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Theoretical Analysis of Plasmon Dispersion in Simple Metals

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Abstract

The dielectric response of an electron gas has been investigated with a view to understanding the small momentum behaviour of the plasmon peak in inelastic electron energy loss spectra. It is shown that the lifetime of the plasmon and the width of the incident beam both contribute to the apparent flattening of the plasmon dispersion which has been seen to occur in some metals (magnesium, aluminium, indium) for plasmon wavevectors less than $\sim 0{\cdot}5\,\text{Å}^{-1}$. Expressions, beyond the random phase approximation, are derived for the dispersion coefficients up to fourth order in the plasmon wavevector. Good agreement with experiment is obtained.

1. Introduction

Several authors have recently drawn attention to the rather flat dispersion of the small momentum bulk plasmon oscillations in some simple metals. Krane (1978) describes the frequency versus momentum (ω versus q) relation for the plasmons in aluminium and indium as having two branches:

$$\omega = \omega_p + \alpha(\hbar^2/m)q^2, \qquad q < 0{\cdot}5\,\text{Å}^{-1}; \tag{1a}$$

$$= \omega_p' + \alpha'(\hbar^2/m)q^2, \qquad q > 0{\cdot}5\,\text{Å}^{-1} \tag{1b}$$

(note that $1\,\text{Å}^{-1} \equiv 10^{10}\,\text{m}^{-1}$). Here ω_p, $\hbar$ and m are the plasmon frequency, Planck's constant divided by 2π and the mass of an electron while α, ω_p' and α' are other constants. Sturm (1978*b*) also drew attention to the small q region, demonstrating that no satisfactory agreement exists between experiment and theory. This behaviour is not explained within the random phase approximation (RPA) even with the inclusion of band structure and core polarization effects (Bross 1978; Sturm 1978*a*).

The purpose of this paper is to re-examine the plasmon dispersion relation and show that we do indeed have a reasonable description of the plasmon dispersion in simple metals. The dispersion relation is significant from several viewpoints: it describes the propagation and properties of plasmons; it gives a direct experimental check on exchange and correlation effects; through the dielectric function it is indirectly related to various transport properties; and the parameters in the dispersion relation are used as an input into some simple but illuminative models (e.g. Mehrotra and Mahanty 1979).

The dispersion relation, in the notation of the hydrodynamic model, may be written as

$$\omega^2 = \omega_p^2 + \bar{\beta}_0^2 q^2 + \bar{\beta}_1^2 q^4 + O(q^6) \tag{2a}$$

or, in terms of dimensionless parameters b, as

$$\omega^2 = \omega_p^2 + b_0(e^4/\hbar^2)q^2 + b_1(\hbar^2/m^2)q^4 + O(q^6), \tag{2b}$$

where $-e$ is the electronic charge. The coefficient b_0 has been examined many times (e.g. Ferrell 1957; Singwi *et al.* 1968) but this is the first time an expression going beyond the RPA has been given for b_1.

The outline of the present paper is as follows. In Section 2 a model dielectric function based on a proposal by Hubbard (1958) is considered, and this leads to a dispersion relation of the form given by equations (2). From a knowledge of the exchange and correlation functional (Kohn and Sham 1965) the coefficients b_0 and b_1 are then evaluated. In Section 3 the model is extended to allow the inclusion of lifetime effects. It is shown that the combination of the energy width of the incident beam and the plasmon lifetime leads to the type of behaviour depicted by equations (1). Thus it is not possible to obtain reliable information about the coefficients b from very small q experimental dispersion relations without a careful theoretical analysis. Hence the recent reassessed values of this parameter by Sturm (1978*b*) are in error. Atomic units of Rydbergs (1 Ry $\equiv$ 13·605 eV) and Bohr radii ($a_0 = 5{\cdot}292 \times 10^{-11}$ m) are used throughout.

2. Basic Model

Quite generally, the dielectric function $\varepsilon(\boldsymbol{q}, \omega)$ for an electron gas may be written as

$$\varepsilon(\boldsymbol{q}, \omega) = 1 + \frac{\varepsilon_{\mathrm{RPA}}(\boldsymbol{q}, \omega) - 1}{1 - G(\boldsymbol{q}, \omega)\{\varepsilon_{\mathrm{RPA}}(\boldsymbol{q}, \omega) - 1\}}, \tag{3}$$

where $\varepsilon_{\mathrm{RPA}}(\boldsymbol{q}, \omega)$ is the RPA dielectric function and $G(\boldsymbol{q}, \omega)$, which represents contributions from the non-bubble diagrams, gives the exchange and correlation corrections. Now the RPA is known to be a good approximation at high electron densities ($r_s < 1$) and also to be 'reasonable' for small q values. However, it is a time-dependent Hartree approach and therefore does not include an exchange hole which must certainly be important for large q. Hubbard's (1958) proposal for overcoming this difficulty was to write

$$G(\boldsymbol{q}, \omega) \approx \tfrac{1}{2}q^2/(q^2 + K_F^2), \tag{4}$$

where $\hbar K_F$ is the Fermi momentum. This is equivalent to assuming that the effective particle–hole interaction is static and of a particular form. In this section it will be assumed likewise that

$$G(\boldsymbol{q}, \omega) \approx G(\boldsymbol{q}, 0). \tag{5}$$

This is the form taken for the dielectric function by Vashistra and Singwi (1972), and the reader is referred to their paper for more details and references associated with this approximation. Note that this is a mean field approximation in the sense that it gives the correct result for $\varepsilon(\boldsymbol{q}, \omega)$ when $\omega = 0$ and $\omega \to \infty$. In Section 3 below this approximation will be extended.

Now (Pines 1964), we may write

$$G(\boldsymbol{q},0) = \gamma_1(q/K_F)^2 + \gamma_2(q/K_F)^4 + O(q^6) \tag{6}$$

and determine the dimensionless coefficients γ_1 and γ_2 by considering the exact static dielectric function. We can then find the plasmon dispersion relation from the zeros of the dielectric function.

In the static limit we may use the density functional theorem (Hohenberg and Kohn 1964). Thus a perturbing charge density $-en_{ext}(\boldsymbol{r})$ disturbs the electron number density $n(\boldsymbol{r}) = n_0 + n_1(\boldsymbol{r})$ in the electron gas in such a way that the variation is given by the density functional 'Poisson' equation

$$\nabla^2\left(\frac{\delta T_s[n(\boldsymbol{r}')]}{\delta n(\boldsymbol{r})} + \frac{\delta E_{xc}[n(\boldsymbol{r}')]}{\delta n(\boldsymbol{r})}\right) = 4\pi e^2\{n_1(\boldsymbol{r}) + n_{ext}(\boldsymbol{r})\}, \tag{7}$$

where $T_s[n(\boldsymbol{r}')]$ is the noninteracting-electron kinetic energy functional, $E_{xc}[n(\boldsymbol{r}')]$ is the exchange and correlation functional, and n_0 is the unperturbed number density. It is convenient to introduce parameters β_i^2 by the linear gradient expansion

$$\frac{n(\boldsymbol{r})}{m}\nabla\left(\frac{\delta T_s[n(\boldsymbol{r}')]}{\delta n(\boldsymbol{r})} + \frac{\delta E_{xc}[n(\boldsymbol{r}')]}{\delta n(\boldsymbol{r})}\right)$$
$$= \beta_0^2\nabla n_1(\boldsymbol{r}) - \beta_1^2\nabla^3 n_1(\boldsymbol{r}) + \beta_2^2\nabla^5 n_1(\boldsymbol{r}) - \ldots. \tag{8}$$

Terms involving products of derivatives are neglected as they do not occur in linear response theory. With the definition of the β parameters in this form one can observe that they are just the static compressibility coefficients that enter the hydrodynamic model. Combination of equations (7) and (8) gives the static dielectric function as

$$\frac{\omega_p^2}{\varepsilon(\boldsymbol{q},0)-1} = -\frac{\omega_p^2\{n_1(\boldsymbol{r}) + n_{ext}(\boldsymbol{r})\}}{n_1(\boldsymbol{r})} = \beta_0^2 q^2 + \beta_1^2 q^4 + O(q^6). \tag{9}$$

The expansion coefficients here are determined by the functionals in equation (8). The functional $T_s[n(\boldsymbol{r}')]$ was evaluated by Kirzhnits (1957) and Hodges (1973) but the exchange and correlation functional $E_{xc}[n(\boldsymbol{r}')]$ is less well known. An expression for the local part of this latter functional is (Nozières and Pines 1958)

$$E_{xc}[n(\boldsymbol{r})] = \int d\boldsymbol{r}\left\{-\tfrac{3}{4}e^2\left(\frac{3}{\pi}\right)^{1/3}\{n(\boldsymbol{r})\}^{4/3}\right.$$
$$\left. + 0{\cdot}00518\frac{me^4}{\hbar^2}n(\boldsymbol{r})\ln\left(\frac{3m^3e^6}{4n(\boldsymbol{r})\hbar^6}\right) - 0{\cdot}0575\frac{me^4}{\hbar^2}n(\boldsymbol{r})\right\}. \tag{10}$$

This result should be valid in the region of metallic densities (Raimes 1972). Substitution of the results for both functionals into equation (8) gives

$$m\beta_0^2 = \tfrac{2}{3}E_F(1 - 0{\cdot}166\,r_s - 0{\cdot}00422\,r_s^2), \tag{11}$$

where $r_s = me^2\hbar^{-2}(3/4\pi n_0)^{\frac{1}{3}}$ and E_F is the Fermi energy. The first gradient correction

to $E_{xc}[n(r)]$ has been calculated by Rasolt and Geldart (1975). The parameter β_1^2 is related to a function $c(r_s)$ which they evaluated numerically by the result

$$n(r)\nabla\{\delta E_{xc}[n(r')]/\delta n(r)\}_{3LG} = -2e^2 n_0^{-\frac{1}{3}} c(r_s)\nabla^3 n_1(r), \tag{12}$$

where the subscript 3LG indicates the third-order linear gradient term. In the region of metallic densities, $c(r_s)$ is well parameterized by

$$c(r_s) = (2{\cdot}68 - 0{\cdot}17 r_s)\times 10^{-3}. \tag{13}$$

Hence from equation (8) we obtain

$$m\beta_1^2 = \tfrac{1}{18}(1 + 0{\cdot}311 r_s - 0{\cdot}0197 r_s^2). \tag{14}$$

The preceding results for β_0^2 and β_1^2 may now be used to calculate the values of γ_1 and γ_2. From equations (3) and (6) we may write

$$\frac{\omega_p^2}{\varepsilon(q,0)-1} = \tfrac{1}{3}V_F^2\left(1 - \frac{\lambda^2\gamma_1}{K_F^2}\right) + \left(\frac{1}{36} - \frac{\lambda^2\gamma_2}{3K_F^2}\right)\frac{\hbar^2 q^2}{m^2} + O(q^4), \tag{15}$$

where λ is the Thomas–Fermi wavevector and V_F the Fermi velocity. Thus comparing equations (9) and (15) one finds

$$\gamma_1 = \tfrac{1}{4} + 0{\cdot}00636 r_s, \qquad \gamma_2 = -0{\cdot}0391 + 0{\cdot}00248 r_s. \tag{16}$$

Within this approximation scheme then the dielectric function is defined by equations (3), (5), (6) and (16). When

$$\varepsilon(q,\omega) = 0, \tag{17}$$

an infinitesimal perturbation will produce a significant change in the charge density; this is the plasmon condition. Equation (17) has a solution of the form given by equation (2a) with

$$\bar{\beta}_0^2 = \frac{3V_F^2}{5} - \frac{\gamma_1\omega_p^2}{K_F^2}, \qquad \bar{\beta}_1^2 = \frac{\hbar^2}{4m^2} + \frac{12V_F^4}{175\omega_p^2} - \frac{\omega_p^2(\gamma_2+\gamma_1^2)}{K_F^4}. \tag{18}$$

Thus for the dimensionless parameters b_0 and b_1 one finds

$$b_0 = \frac{2{\cdot}21}{r_s^2}\left(1 - 0{\cdot}0921 r_s - 0{\cdot}00235 r_s^2\right), \tag{19a}$$

$$b_1 = \frac{0{\cdot}310}{r_s}\left(1 + 0{\cdot}806 r_s - 0{\cdot}004\,18 r_s^2 - 0{\cdot}001\,01 r_s^3 - 0{\cdot}000\,007\,28 r_s^4\right). \tag{19b}$$

Values of b_0 given by equation (19a) agree well with those of previous authors (e.g. Vashistra and Singwi 1972; see Table 1) but they do not agree with the experimental assignments made by Sturm (1978*b*). Nevertheless the corrections beyond the RPA are clearly in the right direction and are of the correct order of magnitude. As we shall see in Section 3, the discrepancy is in the interpretation of the experimental results. When full account of the plasmon lifetime and of the width of the incident beam are included, these results for b_0 and b_1 are found to be in satisfactory agreement with experiment also.

3. Lifetime Effects

In Section 2 the quantity $G(\boldsymbol{q}, \omega)$ was approximated by its static value and the comparison of the results with experiment was sufficiently encouraging that we might expect to obtain a better result with the refined approximation

$$G(\boldsymbol{q}, \omega) \approx G(\boldsymbol{q}, 0) + \omega\left(\frac{\partial G(\boldsymbol{q}, \omega)}{\partial \omega}\right)_{\omega=0} = G(\boldsymbol{q}, 0) + \left(\frac{\omega}{\omega_p}\right) G_1(\boldsymbol{q}). \tag{20}$$

However, one must be rather cautious in adopting a Taylor expansion since the sum rule

$$\{\varepsilon(\boldsymbol{q}, \omega)\}^{-1} \to 1 + \omega_p^2/\omega^2 \qquad \text{as} \qquad \omega \to \infty \tag{21}$$

requires that

$$(\omega_p^2/\omega^2) G(\boldsymbol{q}, \omega) \to 0 \qquad \text{as} \qquad \omega \to \infty. \tag{22}$$

Table 1. Comparison of values of dispersion coefficients

Note that the experimental assignments of the coefficient b_0 made by Sturm (1978*b*) (column 2) differ significantly from those he gave using the results of the calculation by Vashistra and Singwi (1972) (column 3), but the latter results agree well with those obtained from equation (19a) in the present work (column 4). The present results for b_1 from equation (19b) are also listed here (column 5)

(1) Element	(2) b_0(exp)	(3) b_0(VS)	(4) b_0(19a)	(5) b_1(19b)	(1) Element	(3) b_0(VS)	(4) b_0(19a)	(5) b_1(19b)
Al	0·25	0·40	0·41	0·40	Li	0·13	0·14	0·34
Sn	0·43	0·34	0·35	0·39	Na	0·080	0·082	0·32
In	0·29	0·27	0·29	0·37	K	0·035	0·044	0·30
Mg	0·18	0·22	0·23	0·36				

Thus the inclusion of a finite number of terms beyond that given by equation (20) will contradict the sum rule. From the analytic properties of the response function for an electron gas (Pines 1964) it follows that $G_1(\boldsymbol{q})$ must be purely imaginary and given by a simple power series in q^2. Indeed, as with $G(\boldsymbol{q}, 0)$ we should expect the leading term to be of order q^2 (see e.g. du Bois and Kivelson 1969). However, plasmons are found experimentally to have a width $\sim 0{\cdot}5$ eV at $q = 0$ (e.g. Kloos 1973). Mechanisms that can contribute to this finite lifetime include phonon and impurity scattering, interband transitions and plasmon absorption by the incident electron. Thus we find

$$G_1(\boldsymbol{q}) = \mathrm{i}\{\bar{\gamma}_0 + \bar{\gamma}_1 (q/K_F)^2 + \bar{\gamma}_2 (q/K_F)^4 + O(q^6)\}. \tag{23}$$

If $\bar{\gamma}_0$ were zero we would not require $\bar{\gamma}_2$ to evaluate the dispersion to fourth order, and in the following analysis $\bar{\gamma}_2$ and higher terms will be omitted. The zeros of the dielectric function now lie in the complex plane at

$$\omega = \omega_p\{1 - \tfrac{1}{2}\mathrm{i}\bar{\gamma}_0 + (\tfrac{6}{5}(E_F/\hbar\omega_p)^2 - \tfrac{1}{2}\gamma_1 - \tfrac{1}{2}\mathrm{i}\bar{\gamma}_1)(q/K_F)^2 + O(q^4)\}. \tag{24}$$

Causality demands that these solutions lie in the lower half ω plane. A value for $\bar{\gamma}_1$ has been estimated by du Bois and Kivelson (1969) but this is an order of magnitude smaller than the experimental value which is approximately

$$\bar{\gamma}_1 \sim 0{\cdot}15 r_s^{\frac{1}{2}}, \tag{25a}$$

from the results of Kloos (1973). For those metals (e.g. aluminium, sodium and potassium) which seem to fit a quadratic behaviour for their lifetimes, Kloos's results also indicate

$$\bar{\gamma}_0 \sim 0{\cdot}033\, r_s . \tag{25b}$$

Plasmons can be studied experimentally using inelastic electron loss spectroscopy. In this method an energetic electron beam (kinetic energy ~ 50 keV) is fired through a thin film of the metal and the most probable energy loss is plotted against scattering angle. The probability of the incident electron losing energy $\hbar\omega$ is given in the first Born approximation by the loss function

$$W(\boldsymbol{q},\omega) = -(8\pi e^2/\hbar q^2)\,\mathrm{Im}\{\varepsilon(\boldsymbol{q},\omega)\}^{-1} . \tag{26}$$

Using the dielectric function defined by equations (3), (6), (16), (20) and (23), from the relation (26) we obtain to fourth order in q the result

$$W(q,\omega) = (8\pi e^2/\omega_p \hbar q^2)\,\omega\{\bar{\gamma}_0 + \bar{\gamma}_1 (q/K_F)^2\}\, D^{-1} , \tag{27a}$$

where

$$D = \frac{\omega^2}{\omega_p^2}\left[\bar{\gamma}_0 + \bar{\gamma}_1\left(\frac{q}{K_F}\right)^2\right]^2 + \left[1 - \gamma_1\left(\frac{q}{K_F}\right)^2 - \gamma_2\left(\frac{q}{K_F}\right)^4 - \frac{\omega^2}{\omega_p^2}\left\{1 - \frac{3V_F^2}{5\omega^2}q^2 - \left(\frac{12V_F^4}{175\omega^4} + \frac{\hbar^2}{4m^2\omega^2}\right)q^4\right\}\right]^2 . \tag{27b}$$

There is a one to one correspondence between q and the scattering angle θ which is approximately linear (Platzman and Wolff 1973). Thus for a fixed θ (i.e. fixed q) the loss function has a peak located at the solution of

$$\omega^2 = \omega_q^2 - \frac{\omega^2\omega_p^2}{\omega_q^2 + 3\omega^2}\left[\bar{\gamma}_0 + \bar{\gamma}_1\left(\frac{q}{K_F}\right)^2\right]^2\left\{1 - \frac{3V_F^2}{5\omega^2}q^2 - \left(\frac{12V_F^4}{175\omega^4} + \frac{\hbar^2}{4m^2\omega^2}\right)q^4 + \ldots\right\}^{-2} , \tag{28}$$

where ω_q is the plasmon dispersion in the absence of any lifetimes given by equations (2) and (19). It is this peak position which is interpreted as the experimental plasmon frequency. For typical experimental values of the widths, the corrections given by equation (28) are minimal when q is less than $q_c \sim 0{\cdot}9\, r_s^{-\frac{1}{2}}$. For larger q values, damping becomes very important and it flattens the dispersion curve (Höhberger *et al.* 1975). However, the finite lifetime does play another very significant role. Experimentally the incident beam is not monoenergetic but has a finite half-width $\sim 0{\cdot}5$ eV (Krane 1978). Thus the observed spectrum for energy loss ΔE at momentum transfer $\hbar q$ takes the form

$$I(\boldsymbol{q},\Delta E) = \hbar \int_{\omega_p}^{\infty} \mathrm{d}\omega\; W(\boldsymbol{q},\omega)\, \tfrac{1}{2}\Gamma / \{(\tfrac{1}{2}\Gamma)^2 + (\hbar\omega - \Delta E)^2\} , \tag{29}$$

where Γ is the energy width at half-height of the incident electron beam. Note that it is only when both the incident beam and $W(\boldsymbol{q},\omega)$ have finite widths that this integral is nontrivial. The maximum in the energy loss peak now occurs when

$$\int_{\omega_p}^{\infty} \mathrm{d}\omega\; W(\boldsymbol{q},\omega)(\hbar\omega - \Delta E)\Gamma / \{(\tfrac{1}{2}\Gamma)^2 + (\hbar\omega - \Delta E)^2\}^2 = 0 . \tag{30}$$

Note from equation (30) that for $\Delta E > \hbar\omega_p + \Gamma$ and the widths not too large the Lorentzian just serves to broaden the plasmon peak. For ΔE close to ω_p the cutoff in plasmon frequencies at ω_p combined with the finite plasmon lifetime and the width of the incident beam lead to a flattening of the dispersion curve defined by equation (30); this curve will be referred to hereinafter as the broadened dispersion curve. This flattening at small q values is illustrated in Fig. 1 for aluminium ($r_s = 2{\cdot}073$),

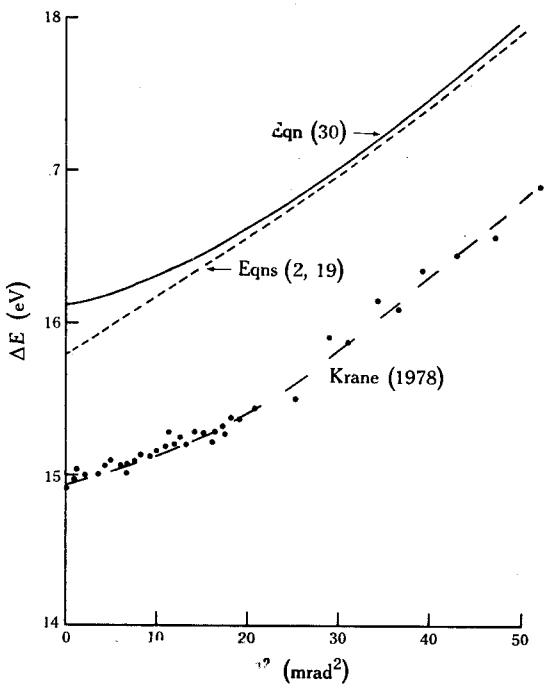

Fig. 1. Position of the peak in the spectrum for electron energy loss ΔE in aluminium, shown as a function of the scattering angle θ. The experimental points from Krane (1978) are compared with the theoretical broadened dispersion curve defined by equation (30) and also the dispersion calculated from equations (2) and (19).

with a plasmon width given by equations (25) and an incident beam half-width of 0·5 eV. As can be seen from the figure, the broadened dispersion curve (defined by equation 30) lies consistently above that given by equations (2) and (19). For comparison the experimental results of Krane (1978) are also shown; these lie well below the theoretical curves since core polarization (Kunz 1965) and band structure (Bross 1978) effects have not been included here. As pointed out by Sturm (1978*b*), the main consequence of these additional contributions is just to shift the dispersion curve to lower energies. Such a shift would bring the broadened dispersion curve and the experimental results into good agreement.

4. Conclusions

The dielectric response function for an electron gas has been considered in a 'mean field' approximation. Within this scheme the plasmon dispersion coefficients b_0 and b_1 were shown to be simply related to the exchange and correlation functional.

Expressions for b_0 and b_1 were derived which could enable a direct experimental check to be made on the exchange and correlation functional expansion coefficients in the region of metallic densities. Both b_0 and b_1 are important in determining the dispersion in the region $0 < q < q_c$ for which plasmons are a well-defined oscillation. The values of b_0 derived here agree well with those found by other authors, but b_1 has not previously been calculated beyond the random phase approximation.

By introducing a finite plasmon lifetime and width of the incident electron beam, we have been able to explain the 'two-branch' behaviour of plasmon dispersion, and hence have found that equations (19) give reasonable values of the parameters b_0 and b_1 for aluminium. We can therefore conclude that plasmon dispersion in simple metals (i.e. those which most closely resemble the jellium model) is reasonably well understood and that it is necessary to take into account the finite width of the incident electron beam in any analysis of inelastic electron energy loss spectra.

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