Pramāṇa, Vol. 24, No. 6, June 1985, pp. 905–910. © Printed in India.

Plasmon dispersion and linewidth in aluminium

R K PAL* and D N TRIPATHY

Institute of Physics, Sachivalaya Marg, Bhubaneswar 751 005, India

*Present address: Department of Physics, Sri Venkateswara College, Dhaula Kuan, New Delhi 110021, India

MS received 28 June 1982; revised 25 June 1984

Abstract. Plasmon dispersion in Al is estimated using the expression for the dynamic structure function, $S_{\mathrm{pl}}(\mathbf{k}, \omega)$, corresponding to the plasmon excitations in a many-electron system derived earlier. An evaluation of its plasmon linewidth is also presented. It is observed that for Al both the dispersion and linewidth agree fairly well with experiments.

Keywords. Dynamic structure function; one-plasmon propagator; dielectric function; synchrotron radiation; pair correlation function.

PACS No. 71·45

1. Introduction

Plasmons are well-defined collective excitations of the interacting electron gas up to a critical wave-vector k_c. For wave-vectors beyond k_c, they decay into particles and holes. Recent measurements (Eisenberger *et al* 1975; Zacharias 1975; Gibbons *et al* 1976; Batson *et al* 1976) on the plasmon dispersion in Al, Li and Na indicate that the frequency of the plasmon mode in all these systems in the long wavelength limit ($k \to 0$) is less than the classical plasma frequency, ω_{pl}. Besides, these experiments also show that plasmons exist to values of wave-vector far beyond k_c^{RPA} where $k_c^{RPA} = 0{\cdot}47\, r_s^{1/2}\, k_F$.

Recently, it has been theoretically shown (Tripathy and Pal 1981; Pal *et al* 1980) that plasmons continue to exist beyond k_c^{RPA}. Tripathy and Pal (1981) obtained the one plasmon propagator for the electron-plasmon system using Bohm-Pines Hamiltonian by treating the electron-plasmon interactions as the perturbation. Analysing the poles of this propagator the observed plasmon frequency shift at $k = 0$ is reproduced in all the above mentioned systems by redefining the critical wave-vector. The value of the new k_c is much larger than k_c^{RPA} implying that the plasmons exist beyond k_c^{RPA}. Having obtained this we also evaluated the dynamic structure function, $S_{\mathrm{pl}}(\mathbf{k}, \omega)$, corresponding to plasmon excitations in Li. Adding this to the dynamic structure function for the quasi-particle excitations denoted by $S_{\mathrm{qp}}(\mathbf{k}, \omega)$ we obtained the total $S(\mathbf{k}, \omega)$ for Li. The $S_{\mathrm{qp}}(\mathbf{k}, \omega)$ used by us was calculated using the dielectric function of Tripathy and Mandal (1977) (TM-dielectric function). Comparing our plasmon dispersion curve for Li with the experimental one it was found that although the agreement with experiment was satisfactory there was some noticeable departure in the case of theoretical dispersion curve from the experimental one. Our calculated value of the plasmon linewidth at half-height, $\Delta E_{1/2}(k)$, was negligibly small. For $K < 0{\cdot}4\, k_F$, this is in contrast to the experiment which gives a finite value even at $k = 0$. The reason for this discrepancy is not clear. Lithium being a comparatively low density system having $r_s \approx 3{\cdot}22$, one might think that our evaluation of $S(\mathbf{k}, \omega)$ is perhaps not so good at low densities. This may be due to two reasons. One is that the derivation of the one-plasmon propagator over which our calculation of $S_{\mathrm{pl}}(\mathbf{k}, \omega)$ depends has perhaps the deficiency of accounting for local field correction effects properly (Tripathy and Pal 1981). Such effects are supposed to be important at low densities. The other reason could be that the $S_{\mathrm{qp}}(\mathbf{k}, \omega)$, evaluated by us using the TM-dielectric function, is not good enough to account for higher-order correlation effects. It has been shown that the TM-dielectric function can be considered to be very reliable in the sense that it has given rise to some interesting structure in the static structure factor, $S(\mathbf{k})$, (Tripathy *et al* 1977; Rao *et al* 1983) which has been verified in the recent experiment by Eisenberger *et al* (1980) using synchrotron radiation. Besides, the TM-dielectric function satisfies the compressibility sum rule and produces a positive value of the pair correlation function, $g(r)$, at $r = 0$ up to $r_s \leqslant 4$. However for $r_s > 4$, this has the unphysical feature of not giving rise to a positive value of $g(0)$. This very deficiency of having $g(0) < 0$ for $r_s \simeq 4$ may be understood as due to the lack of higher-order short range correlation effects in the TM-dielectric function. It is therefore speculated that the theoretical results for plasmon dispersion and linewidth should be in better agreement with the experiment in the case of Al, a relatively high density system with $r_s \simeq 2{\cdot}07$, than in Li whose r_s is $3{\cdot}22$. This is what has been verified in the present paper. Plasmon dispersion and linewidth for Al is calculated using our earlier theory (Tripathy and Pal 1981).

2. Results and discussions

The expression for $S_{\mathrm{pl}}(\mathbf{k}, \omega)$ is given by

$$S_{\mathrm{pl}}(\mathbf{k}, \omega) = -\frac{4k^2}{3\pi^3}(mK_F)\frac{\operatorname{Im}\Sigma(\mathbf{k}, \omega)}{[\omega^2 - \omega_{\mathrm{pl}}^2 - \operatorname{Re}\Sigma(\mathbf{k}, \omega)]^2 + [\operatorname{Im}\Sigma(\mathbf{k}, \omega)]^2}, \quad (1)$$

where $\Sigma(\mathbf{k}, \omega)$ denotes the self-energy operator for the plasmons. We have discussed earlier the diagrammatic representation of $\Sigma(\mathbf{k}, \omega)$ (Tripathy and Pal 1981). Using (1) we evaluate $S_{\mathrm{pl}}(\mathbf{k}, \omega)$ for Al as a function of ω for a set of k values which are plotted in figure 1. To evaluate $S_{\mathrm{qp}}(\mathbf{k}, \omega)$ we choose the TM-dielectric function and make use of the fluctuation dissipation theorem. The reason for choosing the TM-dielectric function has been mentioned earlier. Adding these two contributions we obtain the value of the total $S(\mathbf{k}, \omega)$ as a function of ω for several values of the momentum transfer k. These are shown in figure 2. From the values of ω at the peak positions of these $S(\mathbf{k}, \omega)$ we determine the plasmon frequency as a function of k and hence obtain the plasmon dispersion. Plasmon dispersion in Al is plotted in figure 3 and has been compared with the experimental data of Batson *et al* (1976). As one can see from the dispersion curve the agreement with experiment in Al is reasonably good. Although the plasmon dispersion curve for Al obtained by Holas *et al* (1979) agrees better with experiments as compared to our values, their theory does not explain either the negative frequency shift to ω_{pl} at $k = 0$ or the extension of plasmon mode to a region beyond k_c^{RPA}. Therefore one cannot say that the ω values in their graph beyond k_c^{RPA} correspond to plasmons. Our calculated value for the plasmon linewidth, $\Delta E_{1/2}(k)$, in Al is also given in figure 4. In this figure, the dotted line represents the experimental results of Gibbons *et al* (1976). Let us now compare the plasmon dispersion curve in Al with that obtained by us for Li

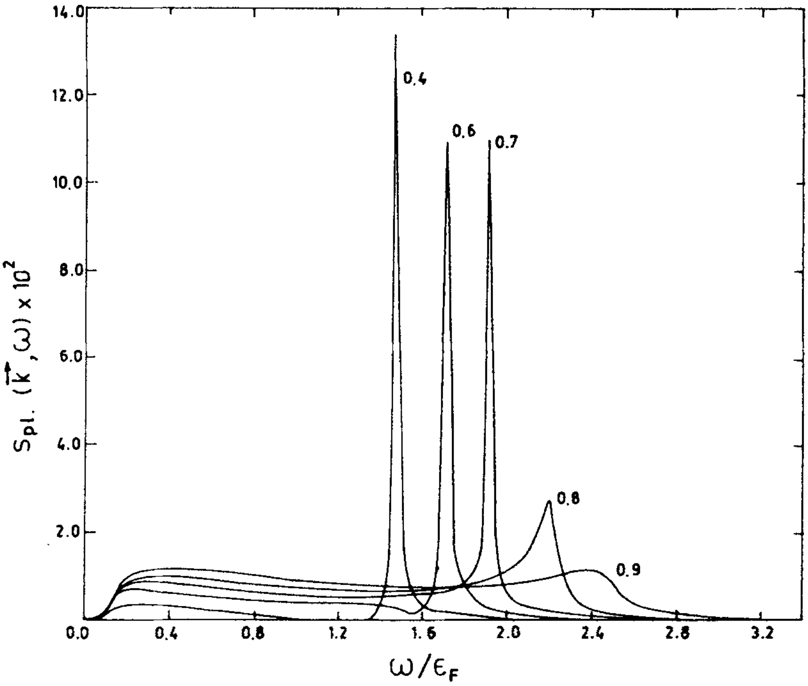

Figure 1. Plots of $S_{pl}(\mathbf{k}, \omega)$ (in units of mk_F) *vs* ω for different k-values.

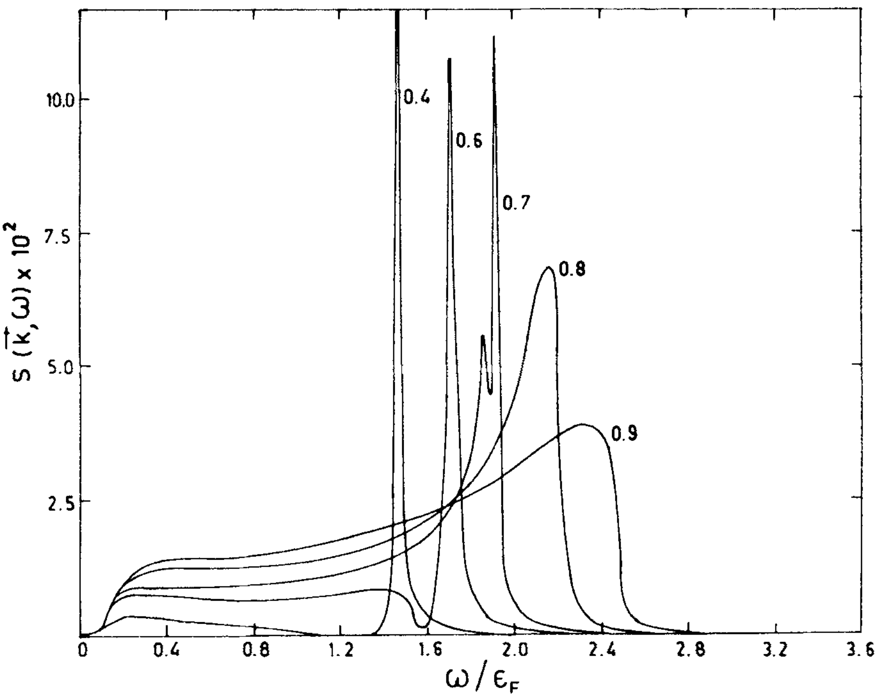

Figure 2. Plots of $S(\mathbf{k}, \omega)$ (in units of mk_F) *vs* ω for different k-values.

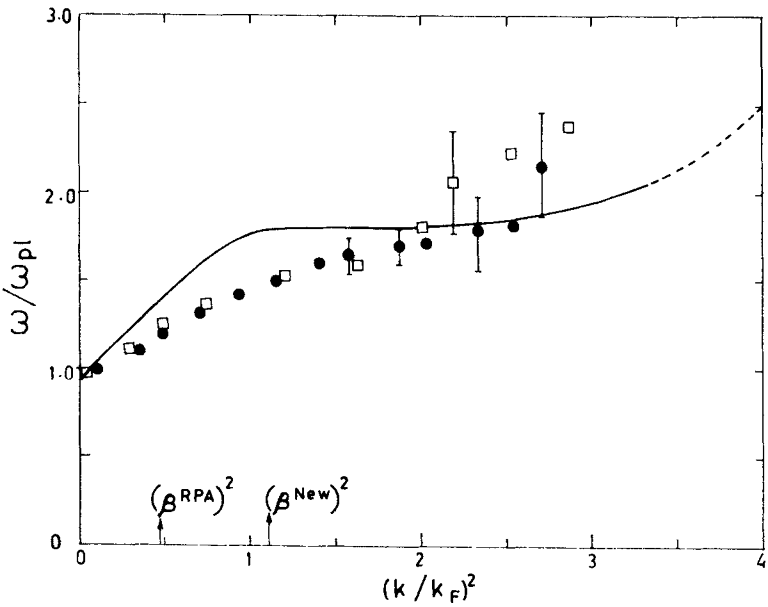

Figure 3. Dispersion relation for the plasmon in Al (——) present theory, (□) and (●) experimental data of Batson *et al* (1976).

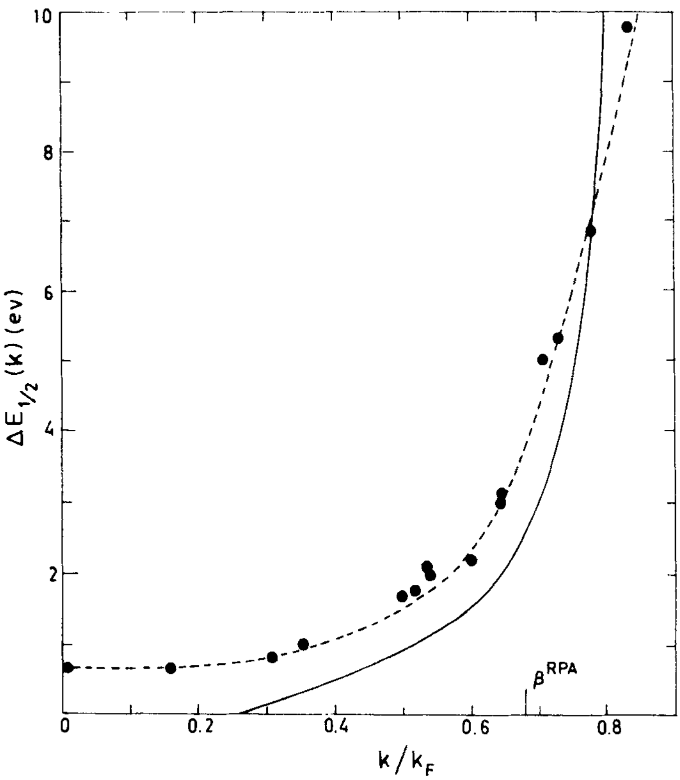

Figure 4. Plasmon linewidth, $\Delta E_{1/2}(k)$ *vs* k in Al (——) present theory (- - -) Experimental data of Gibbons *et al* (1976).

Table 1. Difference (δ) between theoretical and experimental values of the plasmon frequency for Al and Li (Tripathy and Pal 1981) for various values of K.

	Al		Li	
$k(\text{Å}^{-1})$	k/k_F	δ(eV)	k/k_F	δ(eV)
0·3	0·171	0·19	0·273	0·25
0·5	0·286	0·61	0·454	0·70
0·7	0·400	1·14	0·636	1·36
0·9	0·514	1·62	0·818	2·31
1·1	0·629	2·19	1·000	3·52
1·3	0·743	3·34	1·182	3·52

earlier. We notice that our dispersion curve for Al agrees better with experiments over a wider range of k values than that for Li (Tripathy and Pal 1981). To show that, we have calculated the difference, $\delta = (\omega_k^{\text{theory}} - \omega_k^{\text{expt.}})$ for Al and Li for several values of k. This is given in table 1. From the table we notice that for the same values of k, δ, for Al, is less compared to that for Li. This implies that the agreement with experiment is better for Al ($r_s \simeq 2{\cdot}07$) than for Li ($r_s = 3{\cdot}22$). Comparing the plasmon linewidth plot with the plot for Li given eariler we have found that for Li the contribution to $\Delta E_{1/2}(k)$ starts at $k = 0{\cdot}4\,k_F$ whereas for Al it starts at $k \simeq 0{\cdot}25\,k_F$. Any contribution to $\Delta E_{1/2}(k)$ for $k < k_c^{RPA}$ should be visualized as a non-*RPA* effect which is to be considered important for low density systems. Since the non-*RPA* effects, built in, in our earlier theory is not adequate enough for Li, decay of a plasmon into particle-hole pairs starts at a higher k value ($k \simeq 0{\cdot}4\,k_F$) than what is seen for Al ($k \simeq 0{\cdot}25\,k_F$). Thus from the study of plasmon dispersion and linewidth of Al and their comparison with those of Li it is clear that our earlier theory should work better for high density systems ($r_s \leqslant 2$) than for low density systems.

We conclude by saying that our original theory meant for the study of plasmon excitations in a many-electron system may be considered more reliable than the various mean field theories because unlike the mean field theoretic approaches our method reproduces not only the negative frequency shift to ω_{pl} at $k = 0$ but also gives a finite value for plasmon linewidth for $k < k_c^{RPA}$ which for Al goes as close as $k \simeq 0{\cdot}25\,k_F$ where $k_c^{RPA} \simeq 0{\cdot}68\,k_F$. This contribution in the region $0{\cdot}25\,k_F \leqslant k \leqslant k_c^{RPA}$ arises from the bilinear interaction term in the electron-plasmon coupling, that is, from the imaginary part of $\Delta_0(\mathbf{k}, \omega)$ (Tripathy and Pal 1981). Since for Al the experiment indicates a finite value for the linewidth at $k = 0$ it seems that the contribution to the plasmon linewidth for k approaching zero is due to some different mechanism than due to decay of a plasmon into a particle-hole pair.

From our plot of the total $S(\mathbf{k}, \omega)$ for various k shown in figure 2, we observe that there is a double peak structure in $S(\mathbf{k}, \omega)$ around $k \simeq k_c^{RPA}$. This structure is to be interpreted as due to the superposition of plasmon and quasi-particle excitations. The existence of such a structure around k_c^{RPA} has been speculated from the measurements of Batson *et al* (1976). It is worth commenting on the experimentally observed double peak structure in $S(\mathbf{k}, \omega)$ in systems like Al, Li, Be etc. (Eisenberger *et al* 1975; Platzman and Eisenberger 1974; Eisenberger and Platzman 1976). It is noticed that this structure is seen in all these system for values of momentum transfer close to $2\,k_F$. Although our theory shows a continuation of the plasmon mode to a region beyond k_c^{RPA} it does not still exist up to the k-values where the double peak structures are found. We, therefore, think that the structure in $S(\mathbf{k}, \omega)$ for k close to $2\,k_F$ is not due to the superposition of plasmon and quasi-particle spectrum. Rather it may be due to the superpositions of one and multi-pair excitations in the system. Evidence in favour of the latter argument is found from the studies of Green *et al* (1982) and Bhuyan and Tripathy (1982).

References

Batson P E, Chen C H and Silcox J 1976 *Phys. Rev. Lett.* **37** 937
Bhuyan M and Tripathy D N 1982 *Phys. Lett.* **A91** 252
Eisenberger P, Marra W C and Brown G S 1980 *Phys. Rev. Lett.* **45** 1439
Eisenberger P and Platzman P M 1976 *Phys. Rev.* **B13** 934
Eisenberger P, Platzman P M and Schmidt P 1975 *Phys. Rev. Lett.* **34** 18
Gibbons P C, Schnatterly S E, Ritsko J J and Fields J R 1976 *Phys. Rev.* **B13** 2451
Green F, Lowy D N and Szymanski J 1982 *Phys. Rev. Lett.* **48** 638
Holas A, Aravind P K and Singwi K S 1979 *Phys. Rev.* **B20** 4912
Pal R K, Tripathy D N and Mandal S S 1980 *Phys. Status Solidi* **B100** 651
Platzman P M and Eisenberger P 1974 *Phys. Rev. Lett.* **33** 152
Rao B K, Pal R K and Tripathy D N 1983 *Solid State Commun.* **45** 95
Tripathy D N and Mandal S S 1977 *Phys. Rev.* **B16** 231
Tripathy D N and Pal R K 1981 *Phys. Status Solidi* **B104** 345
Tripathy D N, Rao B K and Mandal S S 1977 *Solid State Commun.* **22** 83
Zacharias P 1975 *J. Phys.* **15** 645