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Excitations in the electron liquid

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Starting from Mori's formalism, an expression for the dynamical structure factor for the electron liquid is derived. The memory function is calculated such that no phenomenological collision rates have to be introduced. Using simple approximations for some static correlation functions, good agreement with inelastic x-ray experiments is obtained.

I. INTRODUCTION

Recently, some attention has been focused on experimental results for simple metals, obtained by inelastic x-ray[1-3] and electron[4,5] scattering experiments. In these experiments, the dynamical structure factor $S(q, \omega)$ is measured directly. In the low-momentum-transfer region, a plasmon-like excitation is observed, while for high momentum transfers particle-hole excitations dominate the spectra entirely. For intermediate-wave-vector transfers both excitations are visible. Since the spectra for different metals, such as Li, Al, and Be, are very similar, the observed features should be general properties of the solid-state electron gas.[1]

From a theoretical point of view, these results are not well understood because the experimental results strongly suggest the breakdown of the generalized random-phase approximation (RPA) theories.[1,6,7] A modification of the theory of Ref. 7 was proposed[8] but as the modified theory violates the continuity equation,[8] the rather good agreement with some experimental data should not be regarded as a proof of the usefulness of this modification.

More recent attempts[9,10] to calculate the dynamical structure factor for an electron liquid are based on Mori's formalism.[11] The most important reason to use this formalism is the fact that the finite lifetime of the excitations is introduced in a way such that the continuity equation is not violated and that a number of frequency sum rules are satisfied exactly.[10] In this approach, the original problem of calculating $S(q, \omega)$ is replaced by the equivalent problem of evaluating the memory function. This function is formally the same as $S(q, \omega)$ but the time evolution entering the memory function describes fast decaying processes only and the main problem is then to find a way to approximate the memory function. In Ref. 9, the memory function was first approximated by its free-particle part. In the context of Mori's formalism, this is a strange starting point for an approximation because free-particle excitations have infinite lifetimes while the memory function should account for fast-decaying processes. By introducing a phenomenological collision rate, the memory function was "renormalized" and then the collision rate was determined such that the best possible agreement between theory and experiment was obtained. Nevertheless, the theoretical results could not explain the experimental facts. In Ref. 10 the effect of various modifications of the theory, given in Ref. 7, were studied extensively and the most important conclusion of this approach was that the final result depends on the way the lifetime effects were taken into account. Obviously, RPA-based theories are inconvenient to describe processes where lifetime effects are important and perturbation theory is not valid.[6]

As already mentioned, Mori's formalism is more appropriate to calculate the dynamical structure factor because it is not restricted to weak interacting systems and because the motion of the density operator can be separated from fast fluctuating processes. These fast decaying forces are described by the memory function. Using a method first given and applied to one-dimensional spin systems by the present authors,[12] we calculate the memory function as consistently as possible. The result of this calculation is an expression for the dynamical structure factor as a function of the frequency and some well defined wave-vector-dependent static correlation functions. Using plausible approximations for these static quantities we obtain reasonable agreement with experimental results.

The plan of the paper is as follows. In Sec. II, we discuss the choice of the slow variables and we apply the general theory[12] to the electron liquid

Hamiltonian. In Sec. III, simple approximations for the static quantities appearing in the expression for $S(q, \omega)$ are proposed and comparison with experiments is made. The conclusions of this work are summarized in Sec. IV.

II. DYNAMIC EQUATIONS

The Hamiltonian for an electron liquid with uniform positive background is given by

$$H = \sum_{\vec{k}, s} \epsilon_{\vec{k}} c^{\dagger}_{\vec{k}, s} c_{\vec{k}, s} + \sum_{\substack{\vec{p}, \vec{k}, \vec{q} \\ s, r}} v_{\vec{q}} c^{\dagger}_{\vec{k}+\vec{q}, s} c^{\dagger}_{\vec{p}, r} c_{\vec{p}+\vec{q}, r} c_{\vec{k}, s} , \qquad (2.1)$$

where $v_{\vec{q}}$ is the Fourier transform of the Coulomb potential, including the interaction with the positive background, and $\epsilon_{\vec{k}}$ denotes the kinetic energy of an electron with momentum $\vec{k}$:

$$v_{\vec{q}} = (4\pi e^2/q^2)(1 - \delta_{\vec{q}, 0}) , \qquad (2.2a)$$

$$\epsilon_{\vec{k}} = k^2/2m . \qquad (2.2b)$$

Here and in the following we use units such that $\hbar = 1$. The Fourier transformed density operator is defined by

$$\rho_{\vec{q}} = \sum_{\vec{k}, s} c^{\dagger}_{\vec{k}, s} c_{\vec{k}+\vec{q}, s} . \qquad (2.3)$$

The dynamical structure factor, which can be measured by inelastic x-ray and electron scattering experiments is related to the imaginary part of Kubo's relaxation function[13] by

$$S_{\rho\rho}(\vec{q}, \omega) = [-\omega/(1 - e^{-\beta\omega})]\, \Phi''_{\rho\rho}(\vec{q}, \omega) , \qquad (2.4)$$

where

$$\Phi''_{\rho\rho}(\vec{q}, \omega) = \lim_{\epsilon \to 0} \operatorname{Im} \Phi_{\rho\rho}(\vec{q}, z);$$

$$z = \omega + i\epsilon, \quad \epsilon > 0.$$

Defining a scalar product of two operators by the static susceptibility[11,13]

$$\chi_{AB} = (A, B) = \int_0^{\beta} \langle A^{\dagger} e^{-\lambda H} B e^{\lambda H} \rangle \, d\lambda , \qquad (2.5)$$

the Laplace-transformed relaxation function can be written as[13]

$$\Phi_{\rho\rho}(\vec{q}, z) = (\rho, (z - L)^{-1} \rho)_{\vec{q}} . \qquad (2.6)$$

As usual, $\langle A \rangle$ denotes the thermal average of the operator A. By rotational invariance, we have $(A, B)_{\vec{q}} = (A, B)_q$. The Liouville operator L determines the time evolution in the space of operators and is related to the Hamiltonian by

$$LA = [H, A] \quad \text{or} \quad A(t) = e^{iLt} A(0) = e^{iHt} A(0) e^{-iHt} . \qquad (2.7)$$

If we want to use the memory function formalism, we first have to know which operators belong to the subspace of slow-varying quantities. For the electron liquid, the total density is a conserved quantity. Therefore, the Fourier-transformed density $\rho_{\vec{q}}$ is a slow variable, and consequently all time derivatives are slow variables too. This means that the subspace E of slow variables is given by

$$E = \{\rho_{\vec{q}}, L\rho_{\vec{q}}, L^2\rho_{\vec{q}}, \ldots\} . \qquad (2.8)$$

It is obvious that this set is rather large for practical purposes and therefore we will confine ourselves by taking

$$E = \{\rho_{\vec{q}}, L\rho_{\vec{q}}, L^2\rho_{\vec{q}}\} , \qquad (2.9)$$

as the set of relevant variables. Since $L\rho_{\vec{q}} = \vec{q} \cdot \vec{J}_{\vec{q}}$ ($\vec{J}_{\vec{q}}$ is the current density) and $L^2\rho_{\vec{q}}$ is related to the energy density by the continuity equations, the three operators appearing in Eq. (2.9) have a well-defined physical meaning. It is interesting to note that in the former attempts[9,10] to use Mori's formalism, only the density and the longitudinal current density were taken as relevant variables. A discussion of the consequences of this limitation will be given at the end of this section.

For practical purposes, it is convenient to orthogonalize the operators of E with respect to the scalar product Eq. (2.5) and therefore we replace $L^2\rho_{\vec{q}}$ by

$$A_{\vec{q}} = L^2\rho_{\vec{q}} - \rho_{\vec{q}} (L\rho, L\rho)_q/(\rho, \rho)_q . \qquad (2.10)$$

Following Mori,[11] the matrix of Laplace-transformed relaxation functions

$$\tilde{\Phi}(q, z) = \begin{bmatrix} \Phi_{\rho\rho}(q, z) & \Phi_{\rho L\rho}(q, z) & \Phi_{\rho A}(q, z) \\ \Phi_{L\rho\rho}(q, z) & \Phi_{L\rho L\rho}(q, z) & \Phi_{L\rho A}(q, z) \\ \Phi_{A\rho}(q, z) & \Phi_{AL\rho}(q, z) & \Phi_{AA}(q, z) \end{bmatrix} , \qquad (2.11)$$

is the solution of the matrix equation

$$[z\tilde{1} - \tilde{\Omega}(q) + \tilde{\Sigma}(q, z)]\, \tilde{\Phi}(q, z) = \tilde{\chi}(q) . \qquad (2.12)$$

As a consequence of the orthogonalization, the matrix of static susceptibilities

$$\tilde{\chi}(q)=\begin{bmatrix}(\rho,\rho)_q & 0 & 0\\ 0 & (L\rho,L\rho)_q & 0\\ 0 & 0 & (A,A)_q\end{bmatrix}, \tag{2.13}$$

is diagonal. The frequency matrix is given by

$$\tilde{\Omega}(q)=\begin{bmatrix}0 & 1 & 0\\ \dfrac{(L\rho,L\rho)_q}{(\rho,\rho)_q} & 0 & 1\\ 0 & \dfrac{(A,A)_q}{(L\rho,L\rho)_q} & 0\end{bmatrix}, \tag{2.14}$$

while the projector on the space, spanned by the relevant variables is given by

$$PX_{\vec{q}}=\frac{\rho_{\vec{q}}(\rho,X)_q}{(\rho,\rho)_q}+\frac{L\rho_{\vec{q}}(L\rho,X)_q}{(L\rho,L\rho)_q}+\frac{A_{\vec{q}}(A,X)_q}{(A,A)_q}\ . \tag{2.15}$$

The projector on the space of fast decaying quantities is given by $Q\equiv 1-P$. Since $QL\rho_q=QL^2\rho_q=0$, the matrix of memory functions $\tilde{\Sigma}(q,z)$ contains but a single nonzero element and we have

$$\tilde{\Sigma}(q,z)=\begin{bmatrix}0 & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & \Sigma(q,z)\end{bmatrix}, \tag{2.16}$$

where $\Sigma(q,z)=-(QLA,(z-QLQ)^{-1}QLA)_q/(A,A)_q$. Comparing this expression with Eq. (2.6) we conclude that these functions are essentially the same, but the time evolution entering $\Sigma(q,z)$ is given by QLQ instead of L. By construction, QLQ describes fast-decaying processes. Then the simplest approximation one can think of is to replace $\Sigma(q,z)$ by a constant. Unfortunately, a calculation of this parameter is only possible if it is allowed to use perturbation theory.[14] In a previous paper,[12] the present authors suggested a simple method to circumvent this problem and we will use this approach here.

Denoting $f(q,z)=(QLA,(z-QLQ)^{-1}QLA)_q$ and applying the operator identity $z(z-QLQ)^{-1}=1+QLQ(z-QLQ)^{-1}$ twice, we obtain the equation of motion for the memory function,

$$[z^2+(LA,LA)_q/(A,A)_q]f(q,z)$$
$$=z(QLA,QLA)_q+(QL^3A,(z-QLQ)^{-1}QLA)_q\ . \tag{2.17}$$

Since the last term of the right-hand side of Eq. (2.17) contains higher derivatives than all other terms, it is reasonable to replace it by a complex constant R. Taking the limit $z\to 0$ we have

$$[(LA,LA)_q/(A,A)_q]f(q,0)=R,$$
$$f(q,0)=\lim_{\epsilon\to 0}f(q,i\epsilon)\ , \tag{2.18}$$

and the final equation for $f(q,z)$ reads

$$[z^2+(LA,LA)_q/(A,A)_q]f(q,z)$$
$$=z(QLA,QLA)_q+[(LA,LA)_q/(A,A)_q]f(q,0)\ . \tag{2.19}$$

By symmetry $f(q,0)$ is pure imaginary. Using the sum rule[12]

$$(QLA,QLA)_q=-\frac{1}{\pi}\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty}d\omega\,\mathrm{Im}f(q,z)\ , \tag{2.20}$$

we can calculate $f(q,0)$ and the resulting expression for the memory function is

$$\Sigma(q,z)=-\frac{(QLA,QLA)_q}{(A,A)_q}\,\frac{1}{z+i\,(LA,LA)_q^{1/2}/(A,A)_q^{1/2}}\ . \tag{2.21}$$

Note that in this approach no phenomenological collision rates have to be introduced. Using Eq. (2.12), the solution for $\Phi_{\rho\rho}(q,z)$ is easily obtained and we have

$$\Phi_{\rho\rho}(q,z)=\frac{(\rho,\rho)_q[z^2+z\Sigma(q,z)-(A,A)_q/(L\rho,L\rho)_q]}{z[z^2-(L^2\rho,L^2\rho)_q/(L\rho,L\rho)_q]+\Sigma(q,z)[z^2-(L\rho,L\rho)_q/(\rho,\rho)_q]}\ . \tag{2.22}$$

In order to show that the memory function has been calculated as consistently as possible we expand Eq. (2.22) for large z and we find

$$\Phi_{\rho\rho}(q,z)=\frac{(\rho,\rho)_q}{z}+\frac{(L\rho,L\rho)_q}{z^3}$$
$$+\frac{(L^2\rho,L^2\rho)_q}{z^5}+\frac{(L^3\rho,L^3\rho)_q}{z^7}+\cdots\ . \tag{2.23}$$

This means that our expression for $\Phi_{\rho\rho}(q,z)$ exactly fulfills the sum rules

$$(L^n\rho,L^n\rho)_q=-\frac{1}{\pi}\int_{-\infty}^{+\infty}\omega^{2n}\Phi''_{\rho\rho}(q,\omega)\,d\omega,$$
$$n=0,1,2,3\ . \tag{2.24}$$

The first three sum rules are automatically fulfilled by the choice of our variables. The sum rule for $n=3$, however, is implied by the sum rule on the memory function Eq. (2.20).

In order to show that we can recover the results obtained by Mukhopadyay and Sjölander,[10] we take $\{\rho_{\vec{q}}, L\rho_{\vec{q}}\}$ as the set of relevant variables and we repeat the steps given above. Then we find[12]

$$\Phi_{\rho\rho}(q,z)=\frac{(\rho,\rho)_q[z+\hat{\Sigma}(q,z)]}{z^2+z\hat{\Sigma}(q,z)-(L\rho,L\rho)_q/(\rho,\rho)_q}\ , \tag{2.25}$$

where

$$\hat{\Sigma}(q,z)=-\frac{(QL^2\rho,QL^2\rho)_q}{(L\rho,\ L\rho)_q}\,\frac{1}{z+i\tau^{-1}}\ ,$$

with

$$\tau^{-1}=(L^2\rho,L^2\rho)_q^{1/2}/(L\rho,L\rho)_q^{1/2}\ .$$

The frequency dependence of this relaxation function is exactly the same as the one of Ref. 10, with choice IV for the memory function. The only difference between both relaxation functions is that Mukhopadyay and Sjölander assumed that τ^{-1} is proportional to the plasmon frequency ω_p, while in our case it is not.

The relaxation function given by Eq. (2.25) has three poles in the lower half of the complex z plane. One pole is pure imaginary and the other two are symmetric with respect to the imaginary axes. Therefore this relaxation function can never give a line shape with two peaks at positive energies. This simple but strong argument justifies our particular choice of the variables Eq. (2.9), because the resulting relaxation function Eq. (2.22) is a four-pole approximation.

III. APPROXIMATIONS

The remaining problem is the evaluation of the static quantities appearing in the expression for the relaxation function [see Eq. (2.22)]. Although we have formulated the dynamical theory for arbitrary temperatures, we will restrict ourselves to the zero-temperature limit. This is a good approximation for the electron liquid because the Fermi energy is of the order of electron volts.

Instead of using the results of laborious numerical calculations, we will make simple but plausible approximations for the static quantities, because it turns out that even then, good qualitative agreement with experimental results is obtained.

For the static susceptibility, we use the well-known expression[6,7,15]

$$(\rho,\rho)_q=\frac{-\chi^0(q)}{1-v(q)[1-G(q)]\chi^0(q)}\ , \tag{3.1}$$

where $\chi^0(q)$ denotes the usual Lindhard response function for the noninteracting electron gas. For small wave vectors, the local field factor $G(q)$ can be written as[7,10] $G(q)\approx\gamma q^2$ while for large wave vectors one finds[15] $\frac{1}{3}\leq G(q)\leq\frac{2}{3}$.

The calculation of the susceptibilities $(L\rho,L\rho)_q$, $(L^2\rho,L^2\rho)_q$, and $(L^3\rho,L^3\rho)_q$ is straightforward and the results are summarized in Appendix A. In the following, ω_p denotes the plasma frequency of the noninteracting system in units of the Fermi energy ϵ_F and q stands for the wave vector in units of the Fermi wave vector k_F.

We first consider the region of small wave-vector transfers ($q\to 0$). Restricting ourselves to terms of order q^2 we find

$$\frac{(L\rho,L\rho)_q}{\epsilon_F^2(\rho,\rho)_q}=\omega_p^2[1+q^2(\tfrac{4}{3}\omega_p^{-2}-\gamma)]\ , \tag{3.2a}$$

$$\frac{(L^2\rho,L^2\rho)_q}{\epsilon_F^2(L\rho,L\rho)_q}=\omega_p^2[1+(\tfrac{12}{5}\omega_p^{-2}-\tfrac{2}{5}\bar{\gamma})q^2]\ ,$$

$$\bar{\gamma}=-\frac{1}{2}\int_0^\infty dk\,[S(k)-1]\ , \tag{3.2b}$$

$$\frac{(L^3\rho,L^3\rho)_q}{\epsilon_F^4(L\rho,L\rho)_q}=\tfrac{48}{5}\,\omega_p^2[1-\tfrac{1}{2}g(0)]q^2+\omega_p^4(1-\tfrac{13}{5}\bar{\gamma}q^2)\ . \tag{3.2c}$$

Substituting these expressions in Eq. (2.21) we conclude that the memory function is independent of q:

$$\Sigma(z,q)=\omega_p^2a/(z+i\omega_pa^{1/2})\ ,\quad q\to 0\ , \tag{3.3}$$

where

$$a=\frac{24[1-g(0)]-9\bar{\gamma}\omega_p^2}{\frac{16}{3}+\omega_p^2(5\gamma-2\bar{\gamma})}\ .$$

Thus the inverse collision time $\omega_pa^{1/2}$ is roughly proportional to ω_p.[10] Neglecting the damping for a moment, Eq. (2.22) reduces to[9]

$$\frac{\Phi_{\rho\rho}(q,z)}{(\rho,\rho)_q}=\frac{z}{z^2-\omega_0^2(q)}\ ,\quad q\to 0\ , \tag{3.4}$$

where $\omega_0(q)\approx\omega_p[1+q^2(6/5\omega_p^2-\frac{1}{5}\bar{\gamma})]$.

For large wave vectors ($q\to\infty$) we have

$$(L\rho,L\rho)_q/\epsilon_F^2(\rho,\rho)_q=q^4-\tfrac{4}{5}q^2\ . \tag{3.5a}$$

$$(L^2\rho,L^2\rho)_q/\epsilon_F^2(L\rho,L\rho)_q=q^4+\tfrac{12}{5}q^2\ , \tag{3.5b}$$

$$(L^3\rho,L^3\rho)_q/\epsilon_F^4(L\rho,L\rho)_q=q^8+8q^6\ . \tag{3.5c}$$

Then the memory function reads

$$\Sigma(q,z) = -q^4/(z+iq^2)\,. \tag{3.6}$$

Neglecting the damping, Eq. (2.22) becomes

$$\frac{\Phi_{\rho\rho}(q,z)}{(\rho,\rho)_q} = \frac{z}{[z^2-(q^4+\frac{12}{5}q^2)]}\,, \tag{3.7}$$

which is just the relaxation function for the noninteracting system.[6] Clearly, our dynamical description shows the expected behavior for small and large wave vectors, and is able to give expressions for the damping of the excitations in terms of static quantities which can be calculated.[4,7,10]

In order to compare the theory with experiments, we have plotted $S(k,\omega) = -\omega\,\mathrm{Im}\Phi_{\rho\rho}(k,\omega)$ for various wave vectors and materials. For $G(q)$ we take[6]

$$G(q) = Gq^2/(1+q^2)\,, \quad 0 \leq G \leq \tfrac{2}{3} \tag{3.8}$$

and since $S(q)$ is not known, the simplest thing we we can do is to replace the terms containing $S(q)$ by adjustable parameters. Furthermore, we assume that the pair correlation function in the origin is zero $[g(0)\approx 0]$.[7] Then we have

$$\frac{(L^2\rho,L^2\rho)_q}{\epsilon_F^2(L\rho,L\rho)_q} = q^4+\tfrac{12}{5}q^2+\omega_p^2(1-A)\,, \quad 0 \leq A \leq \tfrac{2}{3}\,, \tag{3.9a}$$

$$\frac{(L^3\rho,L^3\rho)_q}{\epsilon_F^4(L\rho,L\rho)_q} = q^8+8q^6+\tfrac{48}{7}q^4 + 4\omega_p^2(q^4+\tfrac{12}{5}q^2)+\omega_p^4(1-B)\,. \tag{3.9b}$$

By varying the three parameters G, A, and B, it is not difficult to obtain the observed line shape for a particular wave vector. Therefore, our description is only useful if agreement with experiments can be obtained for various wave vectors and materials without changing G, A, or B. By taking $G=\frac{2}{3}$ and $A=0.2$, it is clear from Figs. 1 and 2 that good agreement can be obtained for $1 \leq q \leq 1.6$. Because it turns out that the line shapes are insensitive for modest changes in B, we have taken $B=0$ in all cases. For large wave vectors we take the values for G and A corresponding to the infinite wave vectors limit and we use $G=\frac{2}{3}$ and $A=\frac{2}{3}$. From Fig. 3, we again conclude that there is good agreement.

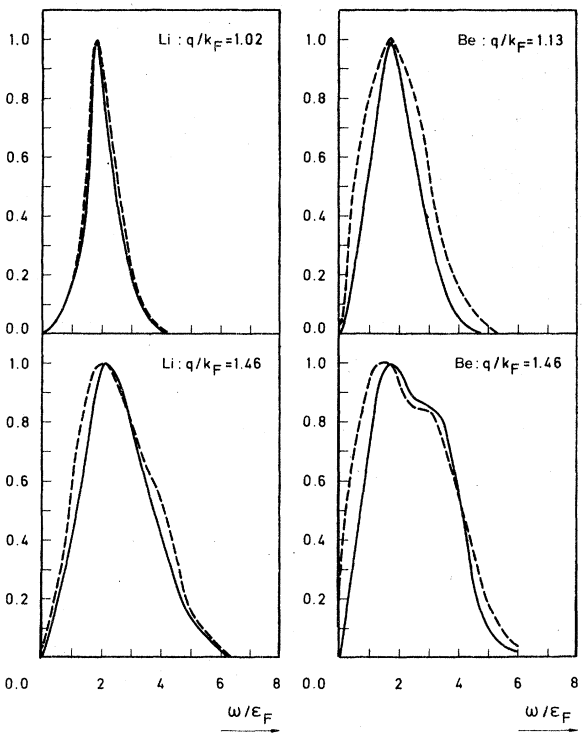

FIG. 1. Comparison between the experimental (dashed line) and theoretical dynamical structure factor for Li and Be.

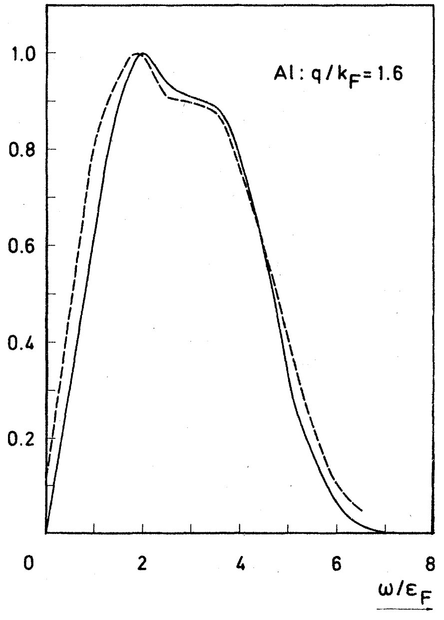

FIG. 2. Experimental and theoretical line shape for Al.

IV. CONCLUSIONS

We have presented a dynamical theory for density excitations in an electron liquid, which can account for the experimentally observed features. Instead of modifying existing theories, we used Mori's formalism in order to obtain an expression for the dynamical structure factor in terms of static sus-

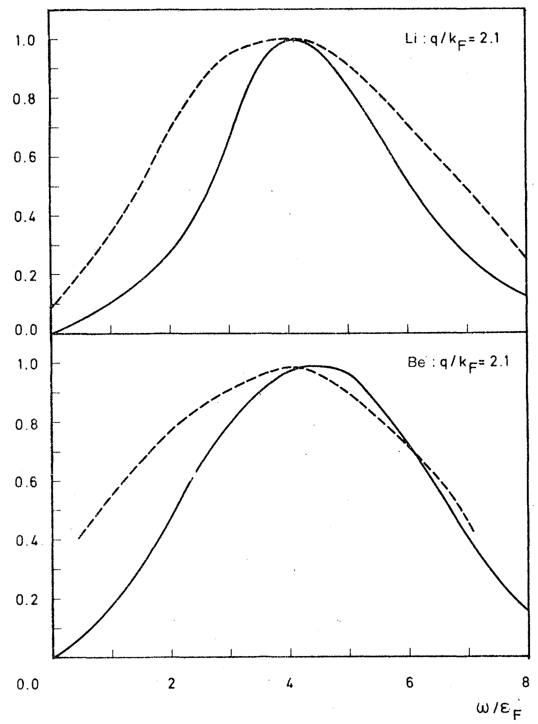

FIG. 3. Line shapes for Be and Li in the large-momentum-transfer region.

ceptibilities and a memory function. This function has been approximated as consistently as possible. The final relaxation function only depends on the frequency and some static correlation functions, and exactly fulfills four frequency sum rules. No phenomenological collision rate has to be introduced. Using simple approximations for the static quantities, good agreement between experiment and theory is obtained.

In our opinion, the dynamical theory given in this paper can be useful if one wants to extract information about static quantities, such as the kinetic energy and the static susceptibility, from experimental data.

By convoluting the experimental resolution function with the expression for the dynamical structure factor, a direct comparison with the experimental data is possible. Then the static quantities appearing in the theoretical dynamical structure factor can be adjusted until good agreement with experimental data is found.

ACKNOWLEDGMENTS

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APPENDIX A

Using the relation

$$(A, LB) = \int_0^\beta \langle A^\dagger e^{-\lambda L} LB\rangle\, d\lambda$$

$$= -\int_0^\beta \frac{d}{d\lambda}\langle A^\dagger e^{-\lambda L} B\rangle\, d\lambda$$

$$= \langle A^\dagger B\rangle - \langle A^\dagger e^{-\beta L} B\rangle = \langle [A^\dagger, B]\rangle\,, \tag{A1}$$

we can write $(L\rho, L\rho)_q$, $(L^2\rho, L^2\rho)_q$, and $(L^3\rho, L^3\rho)_q$ in terms of static correlation functions. In the following, we assume that $T=0$, so that the expectation values must be evaluated in the ground state. The exact expressions for the static susceptibilities now read

$$(L\rho, L\rho)_q = nq^2/m\,, \tag{A2}$$

$$\begin{aligned}(L^2\rho, L^2\rho)_q = {} & 2\sum_{\vec{k}} (\epsilon_{\vec{k}+\vec{q}} - \epsilon_{\vec{k}})^3 \langle c^\dagger_{\vec{k}} c_{\vec{k}}\rangle \\ & + \frac{1}{m^2}\sum_{\vec{m}\vec{n}\vec{k}} [(\vec{k}\cdot\vec{q} + q^2)^2 v_{\vec{k}+\vec{q}} - (\vec{k}\cdot\vec{q})^2 v_{\vec{k}}] \\ & \times \langle c^\dagger_{\vec{m}+\vec{k}} c^\dagger_{\vec{n}} c_{\vec{n}+\vec{k}} c_{\vec{m}}\rangle\,,\end{aligned} \tag{A3}$$

$$\begin{aligned}(L^3\rho, L^3\rho)_q = {} & 2\sum_{\vec{k}} (\epsilon_{\vec{k}+\vec{q}} - \epsilon_{\vec{k}})^5 \langle c^\dagger_{\vec{k}} c_{\vec{k}}\rangle \\ & + \frac{2}{m^2}\sum_{\vec{m}\vec{n}\vec{k}} \{(\epsilon_{\vec{m}} - \epsilon_{\vec{m}+\vec{q}})^2 [\vec{k}\cdot\vec{q} v_{\vec{k}} (2q^2 + 4\vec{m}\cdot\vec{q} + \vec{k}\cdot\vec{q} + \vec{n}\cdot\vec{k} - \vec{m}\cdot\vec{k}) \\ & \qquad - (\vec{k}-\vec{q})\cdot\vec{q} v_{\vec{k}-\vec{q}} (q^2 + 2\vec{n}\cdot\vec{q} + 2\vec{m}\cdot\vec{q} + \vec{k}\cdot\vec{q} + \vec{n}\cdot\vec{k} - \vec{m}\cdot\vec{k})] \langle c^\dagger_{\vec{m}+\vec{k}} c^\dagger_{\vec{n}} c_{\vec{n}+\vec{k}} c_{\vec{m}}\rangle\} \\ & + \frac{1}{m^3}\sum_{\vec{p}\vec{k}} \{(\vec{k}+\vec{q})\cdot(\vec{p}+\vec{q}) [\vec{k}\cdot\vec{q} v_{\vec{k}} - (\vec{k}+\vec{q})\cdot\vec{q} v_{\vec{k}+\vec{q}}] [\vec{p}\cdot\vec{q} v_p - (\vec{p}+\vec{q})\cdot\vec{q} v_{\vec{p}+\vec{q}}] \\ & \qquad + 2\vec{p}\cdot\vec{q} (\vec{k}+\vec{q})\cdot\vec{q} v_{\vec{p}} [\vec{k}\cdot\vec{q} v_{\vec{k}} - (\vec{k}+\vec{q})\cdot\vec{q} v_{\vec{k}+\vec{q}}]\} \sum_{\vec{m}\vec{n}\vec{l}} \langle c^\dagger_{\vec{m}+\vec{k}} c^\dagger_{\vec{n}+\vec{p}} c^\dagger_{\vec{l}} c_{\vec{l}+\vec{p}+\vec{k}} c_{\vec{n}} c_{\vec{m}}\rangle\,.\end{aligned} \tag{A4}$$

For simplicity we have not written the sums over the spin indices explicitly. The density of the electron gas is denoted by n.

As a first approximation we now ignore the difference between the exact single-particle density and the density for the noninteracting system.[9,10] Using Eqs. (A2) and (A3) we can write

$$\frac{(L^2\rho, L^2\rho)_q}{\epsilon_F^2(L\rho, L\rho)_q} = q^4 + \tfrac{12}{5} q^2 + \omega_p^2 \left[1 + \frac{3}{4}\int_0^\infty dk\, k^2[S(k) - 1]\left(\frac{5}{6} - \frac{k^2}{2q^2} + \frac{(k^2-q^2)^2}{4kq^3} \ln\left|\frac{k+q}{k-q}\right|\right)\right]. \tag{A5}$$

In this expression ω_p denotes the plasma frequency in units of the Fermi energy ϵ_F and the wave vectors are in units of k_F. $S(k)$ stands for the static structure factor.[6] In order to obtain a tractable expression for $(L^3\rho, L^3\rho)_q$, more assumptions are necessary. Therefore, the three-particle correlation function is approximated by its uncorrelated part[15] (cf. terms in ω_p^4). As we did not succeed in writing all two-body terms as a function of $S(k)$, we have replaced some difficult terms by their mean value. This approximation does not affect the essential wave-vector dependence. We then obtain

$$\begin{aligned}\frac{(L^3\rho, L^3\rho)_q}{\epsilon_F^4(L\rho, L\rho)_q} = {} & q^8 + 8q^6 + \tfrac{48}{7} q^4 + 2\omega_p^2[2 - g(0)](q^4 + \tfrac{12}{5} q^2) + \omega_p^4 \left[1 + \frac{3}{4}\int_0^\infty dk\, k^2[S(k) - 1]\right. \\ & \left.\times\left(\frac{17}{6} - \frac{2k^2}{q^2} + \frac{3}{2}\frac{q^2}{k^2} + \frac{(k^2-q^2)^2}{kq^3}\ln\left|\frac{k+q}{k-q}\right| + \frac{(k^2-q^2)^2}{4k^3q}\ln\left|\frac{k+q}{k-q}\right|\right)\right].\end{aligned} \tag{A6}$$

Finally, we want to stress the fact that the approximations we have made, have no implications at all as far as frequency sum rules are concerned. Care must be taken, however, that the Schwartz inequality is not violated. In our case this means that our expressions satisfy the inequalities

$$(L^2\rho, L^2\rho)_q(\rho, \rho)_q \geq (L\rho, L\rho)_q^2\,, \tag{A7a}$$

$$(L^3\rho, L^3\rho)_q(L\rho, L\rho)_q \geq (L^2\rho, L^2\rho)_q^2\,. \tag{A7b}$$

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