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Density dependence of the plasmon dispersion in alkali metals

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Abstract. We investigate the role of electron correlations on the plasmon dispersion in alkali metals using a sum rule approach. The single pair contribution to the low-energy part of the excitation spectrum is calculated in the framework of the Landau theory of Fermi liquids and used to estimate the plasmon contribution to the compressibility sum rule. The plasmon contribution to the f-sum rule is calculated employing a non-local effective interaction (g-matrix) recently proposed in the literature. The analysis accounts for the strong density dependence of the dispersion coefficient α exhibited by experimental data. The average energy of multipair excitations is also estimated.

Recent experiments [1] on the volume plasmon dispersion in alkali metals have shown strong deviations from the predictions of current theories for Fermi liquids [2,3] including short-range, exchange and correlation effects. A key quantity in this context is the dispersion coefficient α defined by

$$\omega_{\rm p}(q) = \omega_{\rm p}(0) + \frac{1}{m}\alpha q^2 + {\rm O}(q^4) \tag{1}$$

where $\omega_{\rm p}(0) = \sqrt{4\pi n e^2/m}$ and $n = 3/4\pi r_{\rm s}^3$ is the density. The density dependence of α has been shown to exhibit a rather unexpected behaviour in the experiment of [1] (see figure 1). In particular for $r_{\rm s} \geqslant 5$, the quantity α becomes negative in contrast with all theoretical predictions.

The aim of this work is to show that this behaviour is related to the occurrence of multipair effects that become more and more important as $r_{\rm s}$ increases and in particular to the fact that the plasmon does not exhaust the f-sum rule (a similar behaviour was recently pointed out [4] in the spin response of liquid ^{3}He). Vice versa multipair effects become negligible at small $r_{\rm s}$ where the dynamics of the system is governed by the equations of the random phase approximation accounting only for 1 particle–1 hole (1p–1h) transitions (single pair and collective plasmon excitations)[5].

The q-dependence of the contribution to the density excitation strength $|\rho_{n0}|^2 = |\langle n|\hat{\rho}_q|0\rangle|^2$ arising from the various excitations in the electron system can be deduced [5] on the basis of simple arguments based on conservation laws and sum rules. These excitations include single-pair, collective plasmon and multiparticle–multihole (multipair) transitions. The leading contributions of these transitions, at low q, to the various moments

$$m_k = \sum_n \omega_{n0}^k |\langle n|\hat{\rho}_q|0\rangle|^2 = \int S(q,\omega)\omega^k\,{\rm d}\omega \tag{2}$$

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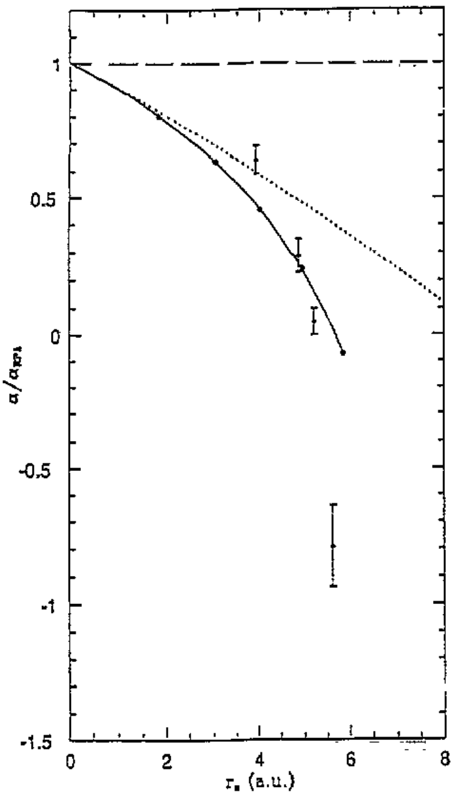

Figure 1. Volume plasmon dispersion coefficients normalized to the RPA value ($\alpha_{\rm RPA} = \frac{3}{5}\epsilon_{\rm F}/\omega_{\rm p}(0)$) against the Wigner–Seitz radius r_s. The solid line is the result of (13). The dotted line gives the dispersion coefficient as calculated from (13) with $K = 0$. Experimental data are from [1].

of the dynamic structure function

$$S(q,\omega) = \sum_n |\langle n|\hat{\rho}_q|0\rangle|^2 \delta(\omega_{n0} - \omega)$$

are reported in table 1. In (2) $|0\rangle$ and $|n\rangle$ are the ground and excited states of the system and

$$\hat{\rho}_q = \sum_i \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i}$$

is the electron density operator.

In the following we will investigate the dispersion of the plasmon in alkali metals with the help of the inverse-energy-weighted sum rule m_{-1} and of the energy-weighted sum rule m_1. With the help of table 1, we can write (up to terms in q^4):

$$m_{-1} = m_{-1}^{\rm pl} + m_{-1}^{\rm sp} + m_{-1}^{\rm mp} \qquad m_1 = m_1^{\rm pl} + m_1^{\rm mp}. \tag{3}$$

Since multipair excitations (mp) are expected to be located at high energies with respect to the plasmon and single-pair excitations, for a first analysis we will neglect their contribution to the inverse-energy-weighted sum rule

$$m_{-1} = \int S(q,\omega)\omega^{-1}\,\mathrm{d}\omega$$

Table 1. Matrix elements, excitation frequencies, and sum rule contributions of density excitations in the long wavelength limit.

	Single-pair	Plasmon	Multipairs
$\sum_n \lvert(\hat{\rho}_q)_{n0}\rvert^1$	q^5	$\frac{Nq^2}{2m\omega_p(0)}$	q^4
ω_{n0}	qv_F	$\omega_p(0)$	ω^{mp}
m_{-1}	$\frac{4}{15}\frac{\epsilon_F N}{m^2\omega_p^4(0)}q^4$	$\frac{Nq^2}{2m\omega_p(0)^2}$	q^4
m_1	q^6	$\frac{Nq^2}{2m}$	q^4
m_3	q^8	$\frac{N\omega_p^2(0)q^2}{2m}$	q^4

where the high-energy part of the excitation spectrum is quenched by the $1/\omega$ factor. Vice versa multipair effects will be explicitly considered in the energy-weighted sum rule

$$m_1 = \int S(q,\omega)\omega\,\mathrm{d}\omega.$$

The inverse-energy-weighted moment m_{-1} is given [5], up to terms in q^4, by

$$m_{-1} = \frac{1}{2}\frac{q^2 N}{m\omega_p^2(0)}\left(1 - \frac{s^2 q^2}{\omega_p^2(0)}\right) \tag{4}$$

where $1/ms^2$ is the compressibility of the system.

An important point of the present analysis is that the single-pair (sp) contribution to m_{-1} can be exactly evaluated. The result is

$$m_{-1}^{sp} = \frac{4}{15}\frac{\epsilon_F N}{m^2\omega_p^4(0)}q^4 \tag{5}$$

where ϵ_F is the Fermi energy. Result (5) follows from the proper application of Landau's theory to the calculation of the low q, ω component of the screened response function. In fact one can show that in this regime, the imaginary part of the dynamic response function obeys the relation

$$\mathrm{Im}\chi(q,\omega)_{q,\omega\to 0} = \frac{q^4}{(4\pi e^2)^2}\mathrm{Im}\frac{1}{\chi_{sc}(q,\omega)} \tag{6}$$

where χ_{sc} is the usual screened response function. Since in the low q,ω limit, one can safely use the equation $1/\chi_{sc}(q,\omega) = 1/\chi_0(q,\omega) + F_0$ holding in Landau's theory (we have set here $F_\ell = 0$ for $\ell \geqslant 2$), one finally gets the useful result

$$\mathrm{Im}\chi(q,\omega)_{q,\omega\to 0} = \frac{q^4}{(4\pi e^2)^2}\mathrm{Im}\frac{1}{\chi_0(q,\omega)}\ , \tag{7}$$

where $\chi_0(q,\omega)$ is the free electron gas response function fixed by the Fermi energy $\epsilon_F = q_F^2/2m^*$. It is worth noting that the Landau parameter F_0 does not enter in the low q, ω behaviour of $\mathrm{Im}\chi(q,\omega)$. From (7) and recalling that $S(q,\omega) = -\frac{1}{\pi}\mathrm{Im}\chi$, one derives the single particle contribution (5) to m_{-1}.

Let us now discuss the energy-weighted moment m_1. This moment is model independent and given by the famous f-sum rule:

$$m_1 = \int S(q,\omega)\omega\,\mathrm{d}\omega = \frac{Nq^2}{2m}. \tag{8}$$

Looking at (3), one concludes that the q^4 correction due to the plasmon and multipair excitation must cancel exactly. The calculation of the plasmon contribution to the m_1 sum rule requires a non-trivial many-body approach. We propose a first estimate of this contribution in the framework of the Brueckner–Hartree–Fock theory. This theory has been so far developed in the local approximation [6]. This is not enough to obtain the contribution to the f-sum rule which originates from explicit non-local and finite-range terms in the effective interaction. In the Brueckner–Hartree–Fock approach, the plasmon contribution to m_1 can be calculated, at low q, through:

$$m_1^{\mathrm{pl}} = \tfrac{1}{2}\langle HF|[\hat\rho_{-q},[H_{\mathrm{eff}},\hat\rho_q]]|HF\rangle \tag{9}$$

where the non-local effective Hamiltonian to be used in (9) has been recently derived in [7] and has the form

$$\begin{aligned} H_{\mathrm{eff}} = {} & \sum_{p\sigma}\frac{p^2}{2m}a^\dagger_{p\sigma}a_{p\sigma} + \frac{1}{2}\sum_{p_1p_2q\sigma\sigma'} v(q)a^\dagger_{p_1+q\sigma}a^\dagger_{p_2-q\sigma'}a_{p_2\sigma'}a_{p_1\sigma} \\ & + \frac{1}{2}\sum_{p_1p_2q\sigma\sigma'} C\left(\frac{1}{2}(p_1-p_2+q)\right)V(q)a^\dagger_{p_1+q\sigma}a^\dagger_{p_2-q\sigma'}a_{p_2\sigma'}a_{p_1\sigma}. \end{aligned} \tag{10}$$

In (10) $v(q) = 4\pi/q^2$ is the Coulomb interaction and $C(\frac{1}{2}(p_1-p_2+q))V(q)$ is the non-local g-matrix correction obtained by solving the Bethe–Goldstone equations [7]. From (9) and (10) one gets

$$m_1^{\mathrm{pl}} = \frac{Nq^2}{2m} - K\frac{Nq^4}{2m} \tag{11a}$$

with

$$K = -\frac{3}{16r_s^3}\left[I_4\int\left[j_1\left(\frac{k_{\mathrm{F}}r}{2}\right)\Big/\frac{k_{\mathrm{F}}r}{2}\right]^2 C(r)r^4\,\mathrm{d}r - \frac{1}{2}C_4\int[j_1(k_{\mathrm{F}}r)/k_{\mathrm{F}}r]^2V(r)r^4\,\mathrm{d}r\right]. \tag{11b}$$

In the above equation $C(r)$ and $V(r)$ are the Fourier transform of $C(P)$ and $V(q)$ of (10), and

$$I_{\mathrm{p}} = 4\pi\int r^pV(r)\,\mathrm{d}r \qquad C_{\mathrm{p}} = 4\pi\int r^pC(r)\,\mathrm{d}r.$$

The values of the coefficient K of the q^4 correction in (11) are reported in table 2 for different r_s.

Equations (5) and (11) are the main results of the present work. In particular result (11) permits us to go beyond the usual mean field theories, such as the time dependent local

Table 2. Values of the coefficient K and of the mean multipair excitation energy ω^{mp} calculated using equations (11) and (16). The values of $\omega_{\mathrm{p}}(0)$ and ϵ_{F} are also shown. For $\frac{1}{2}ms^2$ and E_{kin} and E_{pot} we have used the Monte Carlo values of [10]. The various quantities are in atomic units.

r_s	ϵ_{F}	$\omega_{\mathrm{p}}(0)$	K	ω^{mp}
2	0.460	0.613	0.05	1.52
3	0.205	0.333	0.3	0.68
4	0.115	0.217	0.5	0.48
5	0.074	0.155	1.0	0.30
6	0.051	0.118	2.0	0.20

density approximation (TDLDA), where $K = 0$ and explicitly shows that the f-sum rule is not exhausted by the plasmon mode to the order q^4.

We are now ready to calculate the energy of the plasmon excitation through the equation

$$\omega_{\mathrm{p}}(q) = \sqrt{\frac{m_1^{\mathrm{pl}}}{m_{-1}^{\mathrm{pl}}}} = \omega_{\mathrm{p}}(0)\left[1 + \frac{1}{2}\frac{q^2}{m\omega_{\mathrm{p}}^2(0)}\left(ms^2 + \frac{8}{15}\epsilon_{\mathrm{F}} - Km\omega_{\mathrm{p}}^2(0)\right)\right] \tag{12}$$

where we have used (3)–(5) and (11). From (12) we get the expression

$$\frac{\alpha}{\alpha_{\mathrm{RPA}}} = \frac{5}{3\epsilon_{\mathrm{F}}}\left(\frac{1}{2}ms^2 + \frac{4}{15}\epsilon_{\mathrm{F}} - \frac{1}{2}Km\omega_{\mathrm{p}}^2(0)\right) \tag{13}$$

for the dispersion coefficient normalized to the RPA value $\alpha^{\mathrm{RPA}} = \frac{3}{5}\epsilon_{\mathrm{F}}/\omega_{\mathrm{p}}(0)$.

In figure 1 we report the predictions of (13). For the effective mass entering the Fermi energy, we have taken the free electron mass. We have checked that the predictions for $\alpha/\alpha_{\mathrm{RPA}}$ do not significantly depend on the value of the effective mass. Equation (13) accounts rather well for the general trend indicated by experiments. A crucial role is played by the term in K originating from the q^4 plasmon contribution to the f-sum rule. In the absence of such a term ($K = 0$) one would find rather poor predictions (dotted line), very close to the results of TDLDA calculations [2,3].

Our results also permit us to estimate the average energy of multipair excitations through the ratio $\sqrt{m_3^{\mathrm{mp}}/m_1^{\mathrm{mp}}}$, where m_3^{mp} is the multipair contribution to the cubic energy-weighted sum rule

$$\int S(q,\omega)\omega^3\,\mathrm{d}\omega.$$

This quantity can be calculated using the result (see table 1)

$$m_3^{\mathrm{mp}} = m_3 - m_1^{\mathrm{pl}}\omega_{\mathrm{p}}^2(q) \tag{14}$$

holding at the order q^4, where

$$m_3 = \int \omega^3 S(q,\omega)\,\mathrm{d}\omega$$

is the cubic energy-weighted sum rule given by [8,9]

$$m_3 = \frac{Nq^2}{2m}\left[\omega_{\mathrm{p}}^2(0) + \frac{2q^2}{m}\left(E_{\mathrm{kin}} + \frac{2}{15}E_{\mathrm{pot}}\right)\right] \tag{15}$$

and the second term in the RHS of (14) is the plasmon contribution. In (15) $E_{\rm kin}$ and

$$E_{\rm pot} = \frac{1}{2} n \int {\rm d}r\, \frac{e^2}{r}(g(r) - 1)$$

are the kinetic and potential energy per particle respectively. Using equations (11)–(15), we finally obtain the following expression for the average energy of multipair excitations

$$\omega^{\rm mp} = \sqrt{m_3^{\rm mp}/m_1^{\rm mp}} = \sqrt{(2/mK)[(E_{\rm kin} + \tfrac{2}{15}E_{\rm pot}) - (\tfrac{1}{2}ms^2 + \tfrac{4}{15}\epsilon_{\rm F} - Km\omega_{\rm p}^2(0))]}. \qquad (16)$$

The predicted values for $\omega^{\rm mp}$ are reported in table 2. As expected $\omega^{\rm mp}$ is systematically larger than $\omega^{\rm pl}$ and $\epsilon_{\rm F}$.

In conclusion we have calculated the plasmon contribution to the inverse-energy-weighted and energy-weighted sum rules. We have shown that the contribution of short-range Brueckner correlations to the f-sum rule is sizable and permits us to explain in a natural way the strong density dependence exhibited by the experimental data for the dispersion coefficient α.

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