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The plasmon density of states of a layered electron gas

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Abstract. The plasmon density of states (DOS) $\rho(\omega)$ of a layered electron gas (LEG) is studied theoretically. It is shown that $\rho(\omega)$ is a linear function of frequency ω as ω goes to 0, and then increases more rapidly as ω tends to the screened, three dimensional plasma frequency ω_p. We also study the partial densities of state for constant κ and q_z, where κ and q_z are the magnitude of the transverse and perpendicular momentum transfers. A possible experimental probe for the plasmon DOS is discussed.

1. Introduction

The new superconducting cuprates [1–4] with high superconducting transition temperatures T_c are all characterized by a layered structure of conducting (CuO_2) planes. We propose to use the layered electron gas (LEG) model [5] to describe their collective charge excitation (layer plasmons) and to calculate their density of states (DOS) as a function of frequency in this paper. The structure of the plasmon spectrum in layered, conducting systems differs in a striking way from those of the isotropic ($3D$) conductors. In the latter case, the plasmon branch is described by a dispersion relation $\omega(q)=\omega_0+aq^2$, whereas a layered conductor is characterized by a highly anisotropic plasmon band $\Omega(\kappa, q_z)$ without a gap at $\kappa=0$ except for the single branch for which $q_z=0$ (κ is a two-dimensional wave-vector in the plane of the sheets and q_z has been chosen perpendicular to the layers). That produces noticeable changes in the spectrum of collective modes of these materials. In experimental probes, such as absorption of light, the dielectric response is subject to strong momentum selection rules. As a result only the long-wavelength dielectric response is probed in such experiments. In local experimental probes, such as the sudden creation of a core hole by X-ray photo-electron spectroscopy, one can, in principle, couple to the full spectrum of plasmons. This shows up as a side-band of the core-hole excitation spectrum due to plasmon shake-up. It is conceivable that the plasmon-pole approximation can be used for the calculation of the spectral shape of the plasmon side-band measured with such a local probe. The plasmon-pole approximation replaces the response function with the density of states of the layer plasmons, which is defined by

$$\rho(\omega)=(2\pi)^{-3}\sum_{\kappa,q_z}\delta(\omega-\Omega_p(\kappa,q_z)). \tag{1}$$

The nature of the collective excitations in layered systems has been studied extensively [5, 6] theoretically, because of continuing interest in superlattices and naturally layered materials such as graphite and the transition metal chalcogenides (TaS_2, TeS_2, etc.).

The outline of this paper is as follows: In Sect. 2 we rederive the layer plasmon dispersion relations [6] using the random phase approximation (RPA). In Sect. 3 we calculate the partial density of states, keeping either of the variables κ (the absolute value of the LEG plasmon wavevector parallel to the conducting planes) or q_z (the LEG plasmon wavevector perpendicular to the planes) constant. In Sect. 4 we derive a simple form of the total plasmon DOS, based on an approximate form of the LEG dispersion relations. In Sect. 5 we calculate both analytically and numerically the exact layer plasmon density of states and discuss the various frequency regimes $\omega/\omega_p \ll 1$, $=1$ and >1. Finally – in Sect. 6 – discuss the implications of these results for the shake-up spectra of core levels in a layered compound.

2. Plasmon dispersion relations

We assume that the carriers are localized in $N(N\to\infty)$ independent layers, spaced a distance D apart. The dispersion relation for two-dimensional carriers is assumed to be given by the effective mass approximation: $\varepsilon_k=k^2/2m^*$. Electrons on different sheets are coupled by the Coulomb interaction. We can write an electronic Hamiltonian H^{el}.

$$H^{el}=\sum_{\kappa,l}\varepsilon_{\kappa l}A^+_{\kappa l}A_{\kappa l}+\sum_{\kappa,\kappa',\kappa''}\sum_{l,l'}V^{(0)}_{\kappa''}(l,l')A^+_{\kappa'+\kappa'' l}A^+_{\kappa'-\kappa'' l'}A_{\kappa l'}A_{\kappa' l} \tag{2}$$

where κ, κ' and κ'' are two-dimensional momentum variables, and l, l' label different layers, $A_{\kappa,l}$, $A^{+}_{\kappa,l}$ are carrier annihilation and creation operators and the interaction matrix element $V^0_{\kappa}(l,l')$ is the two-dimensional Fourier transform of the Coulomb interaction between electrons on layers l and l'. For an infinite system $V(l,l')=V(l-l')$.

$$V^{(0)}_{\kappa}(l-l')=\frac{2\pi e^2}{\varepsilon_M \kappa}\, e^{-\kappa D|l-l'|}. \tag{3}$$

If we make a Fourier transformation of the variable $r=l-l'$ we obtain

$$\Phi^0(\kappa;q_z)=\sum_{r=-\infty}^{\infty} V^{(0)}_{\kappa}(r)\exp(\mathrm{i}q_z Dr) = \frac{2\pi e^2}{\varepsilon_M \kappa} F(\kappa D, q_z D), \tag{4}$$

where we use the dimensionless function F (the 'layer form factor'), introduced by Fetter [5b] in the hydrodynamic treatment of the layer plasmon dispersion

$$F(\kappa D, q_z D)\equiv\frac{\sinh \kappa D}{\cosh \kappa D-\cos q_z D}. \tag{5}$$

The study of the screening properties and collective modes requires the knowledge of the fully screened interaction $V(l,l')$. Within the random phase approximation, the latter obeys the equation

$$V(l,l')=V^0(l,l') + \Pi^0(\mathbf{q};\mathrm{i}\omega)\sum_{l''} V^0(l,l'')\,V(l'',l'), \tag{6}$$

where the single layer polarizability Π^0 is given by

$$\Pi^0(\mathbf{q};\mathrm{i}\omega)=\frac{2}{A}\sum_{\kappa}\frac{f(\varepsilon_{\kappa+\mathbf{q}})-f(\varepsilon_k)}{\varepsilon_{k+\mathbf{q}}-\varepsilon_k-\mathrm{i}\omega}. \tag{7}$$

Here A is the area of one layer and $f(\varepsilon_\kappa)$ is the Fermi function. The layer plasmons are defined as the zeroes of the layer dielectric function

$$\varepsilon(\kappa,q_z;\omega)=1-\Phi^0(\kappa,q_z)\,\mathrm{Re}\{\Pi^{(0)}\}. \tag{8}$$

In the frequency regime $\omega > v_F\kappa$, the real part of the polarizability χ_0 of a single sheet is [5, 7]

$$\mathrm{Re}\{\Pi^0(\kappa;\omega)\}=\frac{m^*}{\pi\hbar^2}\left[\frac{\omega}{[\omega^2-(v_F\kappa)^2]^{1/2}}-1\right]. \tag{9}$$

In order to keep the expressions simple we introduce dimensionless variables for the in-plane and out-of-plane wave-vector components κ and q_z and the LEG plasmon frequency ω_p, by defining $y\equiv\kappa D$, $z\equiv q_z D$, $\omega_p(y,z)\equiv\Omega_p(y,z)/\Omega_0$ with $\Omega_0^2=2v_F^2/(Dr_B)$ and $\nu_F\equiv v_F/(D\Omega_0)=(r_B/2D)^{1/2}$. Here $r_B\equiv\frac{\hbar^2\varepsilon_M}{m^* e^2}$ is the effective Bohr radius, m^* the effective carrier mass and ε_M is the background dielectric constant of the medium due to all other high frequency excitations. (A typical value is $\nu_F^2\sim 0.05$). With these definitions, and solving for the roots of $\varepsilon(\kappa,q_z;\omega)=0$, we obtain with (8)

$$0=1-\frac{F(y,z)}{\nu_F^2 y}\left[\frac{\omega_p}{[\omega_p^2-(\nu_F y)^2]^{1/2}}-1\right]. \tag{10}$$

From this equation we solve the layer plasmon dispersion relation

$$\omega_p^2(y,z)=\nu_F^2 y^2+\frac{F^2(y,z)}{\nu_F^2+2F(y,z)/y}. \tag{11}$$

Alternatively we can express this as a dispersion relation for Ω as a function of κ and q_z. When written in this form the dispersion relation reads

$$\Omega_p(\kappa,q_z) = v_F\kappa\left[1+\left[\frac{2F(\kappa D,q_z D)}{r_B\kappa}\right]^2\frac{1}{1+\frac{4F(\kappa D,q_z D)}{r_B\kappa}}\right]^{1/2}.$$

For later use it is convenient to express F as a function of ω_p and y using (10), and also to express z in terms of F and y, by inverting (5). The resulting expressions are found to be

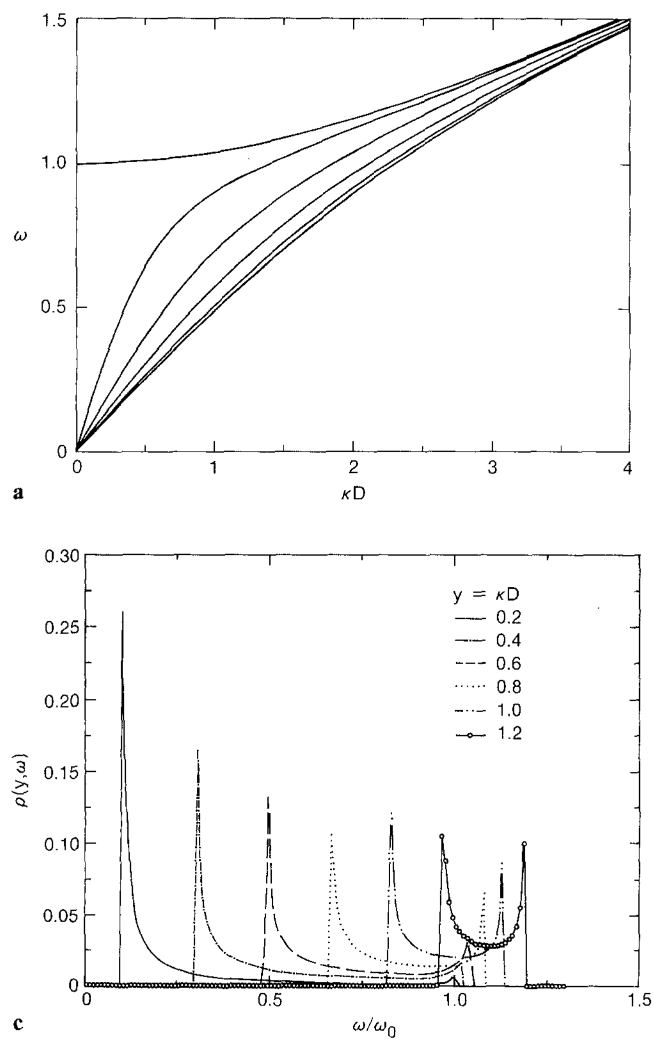
$$F(\omega_p,y)=\frac{\nu_F^2 y\sqrt{\omega_p^2-(\nu_F y)^2}}{\omega_p-\sqrt{\omega_p^2-(\nu_F y)^2}},$$
$$z(\omega_p,y)=\arccos\left(\cosh(y)-\frac{\sinh(y)}{F(\omega_p,y)}\right). \tag{12}$$

It should be noted here that (11) goes beyond the earlier RPA results as it used the exact form of the single sheet polarizability χ_0 (9) and not its expansion valid for $\omega \gg v_F\kappa$ only. On the other hand, we have not included the effects of the non-local exchange and correlation part of the electron-electron interactions. Some exploration of the inclusion of such effects by a local field correction for the in-plane electron-electron interaction has been recently made in [8]. We note that the general consequence of such treatments as the GRPA (generalized random phase approximation) is to reduce the strength of the electron-electron interactions.

3. Plasmon partial density of states

Let us first address the question of the LEG plasmon frequency dependence for $\omega \ll 1$. In this region (see Fig. 1) $y \ll 1$ holds also, and the hyperbolic functions may be expanded in a Taylor series for small y. The LEG plasmon dispersion relations then reduce to the much simpler form

$$\omega_p^2(y,z)=\nu_F^2 y^2 + \frac{y^2}{[\frac{1}{2}y^2+1-\cos z][2+\nu_F^2(\frac{1}{2}y^2+1-\cos z)]}. \tag{13}$$



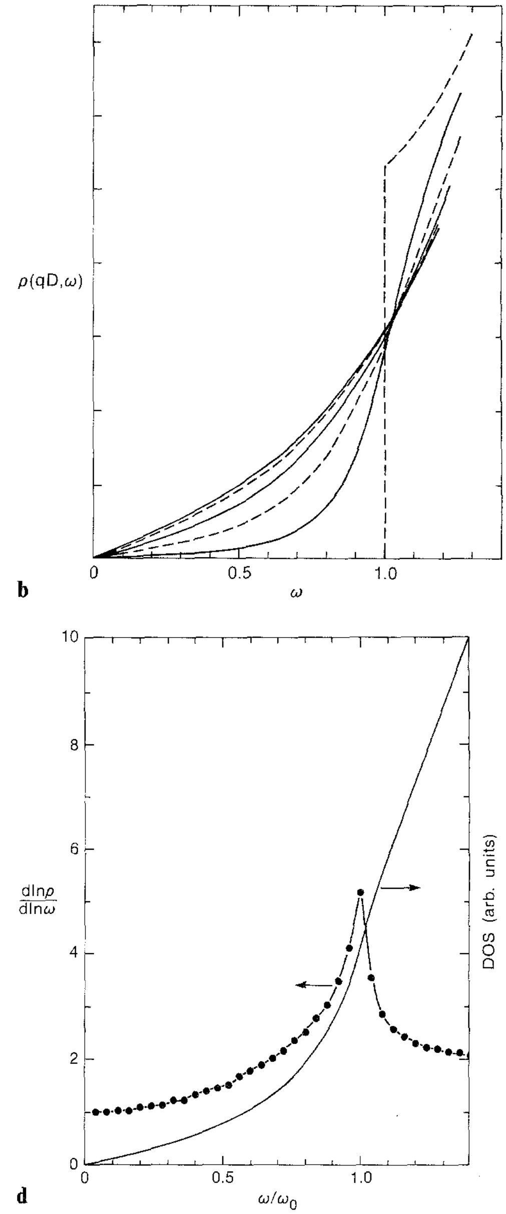


Fig. 1a–d. a Layer plasmon dispersion for $v_F^2=0.05$ and (from top to bottom) $z/\pi=0$, 0.2, 0.4, 0.6, 0.8, 1. **b** Partial DOS with z fixed at (from bottom to top) $z/\pi=0$, 0.2, 0.4, 0.6, 0.8, 1. **c** Partial DOS with y fixed at (from left to right) $y=0.2$, 0.4, 0.6, 0.8, 1.0, 1.2. **d** Total DOS (solid), and exponent β in $\rho=\omega^\beta$ versus frequency (open circles)

For $z \simeq 1$ and $\omega \ll 1$, we obtain the acoustic plasmon dispersion relation:

$$\omega_p(y,z) = y\sqrt{v_F^2+(1-\cos z)^{-1}(2+v_F^2(1-\cos z))^{-1}}. \quad (14)$$

For $z \ll 1$ and $\omega \ll 1$, and retaining only leading orders in z and y (11) becomes:

$$\omega_p^2(y,z)=v_F^2 y^2+\frac{y^2}{y^2+z^2}. \quad (15)$$

It is not transparent from this form that the dispersion is still acoustic for small ω. We rewrite the formula in the following form

$$\omega_p^2(y,z)=\frac{y^2}{z^2}(1+v_F^2(y^2+z^2))-\omega_p^2\frac{y^2}{z^2}. \quad (16)$$

Clearly for $\omega_p \ll 1$ the second term is much smaller than the first term and can be neglected, hence (11) has ω proportional to y for all values of the wavevector perpendicular to the planes (except for the special point $z=0$). As the integral over y is two-dimensional, a direct consequence of this is that the partial DOS for each fixed value of z is proportional to ω in this limit. In Fig. 1 we plot a set of dispersion curves for a number of z-values for $v_F^2=0.05$, which is a typical value. Also indicated for the same set of z-values is the partial DOS obtained by integrating numerically over y, while keeping z fixed. It is clear from this plot, that for each value of z the partial DOS rises linearly with frequency for ω below about 0.4. A singular point is reached at $z=0$, where the dispersion sets in at $\omega=1$. As the total DOS is obtained by integrating over z between $-\pi$ and π one expects to obtain a linear dependency on ω for the plasmon DOS. This is not so obvious if one considers instead the partial DOS for fixed y, while integrating over all z. This is indicated in Fig. 1c. One clearly sees the 1-dimensional nature of this partial DOS reflected in the divergencies at the points where the energy corresponds to the lower and higher branches ($z = \pm\pi$ and $z = 0$ respectively).

4. Plasmon DOS using an approximation to the dispersion formula

We first derive the DOS using a simplified dispersion formula, which nevertheless possesses the most relevant physical properties. If v_F^2 is of the order of 0.05 as usual, one may neglect the first term in (11). This only decreases the group velocity somewhat, without affecting the quasi-acoustic nature of the dispersion relation. We also replace the second term in (11) with $yF/2$, which corresponds to neglecting some higher order corrections. We furthermore replace the $\sinh(y)$ and $\cosh(y)$ in F with their limiting values for small y, which is a good approximation for small frequencies. For larger frequencies it is still reasonable and only results in a pile-up of states near the bulk plasma frequency. The result is

$$\omega_p^2(y,z) = \frac{y^2}{y^2 + 2(1-\cos(z))}. \tag{17}$$

We will first calculate the number of states per unit volume with $\omega_p(y,z) < \omega$ using the expression:

$$N(\omega) = (2\pi)^{-3} \int_{\omega_p(y,z)<\omega} \mathrm{d}^3k$$

$$= (2\pi D)^{-3} \int_{-\pi}^{\pi} \mathrm{d}z \int_{\omega_p(y,z)<\omega} \mathrm{d}^2\mathbf{y}. \tag{18}$$

The integral over $\mathbf{y}$ is equal to the area πy^2 enclosed by the contour of constant y for given z and ω. Hence we rewrite (17) as

$$y^2 = \frac{2(1-\cos(z))\,\omega^2}{1-\omega^2}. \tag{19}$$

After integration over z we obtain

$$N(\omega) = \frac{1}{2\pi D^3} \frac{\omega^2}{1-\omega^2}. \tag{20}$$

Differentiating once gives us the density of states:

$$\rho(\omega) = \frac{1}{\pi D^3} \frac{\omega}{(1-\omega^2)^2}. \tag{21}$$

5. Plasmon DOS using the full dispersion formula

We now calculate the DOS per unit volume using the relation

$$\rho(\omega) = V^{-1} \sum_{\mathbf{y},z} \delta(\omega_p(y,z) - \omega)$$

$$= \frac{2}{(2\pi D)^3} \int_0^{y_m} \left|\frac{\mathrm{d}z}{\mathrm{d}\omega_p}\right|_{\omega,y} 2\pi y\,\mathrm{d}y, \tag{22}$$

where y_m corresponds to the value of y for which $\omega_p = \omega$ on the lowest branch ($\cos(z) = -1$) of the dispersion curves for fixed z. With (11) we obtain

$$v_F^2 y_m^2 + \frac{\left[\dfrac{\sinh(y_m)}{1+\cosh(y_m)}\right]^2}{v_F^2 + \dfrac{2}{y_m}\dfrac{\sinh(y_m)}{1+\cosh(y_m)}} = \omega^2. \tag{23}$$

We now make a transformation of variables $x \equiv y/\omega$ in the integration of (22), and define $x_m \equiv y_m/\omega$. We will be interested in the behaviour for $\omega \to 0$, where the above expression reduces to

$$x_m^2 = 4\frac{1+v_F^2}{1+4v_F^2(1+v_F^2)}. \tag{24}$$

By combining (5), (11) and (12) we calculate $\mathrm{d}z/\mathrm{d}\omega_p$ and express it as a function of x and ω

$$\left|\frac{\mathrm{d}z}{\mathrm{d}\omega_p}\right|_{\omega,x} = \frac{2\omega}{(\partial\omega^2/\partial F)(\partial F/\partial\cos z)\sin z}$$

$$= \frac{x\sinh(x\omega)}{\omega^2(1-v_F^2x^2)^{3/2}\sin z}. \tag{25}$$

With the help of (12) we express $\sin z$ as a function of x and ω

$$\sin^2 z = 1 - \left[\cosh(x\omega) - \frac{\sinh(x\omega)\left(1-\sqrt{1-v_F^2x^2}\right)}{v_F^2 x\omega\sqrt{1-v_F^2x^2}}\right]^2, \tag{26}$$

which, in the limit $\omega \ll 1$ becomes

$$\sin z \simeq \left\{1 - \left[1 + \frac{1}{v_F^2}\left(\frac{1}{\sqrt{1-v_F^2x^2}} - 1\right)\right]^2\right\}^{1/2}.$$

Here we note, that in this limit $\sin z$ is a function of x and does not depend on ω. For $v_F \to 0$ the above expression reduces to

$$\sin z \simeq x\sqrt{1-\tfrac{1}{4}x^2}. \tag{28}$$

We are now ready to write down the expression for the density of states in the limit where $\omega \ll 1$. As this implies $y \ll 1$, we also substitute $\cosh(y)$ and $\sinh(y)$ with their limiting behaviour for small y. Combining (22), (24), (25) and (27) we obtain

$$\rho(\omega) = \frac{\omega}{2\pi^2 D^3} \int_0^{x_m} \frac{x^3\,\mathrm{d}x}{(1-v_F^2x^2)^{3/2}\sin z}. \tag{29}$$

As the integrand and x_m are independent of ω we see that the plasmon DOS is proportional to ω for $\omega \ll 1$. We can check this expression by choosing $v_F = 0$, as discussed in the preceding section. From (24) we see that $x_m = 2$. Expression (29) can now be calculated analytically and becomes, using (28)

$$\rho(\omega)=\frac{4\omega}{\pi^2 D^3}\int_0^1 \frac{u^2}{\sqrt{1-u^2}}\,du=\frac{\omega}{\pi D^3}, \tag{30}$$

which is equal to the limiting behaviour for small ω that we already derived in (21).

Finally we compare these analytical results to computations made by integrating the plasmon dispersion relation of (11) numerically. In Fig. 1d we display the total plasmon DOS $\rho(\omega)$, as well as the function $\omega/\rho * d\rho/d\omega$, which corresponds to the exponent β in $\rho \simeq \omega^{\beta}$. Also the numerical results show that the density of states depends linearly on ω for small frequencies in this numerical calculation. Interestingly there is only a weak discontinuity in the first derivative of the DOS at $\Omega_p/\Omega_0=1$. In reality a cutoff of the plasmon DOS must be found at larger values of the in-plane wavevector, due to non-parabolic dispersion of the electronic bands near the Brillouin-zone boundary. These effects have not been included in this study. The DOS based on the approximation for the dispersion relations (21), has a singularity at $\Omega_p/\Omega_0=1$, due to the fact that with this approximation the plasmon dispersion saturates for large values of the in-plane wavevector.

6. Discussion

In the above sections we have shown that, under conditions where the experimental probe couples to the plasmon DOS, one should observe this as a linear frequency dependency of the response function. One such possible probe would be core level spectroscopy of a 'probing" atom embedded in a LEG. The plasmon DOS should then show up as the shake-up spectrum of the core line. A necessary requirement would be to have a very narrow core-line, with an intrinsic line-width much smaller that the width of the plasmon side-band. Usually the intrinsic width of a core line is of the order of one eV due to lifetime effects, about equal to the screened plasmon energy of the high T_c cuprates (1 eV). With a proper choice of the core-line and deconvolution techniques it might be possible to determine the plasmon DOS with such an experiment.

Another possibility is, that the plasmon DOS shows up as the inverse life-time of an electron-hole pair, due to electron-electron interactions. This possibility merits, in our view, further study as a possible microscopic route to the marginal Fermi liquid model [9].

7. Conclusions

We calculated the plasmon density of states of a layered electron gas. It is shown, that this DOS is proportional to frequency ω for sufficiently small ω. This result can in principle be verified with core-level spectroscopy, where the shake-up spectrum corresponds to the density of states of collective excitations.

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