

with

$$I_1=\int_0^\infty dk\left[\frac{24}{k^2}+\frac{64}{\Omega}-\frac{36k^2}{\Omega^2}\right]\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}} d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}} d^3p_3\delta(\mathbf{k}\cdot\bar{\mathbf{k}}-\Omega_p),$$

$$I_2=\frac{64}{\Omega_p{}^2}\int_0^\infty dk\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}} d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}} d^3p_3\delta(\bar{\mathbf{k}}\cdot\mathbf{k}-\Omega_p)(p_1{}^2+\mathbf{p}_1\cdot\mathbf{p}_3).$$

The integrals over $\mathbf{p}_1$, $\mathbf{p}_3$ can be performed by the method of DuBois. We refer the reader to Eqs. A(1) to A(6) of his paper, from which it follows that, for $k<2$

$$I_1=2\pi\int_0^2 kdk\left[\frac{24}{k^2}+\frac{64}{\Omega}-\frac{36k^2}{\Omega^2}\right]\int_{-\infty}^\infty dt\,\exp[it(k-(\Omega/k))]f_k{}^2(t),$$

where

$$f_k(t)=\int_0^1 d\alpha\int_{k\alpha/2}^1 xdx\,e^{it(x-k\alpha)}=\frac{1}{kt^2}\left(1+\frac{i}{t}\right)[e^{it(1-x)}-e^{it}]-\frac{1}{t^2}e^{-itk/2},$$

and

$$I_2=\frac{64}{\Omega^2}\int_0^2 2\pi kdk\int_{-\infty}^\infty dt\,\exp[it(k-(\Omega/k))]\left[f_k{}^2(t)-f_k(t)h_k(t)-\frac{1}{2}\frac{\partial^2}{\partial t^2}(f_k{}^2(t))\right]$$

$$=\frac{64}{\Omega^2}\int_0^2 2\pi kdk\int_{-\infty}^\infty dt\,\exp[it(k-(\Omega/k))]\left[f_k{}^2(t)\left(1+\frac{k^2}{2}-\Omega+\frac{\Omega^2}{2k^2}\right)-f_k(t)h_k(t)\right],$$

where

$$h_k(t)=\int_0^1 d\alpha\int_{k\alpha/2}^1 x^3dx\,e^{it(x-k\alpha)}=\frac{1}{kt^2}(e^{it(1-k)}-e^{it})\left(1+\frac{3i}{t}-\frac{6}{t^2}-\frac{6i}{t^3}\right)+\left(\frac{6}{t^4}-\frac{k^2}{4t^2}\right)e^{-itk/2}.$$

We do not need the integrals for $k>2$, for small Ω. Collecting together these expressions, we have

$$\Gamma_1(q)=\frac{9\Omega_p{}^3q^2}{80}\int_0^2 kdk\int_{-\infty}^\infty dt\,\exp[it(k-(\Omega/k))]\left\{f_k{}^2(t)\left[\frac{56}{k^2}+\frac{64}{\Omega^2}-\frac{4k^2}{\Omega^2}\right]-\frac{64}{\Omega^2}f_k(t)h_k(t)\right\}. \tag{9}$$

The remaining integrals are very tedious, and using the method of evaluation shown in the Appendix, the following result is finally obtained:

$$\Gamma_1(q)=\tfrac{3}{5}\pi q^2\Omega_p{}^3[10\ln2+2-4.5\Omega+O(\Omega^2)]. \tag{10}$$

Had we expanded the denominators in $(\mathbf{k}-\mathbf{q})^2$, $(\bar{\mathbf{k}}-\mathbf{q})^2$ of the matrix element in Eq. (7), we would have obtained for the coefficient in square brackets the value $[34-22\ln2-4.5\Omega+O(\Omega^2)]$. This result [Eq. (10)] is still too large, and we must now modify our calculation to include polarization effects. DuBois, in writing down the matrix element which describes the decay process, asserts that these effects can be ignored. His justification for this appears to be that the integral which remains converges. To see that this is not so, consider the complete matrix element $\langle 2\text{ pairs}|M|1\text{ plasmon}\rangle$ which includes the modified interaction instead of the bare interaction in the internal Coulomb line of the eight indistinguishable diagrams.[7] The corrected matrix element which corresponds to our Eq. (5) is

$$\langle 2\text{ pairs}|M|1\text{ plasmon}\rangle=\frac{(2\pi)^{-7/2}g_p(q)4\pi\alpha r_s i}{(2\Omega_0(q))^{1/2}}\Bigg[\frac{(X_2{}^\dagger X_1)(X_4{}^\dagger X_3)C(\mathbf{p}_1,\mathbf{p}_2)}{(\mathbf{p}_4-\mathbf{p}_3)^2+(\alpha r_s/\pi^2)Q_{r_s}(|\mathbf{p}_4-\mathbf{p}_3|,\frac{1}{2}(p_4{}^2-p_3{}^2))}$$
$$-\frac{(X_2{}^\dagger X_3)(X_4{}^\dagger X_1)C(\mathbf{p}_3,\mathbf{p}_2)}{(\mathbf{p}_4-\mathbf{p}_1)^2+(\alpha r_s/\pi^2)Q_{r_s}(|\mathbf{p}_4-\mathbf{p}_1|,\frac{1}{2}(p_4{}^2-p_1{}^2))}-\frac{(X_4{}^\dagger X_1)(X_2{}^\dagger X_3)C(\mathbf{p}_1,\mathbf{p}_4)}{(\mathbf{p}_2-\mathbf{p}_3)^2+(\alpha r_s/\pi^2)Q_{r_s}(|\mathbf{p}_2-\mathbf{p}_3|,\frac{1}{2}(p_2{}^2-p_3{}^2))}$$
$$+\frac{(X_4{}^\dagger X_3)(X_2{}^\dagger X_1)C(\mathbf{p}_3,\mathbf{p}_4)}{(\mathbf{p}_2-\mathbf{p}_1)^2+(\alpha r_s/\pi^2)Q_{r_s}(|\mathbf{p}_2-\mathbf{p}_1|,\frac{1}{2}(p_2{}^2-p_1{}^2))}\Bigg].$$

Q_{r_s} includes all types of proper polarization graphs. In the notation of DuBois, we have

$$Q_{r_s}=Q^{(0)}+Q_{r_s}{}^{(1a)}+Q_{r_s}{}^{(1b)}+\cdots.$$

$Q_{r_s}{}^{(1)}$ and higher order propagators should be negligible for small r_s, being of order r_s with respect to $Q^{(0)}$, the pair propagator, so that we retain only the pair propagator whose explicit form is given by DuBois in his first paper,[10] Eqs. (1.43) to (1.46). At the densities which concern us it is by no means certain that higher order propagators are completely negligible, but the effect of retaining these processes should be to reduce our result still further. We proceed as before, to find

$$\Gamma_1(q)_{r_s}=\frac{27\Omega_p{}^7}{2^9q^2\pi^2}\int d^3k\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}}d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}}d^3p_3\delta(\mathbf{k}\cdot\bar{\mathbf{k}}-\Omega)$$
$$\times\sum_{\text{spins}}\Bigg|\left[\frac{(X_2{}^\dagger X_1)(X_4{}^\dagger X_3)}{(\mathbf{k}-\mathbf{q})^2+(\alpha r_s/\pi^2)Q(|\mathbf{k}-\mathbf{q}|,\Omega-\frac{1}{2}(2\mathbf{p}_1\cdot\mathbf{k}+k^2))}\right]\left(\frac{q^2}{\Omega^2}-\frac{\mathbf{q}\cdot\mathbf{k}}{\Omega^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_1)^2-[\mathbf{q}\cdot(\mathbf{p}_1+\mathbf{k})]^2}{\Omega^3}\right)$$
$$-\left[\frac{(X_2{}^\dagger X_3)(X_4{}^\dagger X_1)}{(\bar{\mathbf{k}}-\mathbf{q})^2+(\alpha r_s/\pi^2)Q(|\bar{\mathbf{k}}-\mathbf{q}|,\Omega+\frac{1}{2}(2\mathbf{p}_3\cdot\bar{\mathbf{k}}-\bar{k}^2))}\right]\left(\frac{q^2}{\Omega^2}-\frac{\mathbf{q}\cdot\bar{\mathbf{k}}}{\Omega^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_3)^2-(\mathbf{q}\cdot(\mathbf{p}_1+\mathbf{k}))^2}{\Omega^3}\right)$$
$$-\left[\frac{(X_4{}^\dagger X_1)(X_2{}^\dagger X_3)}{\bar{k}^2+(\alpha r_s/\pi^2)Q(\bar{k},\frac{1}{2}(\bar{k}^2-2\mathbf{p}_3\cdot\bar{\mathbf{k}}))}\right]\left(\frac{\mathbf{q}\cdot\bar{\mathbf{k}}}{\Omega^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_1)^2-(\mathbf{q}\cdot(\mathbf{p}_3+\mathbf{k}-\mathbf{q}))^2}{\Omega^3}\right)$$
$$+\left[\frac{(X_2{}^\dagger X_1)(X_4{}^\dagger X_3)}{k^2+(\alpha r_s/\pi^2)Q(k,\frac{1}{2}(k^2+2\mathbf{k}\cdot\mathbf{p}_1))}\right]\left(\frac{\mathbf{q}\cdot\mathbf{k}}{\Omega^2}+\frac{(\mathbf{q}\cdot\mathbf{p}_3)^2-(\mathbf{q}\cdot(\mathbf{p}_3+\mathbf{k}-\mathbf{q}))^2}{\Omega^3}\right)\Bigg|^2.$$

The denominators of the terms in square brackets are of the form $k^2+(\alpha r_sQ(k,u))/\pi^2$ and the principal contribution to the integral, for small Ω, comes from the region $\Omega/2<k<\Omega$. Since in our dimensionless variables we have $\Omega_p=(4\alpha r_s/3\pi)^{1/2}$, we see that polarization effects can certainly not be ignored. The two terms, k^2, $\alpha r_sQ(k,u)/\pi^2$, should be at least of equal magnitude in the region of principal importance. To simplify this expression, we make use of the restrictions imposed by the step functions and the delta function, which require that for small values of k, the arguments of the various $Q(k,u)$ are $u=\frac{1}{2}\Omega+O(\Omega^2)$. We can then expand the several $Q(k,u)$ about $u=\frac{1}{2}\Omega$, and retain only the leading term. We obtain

$$\frac{1}{(\mathbf{k}-\mathbf{q})^2+\alpha r_sQ(|\bar{\mathbf{k}}-\mathbf{q}|,\frac{1}{2}\Omega)/\pi^2}=\frac{1}{k^2+\alpha r_sQ(k,\frac{1}{2}\Omega)/\pi^2}+\frac{k^2-(\mathbf{k}-\mathbf{q})^2+\alpha r_s[Q(k,\frac{1}{2}\Omega)-Q(|\mathbf{k}-\mathbf{q}|,\frac{1}{2}\Omega)]/\pi^2}{[k^2+\alpha r_sQ(k,\frac{1}{2}\Omega)/\pi^2]^2}+O(q^2),$$

and a similar expression for the denominator in $(\bar{\mathbf{k}}-\mathbf{q})^2$.

The term of order q in this expansion should be retained, but a detailed analysis shows that if we keep this term our result is only slightly modified, so that in view of our approximations, it can be ignored. Proceeding as before, following Eq. (7), we then find

$$\Gamma_1(q)_{r_s}=\frac{27\Omega_p{}^3q^2}{2^9\pi^2}\int d^3k\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}}d^3p_1\int_{\substack{p_3<1\\|\mathbf{p}_3+\mathbf{k}|>1}}d^3p_3\delta(\mathbf{k}\cdot\bar{\mathbf{k}}-\Omega_p)\left\{\frac{32}{A^2+B^2}-\frac{4(A\bar{A}+B\bar{B})}{(A^2+B^2)(\bar{A}^2+\bar{B}^2)}\right\}\left(1-\frac{2}{q^2\Omega_p}(\mathbf{q}\cdot\mathbf{k})(\mathbf{q}\cdot\bar{\mathbf{k}})\right)^2,$$

where

$$A=A(k)=\mathrm{Re}[k^2+(\alpha r_sQ(k,\tfrac{1}{2}\Omega)/\pi^2)],$$
$$B=B(k)=\mathrm{Im}[\alpha r_sQ(k,\tfrac{1}{2}\Omega)/\pi^2],\quad \bar{A}=A(\bar{k}),\quad \bar{B}=B(\bar{k}).$$

We are interested in $k=O(\Omega)$, and in this region our step functions and delta function imply that $\bar{k}\approx 2$, so that we can put $\bar{A}=\bar{k}^2$, and ignore $\bar{B}$ entirely. Then, repeating our previous analysis, we have

$$\Gamma_1(q)_{r_s}=\frac{9\Omega_p{}^3q^2}{80}\int_0^2 kdk\int_{-\infty}^{\infty}dt\,\exp[it(k-(\Omega/k))]\left[\frac{k^4}{(A^2+B^2)}\right]\left\{f_k{}^2(t)\left[\frac{56}{k^2}+\frac{64}{\Omega^2}-\frac{4A}{\Omega^2}\right]-\frac{64}{\Omega^2}f_k(t)h_k(t)\right\}.$$

We have computed the ratio $k^4/(A^2+B^2)$ for $\frac{1}{2}\Omega<k<\Omega$, using

$$\mathrm{Re}Q(q,u)=2\pi\left[1-\frac{1}{2q}\left(1-\left(\frac{u}{q}-\frac{q}{2}\right)^2\right)\ln\left|\left(\frac{u}{q}-\frac{q}{2}+1\right)\Big/\left(\frac{u}{q}-\frac{q}{2}-1\right)\right|\right.$$
$$\left.+\frac{1}{2q}\left(1-\left(\frac{u}{q}+\frac{q}{2}\right)^2\right)\ln\left|\left(\frac{u}{q}+\frac{q}{2}+1\right)\Big/\left(\frac{u}{q}+\frac{q}{2}-1\right)\right|\right],$$

and

$$\mathrm{Im}Q(q,u)=\pi\int_{\substack{p<1\\|\mathbf{p}+\mathbf{q}|>1}}d^3p\;\delta(\tfrac{1}{2}q^2+\mathbf{q}\cdot\mathbf{p}-u)+\int_{\substack{p<1\\|\mathbf{p}+\mathbf{q}|>1}}d^3p\;\delta(\tfrac{1}{2}q^2+\mathbf{q}\cdot\mathbf{p}+u)$$
$$=2\pi^2\left\{\frac{u}{q}+\frac{1}{q}\eta\left(\frac{q}{2}+\frac{u}{q}-1\right)\right.$$
$$\times\left[-\left(\frac{q}{2}+\frac{u}{q}-1\right)-\frac{1}{2}\left(\frac{q}{2}+\frac{u}{q}-1\right)^2\right]-\frac{1}{q}\eta\left(\frac{u}{q}-\frac{q}{2}-1\right)\left[-\left(\frac{u}{q}-\frac{q}{2}-1\right)-\frac{1}{2}\left(\frac{u}{q}-\frac{q}{2}-1\right)^2\right]$$
$$\left.+\frac{1}{q}\eta\left(\frac{q}{2}-\frac{u}{q}-1\right)\left[-\left(\frac{q}{2}-\frac{u}{q}-1\right)-\frac{1}{2}\left(\frac{q}{2}-\frac{u}{q}-1\right)^2\right]\right\},\quad u>0,$$

and for $\Omega_p=\frac{2}{3}$, which is very nearly the value for aluminum. It turns out to be very nearly constant: $k^4/(A^2+B^2)\approx 1/36$, and we can estimate $\Gamma_1(q)_{r_s}$ at once by simply dividing our Eq. (10) by this factor. For different values of Ω_p the modifying factor will of course vary, but for aluminum we have

$$\Gamma_1(q)_{r_s}=(\pi/60)q^2\Omega_p{}^3\{[10\ln 2+2]-O(\Omega)\}.\tag{11}$$

In view of our approximations, this result must be regarded as fairly crude, and since we have neglected higher order polarization processes, Eq. (11) represents an upper bound to the true plasmon level width. DuBois also considers plasmon decay via excitation of a pair plus another plasmon. Unfortunately, his result for this contribution to the level width is also incorrect, the error being due to incorrect evaluation of the integral and to his dispersion relation, Eq. (1.10), which is incorrect and should read

$$\Omega_0(q)=\Omega_p+3q^2(1-\tfrac{1}{4}\Omega^2)/10\Omega_p+\cdots.\tag{12}$$

For this decay process we have, from DuBois' Eq. (2.20),

$$\Gamma_2(q)=\frac{g_p{}^2(q)}{(2\pi)^5\Omega_0(q)}\int d^3k\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{q}-\mathbf{k}|>1}}d^3p_1\frac{g_p{}^2(k)}{2\Omega_0(k)}C^2(\mathbf{p}_1,\mathbf{p}_1+\mathbf{q}-\mathbf{k})\delta[(\mathbf{q}-\mathbf{k})\cdot\mathbf{p}_1+\tfrac{1}{2}(\mathbf{q}-\mathbf{k})^2+\Omega_0(k)-\Omega_0(q)].\tag{13}$$

If we use the expansion, Eq. (6), and the correct dispersion relation, Eq. (12), and if we change variables to

$\mathbf{k}'=\mathbf{q}-\mathbf{k}$ and drop the prime, Eq. (13) becomes

$$\Gamma_2(q)=\frac{9\Omega^2}{64q^2\pi}\int\frac{d^3k}{(\mathbf{q}-\mathbf{k})^2}\int_{\substack{p_1<1\\|\mathbf{p}_1+\mathbf{k}|>1}}d^3p_1$$

$$\times\left[\left(q^2-\mathbf{q}\cdot\mathbf{k}-\frac{(\mathbf{q}\cdot\mathbf{k})^2}{\Omega}\right)^2-\frac{4(\mathbf{q}\cdot\mathbf{p}_1)(\mathbf{q}\cdot\mathbf{k})}{\Omega}\left(q^2-\mathbf{q}\cdot\mathbf{k}-\frac{(\mathbf{q}\cdot\mathbf{k})^2}{\Omega}\right)+\frac{4(\mathbf{q}\cdot\mathbf{p}_1)^2(\mathbf{q}\cdot\mathbf{k})^2}{\Omega^2}\right]\delta\left(\mathbf{k}\cdot\mathbf{p}_1+\tfrac{1}{2}k^2+\frac{\beta}{\Omega}(-2\mathbf{q}\cdot\mathbf{k}+k^2)\right),$$

where $\beta=(3/10)(1-\frac{1}{4}\Omega^2)$.

The step function, $|\mathbf{p}_1+\mathbf{k}|>1$, and the delta function together imply that $p_1^2>[1-(2\beta(2\mathbf{q}\cdot\mathbf{k}-k^2)/\Omega)]$. The integral over $\mathbf{p}_1$ will therefore give no contribution unless $2\mathbf{q}\cdot\mathbf{k}-k^2>0$, and we have, after carrying out one of the angular integrations,

$$\Gamma_2(q)=\frac{9\Omega^2\pi}{16q^2}\int_0^{2q}k^2dk\int_{k/2q}^1\frac{dx_2}{(q^2-2\mathbf{q}\cdot\mathbf{k}+k^2)}\int_y^1 p_1^2dp_1\eta(y^2)\int_{-1}^{+1}dx_1\delta\left(kp_1x_1+\frac{k^2}{2}+\frac{\beta}{\Omega}(-2\mathbf{q}\cdot\mathbf{k}+k^2)\right)$$

$$\times\left\{\left[q^2-\mathbf{q}\cdot\mathbf{k}-\frac{(\mathbf{q}\cdot\mathbf{k})^2}{\Omega}\right]^2-\frac{4\mathbf{q}\cdot\mathbf{k}}{\Omega}\left(q^2-\mathbf{q}\cdot\mathbf{k}-\frac{(\mathbf{q}\cdot\mathbf{k})^2}{\Omega}\right)qp_1x_1x_2+4q^2p_1^2\frac{(\mathbf{q}\cdot\mathbf{k})^2}{\Omega^2}\left(x_1^2x_2^2+\frac{(1-x_1^2)(1-x_2^2)}{2}\right)\right\},\quad(14)$$

where $x_2=\hat{q}\cdot\hat{k}$, $x_1=\hat{p}_1\cdot\hat{k}$, and $y=[1-(2\beta(2\mathbf{q}\cdot\mathbf{k}-k^2)/\Omega)]^{1/2}$. We put $k=2qz$, and drop terms in q^5 and q^6, and Eq. (14) becomes

$$\Gamma_2(q)=\frac{9\pi\Omega^2}{2q}\int_0^1 z^2dz\int_z^1\frac{dx_2}{(1-4zx_2+4z^2)}\int_y^1\eta[1-(\beta q^2(zx_2-z^2)/\Omega)]p_1^2dp_1\times\int_{-1}^{+1}dx_1$$

$$\times\delta(2qzp_1x_1+2q^2z^2+(4\beta q^2(-zx_2+z^2)/\Omega))\{q^4(1-2zx_2)^2\}.\quad(15)$$

For small q, the step function and delta function are always satisfied, the remaining quadratures are trivial, and Eq. (15) reduces to

$$\Gamma_2(q)=13\pi\Omega q^4(1-\tfrac{1}{4}\Omega^2)/400.\quad(16)$$

This does not yet complete the calculation, as we have so far neglected the q^4 dependence of $\Gamma_1(q)_{r_s}$. The matrix element [Eq. (5)], which prescribes $\Gamma_1(q)$ involves the effective coupling of the plasmon, $g_p(q)$, which has been expanded incorrectly by DuBois [Eq. (1.15)] and should read

$$\frac{g_p^2(q)}{(2\pi)^3}=\frac{3\Omega_p^4}{8\pi q^2}\left\{1+\frac{9q^2}{5\Omega_p^2}+\cdots\right\}.\quad(17)$$

If we retain the second term in this expansion throughout, we see from Eq. (11) that the additional term in q^4 is

$$\Gamma_2(q)_{r_s}=\frac{9\pi q^4\Omega}{300}\{(10\ln 2+2)+O(\Omega)\}.\quad(18)$$

Combining Eqs. (11), (16), and (18) we have for the plasmon level width $\Gamma(q)$

$$\Gamma(q)=(\pi q^2\Omega_p^3/60)[(10\ln 2+2)-O(\Omega)]$$

$$+q^4\Omega\pi\left[\frac{9}{300}(10\ln 2+2)+\frac{13}{400}\left(1-\frac{\Omega^2}{4}\right)\right]+\cdots.\quad(19)$$

Assuming as before that $\Delta E_{1/2}=\frac{1}{2}\Gamma(q)$,

$$\frac{\Delta E_{1/2}}{\Delta E}=\frac{\pi\Omega_p^2(10\ln 2+2)q^2}{120}$$

$$+\frac{\pi}{100}\left[15\ln 2+3+\frac{13}{8}\left(1-\frac{\Omega^2}{4}\right)\right]q^4+\cdots.\quad(20)$$

DuBois[7] used his expression for $\Gamma(q)$ to estimate a value for q_c. In view of the differences between Eqs. (2) and (19), his evaluation and discussion of q_c are now not significant.

3. EXPERIMENT

Measurements have been made of the characteristic loss spectra of 20-keV electrons passing through a 1000-Å film of aluminum as a function of electron-scattering angle.[6] The energy resolution of the apparatus was ≈ 1 eV and the angular resolution ≈ 1 mrad. For Al and 20-keV electrons, $\theta_c=q_c/P\approx 20$ mrad.

Spectra were measured at angular intervals of about 1.9 mrad for scattering angles between 0 and 23 mrad; some of the spectra are shown in Fig. 1. The full-width at half-maximum intensity of the dispersed $\approx$15-eV loss was measured at each angle and plotted as a function of θ^2, as shown in Fig. 2. At the larger scattering angles, it was necessary to assume a level for the continuum of single-electron excitations, a line shape for the undispersed peak at ≈ 15 eV, and that the dispersed loss peak was symmetrical.[6] While it is believed that these assumptions were reasonable, the spread of the experimental points in Fig. 2 indicates the degree of uncertainty in the measurements.

4. COMPARISON OF THEORY AND EXPERIMENT

The theoretical expressions and the experimental results for the broadening of the Al plasmon loss peak can be expressed as a function of scattering angle θ by

$$\Delta E_{1/2} = A + B\theta^2 + C\theta^4, \tag{21}$$

where the constants B and C are appropriate to the primary electron energy of 20 keV. Values of the constant A have been obtained recently for Al and two other elements and have been compared with the relevant theory.[11]

The data points shown in Fig. 2 have been fitted to Eq. (21) by the method of least squares. Similarly, the data points plotted by Kunz[9] have been fitted by us

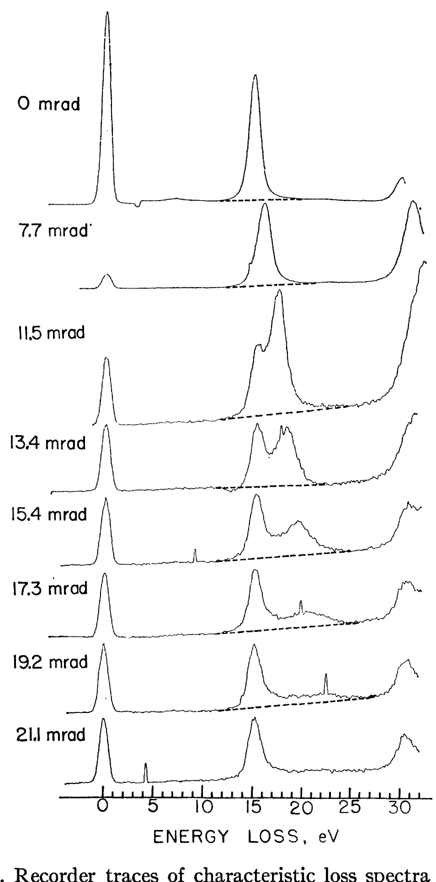

FIG. 1. Recorder traces of characteristic loss spectra obtained with a 1000-Å aluminum specimen for the various electron scattering angles indicated. The occasional spikes are noise pulses due to electrical breakdown. The dashed lines indicate the continuum levels assumed in obtaining the half-width of the dispersed peak.

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

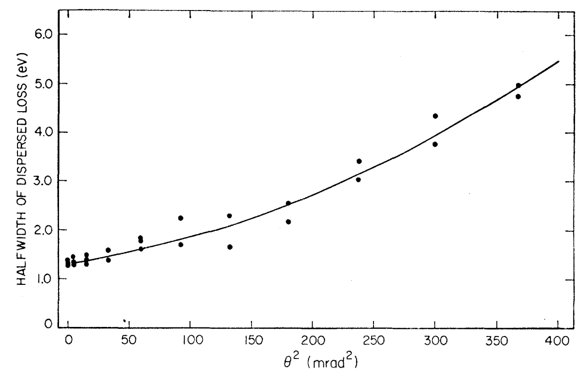

FIG. 2. Plot of measured full-width at half-maximum intensity of the dispersed plasmon loss in aluminum as a function of the square of the scattering angle θ. The solid line is the computed least-squares quadratic fit to the experimental points.

and the two sets of experimental values of B and C are shown in Table I. Two qualifications need to be mentioned in using these experimental values of B and C. Firstly, no correction has been made to either set of experimental data to account for the energy width of the primary electron beam and the finite energy resolution of the analyzer appropriate to each experiment. An unfolding operation has recently been performed on an Al spectrum obtained at zero scattering angle from which it was found that the unfolded half-width was 1.05 eV compared to the measured width of 1.35 ± 0.04 eV.[11] It might then be expected that the value of B found experimentally in this work could be too low by up to 20% and that the value of C could be smaller than indicated. Secondly, it is difficult to estimate any error at the larger scattering angles associated with the unknown shape and level of the continuum and with the unknown plasmon line shape. From recent measurements of the differential cross section for the Al plasmon loss,[12] it would seem that any errors of the latter types are unlikely to be appreciable.

Theoretical estimates of the values for B and C using Eqs. (1), (3), and (20) are also shown in Table I. It is seen that there is satisfactory agreement between the theoretical and experimental results of the present

TABLE I. Theoretical and experimental values of the constants B and C in Eq. (20). The errors quoted for the experimental values are the standard errors derived from the least-squares fits of the raw experimental data; no contributions have been included for the sources of systematic error discussed in the text.

	B	C
Theory:		
Nozières and Pines [Eq. (1)]	1.87×10^4	
DuBois [Eq. (3)]	1.33×10^5	6.83×10^7
This work [Eq. (19)]	2.96×10^3	2.25×10^7
Experiment:		
Kunz (Ref. 9)	$-(9.5\pm5.2)\times10^3$	$(1.5\pm0.3)\times10^8$
This work	$(3.5\pm1.1)\times10^3$	$(1.7\pm0.3)\times10^7$

[12] Reference 6, Fig. 3.

work. Values of B obtained using Eqs. (1) and (3) are much higher than those found here experimentally, while the value of B found from Kunz's data is negative; this latter nonphysical result might possibly be due to random errors in Kunz's data points or to a systematic error in his selection of a continuum level. It would be reasonable to assume that the discrepancies between the theoretical and experimental values of B and C found in the present work are due to the experimental errors discussed above, to the approximations used in Sec. 2, and to the assumption $\Delta E_{1/2}=\frac{1}{2}\Gamma(q)$, and to departures from the free-electron theory caused by the lattice.

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APPENDIX

We first carry out the t integrations in Eq. (9) using the identity

$$\int_{-\infty}^{\infty} dt\, e^{-ita(k)}t^{-n}=\frac{\pi(-1)^n}{(n-1)!}[a(k)]^{n-1}\operatorname{sgn} a(k)\,,$$

and find after some algebra that

$$\int_{-\infty}^{\infty} dt\, f_k^2(t)\exp\{it[k-(\Omega/k)]\}=(2\pi/k^2)\{-g(2-k)+2g(2)-g(k+2)+2k[j(1-\tfrac{1}{2}k)-j(\tfrac{1}{2}k+1)]\}\,, \tag{A1}$$

where

$$g(z)=\eta\left(-\frac{\Omega}{k}+z\right)\left[\frac{1}{3!}\left(\frac{\Omega}{k}-z\right)^3+\frac{2}{4!}\left(\frac{\Omega}{k}-z\right)^4+\frac{1}{5!}\left(\frac{\Omega}{k}-z\right)^5\right],$$

$$j(z)=\eta\left(-\frac{\Omega}{k}+z\right)\left[\frac{1}{3!}\left(\frac{\Omega}{k}-z\right)^3+\frac{1}{4!}\left(\frac{\Omega}{k}-z\right)^4\right] \tag{A2}$$

and

$$\int_{-\infty}^{\infty} dt\, f_k(t)h_k(t)=(2\pi/k^2)\{-l(2-k)+2l(2)-l(2+k)+k[s(1-\tfrac{1}{2}k)-s(1+\tfrac{1}{2}k)]\}\,,$$

where

$$l(z)=\eta\left(-\frac{\Omega}{k}+z\right)\left[\frac{1}{3!}\left(\frac{\Omega}{k}-z\right)^3+\frac{4}{4!}\left(\frac{\Omega}{k}-z\right)^4+\frac{9}{5!}\left(\frac{\Omega}{k}-z\right)^5+\frac{12}{6!}\left(\frac{\Omega}{k}-z\right)^6+\frac{6}{7!}\left(\frac{\Omega}{k}-z\right)^7\right],$$

$$s(z)=\eta\left(-\frac{\Omega}{k}+z\right)\left[\left(1+\frac{k^2}{4}\right)\frac{1}{3!}\left(\frac{\Omega}{k}-z\right)^3+\left(3+\frac{k^2}{4}\right)\left(\frac{\Omega}{k}-z\right)^4+\frac{12}{5!}\left(\frac{\Omega}{k}-z\right)^5+\frac{12}{6!}\left(\frac{\Omega}{k}-z\right)^6\right].$$

The remaining integrals over k are elementary, but so many are involved that it is very easy to make errors. We therefore appeal to the method of Mellin transforms. We shall illustrate the technique for one of the k integrals of Eq. (9). Consider

$$L(\Omega)=\int_0^2\left(\frac{dk}{k}\right)\int_{-\infty}^{\infty} dt\,\exp[it(k-(\Omega/k))]f_k^2(t)\,.$$

Using Eqs. (A1), (A2), and the identity

$$\int_0^{\infty}\Omega^{p-1}\eta\left(z-\frac{\Omega}{k}\right)\left(\frac{\Omega}{k}-z\right)^n d\Omega=\frac{k^p z^{p+n}(-1)^n(p-1)!\,n!}{(p+n)!}\,,\quad \operatorname{Re}p>0\,,$$

the Mellin transform of $L(\Omega)$ is

$$M(p)=\int_0^{\infty}\Omega^{p-1}L(\Omega)d\Omega=2\pi(p-1)!\int_0^1 dx$$
$$\times\{x^{p-3}2^{2p+1}[f(p+3)-4f(p+4)+4f(p+5)]-x^{p-2}2^p[f'(p+3)-f'(p+4)]\}\,,\quad \operatorname{Re}p>0\,, \tag{A3}$$

where

$$f(p+m)=\frac{(1-x)^{p+m}-2+(1+x)^{p+m}}{(p+m)!};\quad f'(p+m)=\frac{(1-x)^{p+m}-(1+x)^{p+m}}{(p+m)!}.$$

Hence, by the Mellin Inversion formula

$$L(\Omega)=(1/2\pi i)\int_{C-i\infty}^{C+i\infty}M(p)\Omega^{-p}dp;\quad C=\operatorname{Re}p>0.$$

The integrals over x in Eq. (A3) have a pole at $p=0$. Thus, we have

$$\int_0^1 x^{p-3}f(p+m)dx=\int_0^1 x^{p-3}[(1-x)^{p+m}-2+(1+x)^{p+m}]dx/(p+m)!$$

$$=(1/p(p+m-2)!)+\int_0^1[x^{p-3}f(p+m)-x^{p-1}(p+m-2)!]dx, \tag{A4}$$

where we have expanded the integrand about $x=0$ to exhibit the pole explicitly. The second term of Eq. (A4) is regular at $p=0$. Similarly

$$\int_0^1 x^{p-2}f'(p+m)dx=-\frac{2}{(p+m-1)!}+\int_0^1\left[x^{p-2}f'(p+m)+\frac{2x^{p-1}}{(p+m-1)!}\right]dx,$$

so that we can write

$$L(\Omega)=\frac{1}{i}\int_{C-i\infty}^{C+i\infty}\frac{(p-1)!}{p}\left\{2^{2p+1}\left[\frac{1}{(p+1)!}-\frac{4}{(p+2)!}+\frac{4}{(p+3)!}\right]+2^{p+1}\left[\frac{1}{(p+2)!}-\frac{1}{(p+3)!}\right]\right\}\Omega^{-p}dp$$

$$+\frac{1}{i}\int_{C-i\infty}^{C+i\infty}(p-1)!\int_0^1 dx\left\{2^{2p+1}x^{p-3}\left(\left[f(p+3)-\frac{x^2}{(p+1)!}\right]-4\left[f(p+4)-\frac{x^2}{(p+2)!}\right]+4\left[f(p+5)-\frac{x^2}{(p+3)!}\right]\right)\right.$$

$$\left.-2^px^{p-2}\left(\left[f'(p+3)+\frac{2x}{(p+2)!}\right]-\left[f'(p+4)+\frac{2x}{(p+3)!}\right]\right)\right\}\Omega^{-p}dp.$$

The first integral has a double pole at $p=0$, and single poles at $p=-1,-2$, while the second has single poles only which concern us. Evaluating the residues at these poles, we have

$$L(\Omega)=\tfrac{4}{3}\pi(1-\ln 2)+O(\Omega^3).$$

Similarly, we find the following results:

$$\int_0^2 kdk\int_{-\infty}^{\infty}dt\,\exp[it(k-(\Omega/k))]f_k^2(t)=\tfrac{1}{2}\pi\Omega^2-\tfrac{1}{6}\pi\Omega^3+O(\Omega^4),$$

$$\int_0^2 k^3dk\int_{-\infty}^{\infty}dt\,\exp[it(k-(\Omega/k))]f_k^2(t)=\tfrac{2}{3}\pi\Omega^3,$$

$$\int_0^2 kdk\int_{-\infty}^{\infty}dt\,\exp[it(k-(\Omega/k))]f_k(t)h_k(t)=2\pi[\Omega^2(\tfrac{3}{4}-\ln 2)+(\Omega^3/12)].$$

The above results may be collected and substituted into Eq. (9) to obtain Eq. (10).