Dynamical correlations of simple metals: ε(k,ω) for arbitrary k and ω

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Dynamical correlations in the homogeneous electron gas are central to time dependent density functional theory, a very active area of research. The aim of the present investigation is to obtain exact results for dynamical correlation effects with the $\mathbf{k}$ and ω dependent dielectric function $\epsilon(\mathbf{k},\omega)$ of the jellium model in the high density limit. We write $\epsilon(\mathbf{k},\omega)$ in terms of the proper polarizability $\pi_p(\mathbf{k},\omega)$, i.e.

$$\epsilon(\mathbf{k},\omega) = 1 + v(\mathbf{k})\pi_p(\mathbf{k},\omega) \qquad (1)$$

In many body perturbation theory $\pi_p(\mathbf{k},\omega)$ is defined as the sum of irreducible diagrams. The leading correction to the RPA bubble are depicted in Fig.1 It was

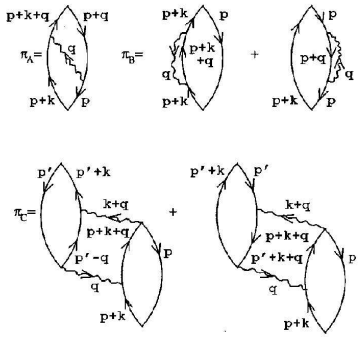

FIG. 1: *The π_A, π_B and π_C diagrams are lowest order corrections to the RPA bubble. The wavy lines signify the dynamically screened Coulomb interaction. p, p′, q and k are 4 dimensional vectors including frequency.*

demonstrated long ago [1] that a consistent set of diagrams generated in selfconsistent approximation (SCA) must include the π_C diagrams. It will then give the correct r_s dependence of the compressibility sum rule. Thus we have

$$\begin{aligned}\epsilon(\mathbf{k},\omega) &= 1 + v(\mathbf{k})[\pi_0(\mathbf{k},\omega + \\ &\quad + \pi_A(\mathbf{k},\omega) + \pi_B(\mathbf{k},\omega) + \pi_c(\mathbf{k},\omega) + \cdots] \qquad (2)\end{aligned}$$

$\pi_0(\mathbf{k},\omega)$ defines the well knonw Lindhard dielectric function $\epsilon_L(\mathbf{k},\omega) = 1 + v(\mathbf{k})\pi_0(\mathbf{k},\omega)$. Although these formal results were obtained long ago [2] the corresponding expressions of π_A, π_B and π_C involve seven-dimensional integrals whose numerical evaluations is a formidable task that prevented computation for arbitrary $\mathbf{k}$ and ω until recently [3]. There it was realized that the seven-dimensional integrals could be reduced to three-dimensional integrals by analytical means. Nevertheless the numerical evaluation of the diagrams is very complicated and as a first step the imaginary part $Im\,\epsilon(\mathbf{k},\omega)$ for $(\mathbf{k},\omega)$ *outside* the ph spectrum was computed, cf Fig.2.

Since $Im\,\pi_0(\mathbf{k},\omega)$ vanishes in that regime we have

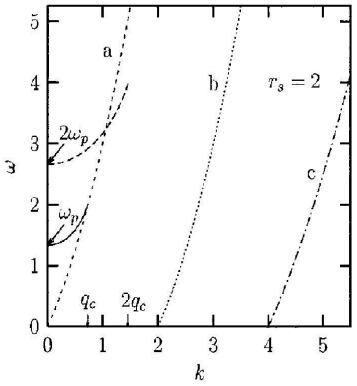

FIG. 2: *Excitation spectrum of the homogeneous electron gas. a: upper bound of the ph spectrum $\omega = k^2 + 2k$; b: lower bound of the ph spectrum $\omega = k^2 - 2k$; c: lower bound of the ph-ph spectrum $\omega = k^2/2 - 2k$. ω_{pl} denotes the onset of the RPA plasmon dispersion $\omega_{pl}(\mathbf{k})$ with the cutoff wavenumber q_c. The line $2\omega_{pl}(k/2)$ marks the onset of pl-pl excitations [4].*

$$\begin{aligned}Im\,\epsilon(\mathbf{k},\omega) &= v(\mathbf{k})Im\,[\pi_A(\mathbf{k},\omega) + \pi_B(\mathbf{k},\omega) + \pi_C(\mathbf{k},\omega)] \\ &\equiv Im\,(\epsilon_{ABC}(\mathbf{k},\omega) \qquad (3)\end{aligned}$$

Dynamical correlations that constitute $Im\,\epsilon_{ABC}(\mathbf{k},\omega)$ outside the ph spectrum were found to consist of ph-ph excititions, ph-plasmon (ph-pl) excitations and pl-pl excitations. Comparisons with existing experimental data on simple metals were very satisfying. Contributions to the plasmon half width [5]

$$\Delta E_{1/2}(\mathbf{k}) = 2\left(Im\,\epsilon_{ABC}(\mathbf{k},\omega) \times \left|\frac{\partial\epsilon_L(\mathbf{k},\omega)}{\partial\omega}\right|^{-1}\right)_{\omega=\omega_{pl}(\mathbf{k})} \qquad (4)$$

the high frequency tail of the dynamical structure factor

$$S(\mathbf{k},\omega) = \frac{\hbar k^2}{4\pi^2 e^2 n_0} Im\left[\frac{-1}{\epsilon(\mathbf{k},\omega)}\right], \qquad (5)$$

$$\sigma(\omega) = \frac{4}{3}\frac{|V^s_{\mathbf{G}_{110}}|^2|\mathbf{G}_{110}|^4}{\omega^3} Im\,\epsilon_{ABC}(\mathbf{G}_{110},\omega) \qquad (6)$$

and optical absortion of alkali metals [6] were considered. These successes stimulated further experimental investigations on simple metals. In particular, the predicted peaklike structures in the high frequency tail of $S(\mathbf{k},\omega)$ resulting form pl-pl excitations were recently observed experimentally by IXSS [7] on *Al* and *Na*, cf. Fig.3 and Fig.4. The rather successful comparison with various experimental obseravations is the impetus behind our effort to complete the evaluation of $\epsilon(\mathbf{k},\omega)$ by computing $Im\,\epsilon(\mathbf{k},\omega)$ inside the ph spectrum as well as $Re\,\epsilon(\mathbf{k},\omega)$.

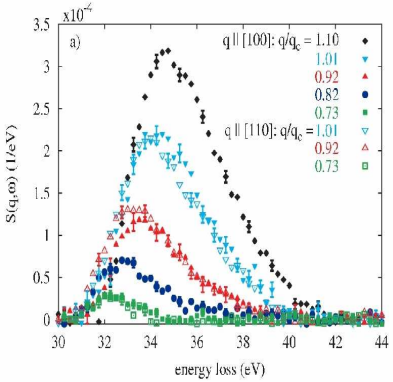

FIG. 3: *Al pl-pl correlation peak extracted from the experimental $S(\mathbf{q},\omega)$ spectra.*

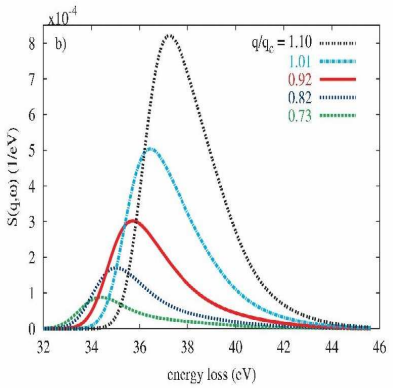

FIG. 4: *Calculated Al pl-pl correlation peak for $r_s = 2.07$ on an absolute scale, broadened by the experimental resolution.*

This is the most difficult part of the computation because of the singular nature of the integrands in this $(\mathbf{k},\omega)$ regime. Work is in progress. When this is achieved not only structures *inside* the ph spectrum observed in experiments can be analysed and compared with the calculated result but also the quality of the considered approximation could be assessed by checking Kramers-Kronig relations and sum rules such as the f-sum rule, the conductivity sum rule, the third frequency moment sum rule and the ground state theorem that relates the excitation spectrum to the ground state energy [8]. This will also shed light on the importance of diagrams not considered in the present approximation, such as e.g. second order exchange diagrams which might be responsible for the discrepancy in the intensity of pl-pl excitations between theory and experiment, cf. Fig.3 and Fig.4.

Summary

Dynamical correlations in the homogeneous electron gas are investigated by evaluating exactly the leading correction to the RPA proper polarizability that defines the frequency ω and wavevector $\mathbf{k}$ dependent dielecric function $\epsilon(\mathbf{k},\omega)$. *Outside* the particle-hole (ph) spectrum predicted peaklike structures in the high frequency tail of the dynamic structure factor due to double plasmon excitations were recently observed in inelastic x ray scattering spectroscopy (IXSS) on *Al* and *Na*. The evaluation of $\epsilon(\mathbf{k},\omega)$ *inside* the ph spectrum is in progress. This will enable us to analyze further experimental observations inside the ph spectrum of simple metals and assess the quality of the approximation by checking sum rules and the ground state theorem.

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[4] Frequencies are measured in units of the free electron Fermi energy and the wavevectors in units of the Fermi wave number.

[5] The plasma frequemcy $\omega_{pl}(\mathbf{k})$ is the solution of $\epsilon_L(\mathbf{k},\omega) = 0$

[6] $V^s_{\mathbf{G}_{110}}$ is the screened psudopotential, $\mathbf{G}_{110}$ is a BCC reciprocal lattice vector.

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