Eur. Phys. J. B **37**, 3–6 (2004)
DOI: 10.1140/epjb/e2004-00023-3

THE EUROPEAN PHYSICAL JOURNAL B

Correlation in the Al plasma excitation

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Received 24 July 2003
Published online 19 February 2004 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2004

Abstract. The density-density correlation function in elementary excitations of the interacting electron gas is calculated from a simple model of the dynamic form factor. For the Al plasma excitation, a correlation length of ≈0.2 nm is found. It is shown that the small correlation length does not contradict the surprisingly large coherence length of almost 10 nm recently found in inelastic interference experiments. The difference of nearly 2 orders of magnitude can be traced back to the long range Coulomb interaction between probe and target electrons.

PACS. 34.80.Pa Coherence and correlation in electron scattering – 71.45.Gm Exchange, correlation, plasmons – 82.80.Pv Electron spectroscopy

1 Introduction

The correlation length in solid state plasmas has been a matter of concern since many decades. It is reasonable to assume that the movement of electrons is correlated over a distance of the order of magnitude of the Thomas-Fermi screening length. A word of caution is in place here because the Thomas-Fermi screening length is a static quantity whereas the electrons in a solid state plasmon oscillate at a frequency of 10^{15} Hz. Since the plasmon is a coherent superposition of electron-hole pairs [1] the exchange-correlation hole [2] might serve as another length scale. In metals, both the static screening length and the extension of the exchange-correlation hole amount to some Ångstroms, roughly speaking the inter-electron distance.

A simple classical argument suggests to define the extension of the plasmon as the product of the lifetime and the group velocity. For Al, this was found to be 0.4 nm [3], a value seemingly in agreement with early experiments on the spatial resolution in plasmon energy filtered images [4]. However, more recently Lichte and coworkers [5,6] reported interference experiments with fast electrons having excited a plasmon in Al. Contrary to the early predictions they found a coherence length of almost 10 nm from fringe contrast in the inelastic hologram.

The situation appears even more confusing when the uncertainty principle is used to determine the (coherent) extension of the plasmon scattered electron: this is by definition $\delta x = \hbar/\sqrt{\langle \delta q^2 \rangle}$. Since the scattering distribution is a Lorentzian (the variance of which is infinite) the cohrence length of the scattered electron comes out as $\delta x = 0$.

Applying the Wiener-Khinchin theorem it is shown that the density-density correlation function of a charge oscillation in a medium is related to its dynamic form factor which in turn is accessible by inelastic scattering experiments with fast electrons. We are then in a position to work out the relationship between the density correlation function, experimentally determined scattering cross sections and the coherence of the probe electrons. We find that the experimentally determined coherence length is precisely predicted by theory, and that this is consistent with the small correlation length in solid state plasmas.

With the time-dependent density autocorrelation in the ground state of a quantum mechanical system

$$p(\boldsymbol{r},t) = \int d^3r' \langle n(\boldsymbol{r}+\boldsymbol{r}',t) n(\boldsymbol{r}',0)\rangle \tag{1}$$

where $n(\mathbf{r}) = \sum_{\mathbf{i}} \delta^{\mathbf{3}}(\hat{\mathbf{r}}_{\mathbf{i}} - \mathbf{r})$ is the particle density operator we can write the density autocorrelation function of an eigenmode of energy E of this system as

$$p_E(\mathbf{r}) = \frac{1}{2\pi\hbar} \int dt e^{iEt/\hbar} p(\boldsymbol{r},t). \tag{2}$$

2 The density-density correlation function in the plasmon

According to the Wiener-Khinchin theorem and the fact that the operator FT of the Fourier transformation is linear we have

$$FT_{\mathbf{r}}[p_E(\mathbf{r})] = \frac{1}{2\pi\hbar} \int dt\, e^{iEt/\hbar} \langle n_{\boldsymbol{q}}(t) n_{-\boldsymbol{q}}(0)\rangle. \tag{3}$$

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where

$$n_{\mathbf{q}} = FT_{\mathbf{r}}[n(\mathbf{r},t)] \tag{4}$$

is the Fourier transform of the particle density operator.

It can be shown [7] that the dynamic form factor is

$$S(\boldsymbol{q},E) = \frac{1}{2\pi\hbar}\int dt e^{iEt/\hbar}\langle n_{\boldsymbol{q}}(t)n_{-\boldsymbol{q}}(0)\rangle. \tag{5}$$

Comparing equations (3) and (5) we see that

$$p_E(\mathbf{r}) = FT_{\mathbf{q}}[S(\mathbf{q},E)]. \tag{6}$$

Note that we applied the Wiener-Khinchin theorem to operators. Although equation (5) has a product of density operators on the right hand side the left hand side is not a product any longer due to the expectation value. Only the static form factor $S(\mathbf{q},0)$, i.e. the quantity describing elastic scattering decomposes into a product, and one retrieves the well-known theorem that the Fourier transform of the absolute square of the scattering amplitude is the autocorrelation of the static charge density.

Knowledge of the dynamic form factor $S(\mathbf{q},E_{pl})$ for the plasma oscillation at energy E_{pl} immediately gives the density autocorrelation, equation (6). One can either calculate $S(\mathbf{q},E_{pl})$ for a given system, or derive it from experiment since the dynamic form factor relates to the double differential inelastic scattering cross section as [7]

$$\frac{\partial^2\sigma}{\partial E\partial\Omega} = \frac{4\gamma^2}{q^4a_0^2}\frac{k}{k_0}S(\mathbf{q},E), \tag{7}$$

where $a_0 = 4\pi\varepsilon_0\hbar^2/me^2$ is the Bohr radius, and γ is a relativistic factor. Equation (7) is valid in the first Born approximation for an incident plane wave, and for single scattering [8,9].

Experimentally, the inelastic electron scattering cross section for cubic systems is isotropic and is to a good approximation Lorentzian in wave number transfer q, with a cutoff q_c. It then follows that the dynamic form factor must be proportional to q^2 up to the cutoff and vanish thereafter.

The plasmon in simple metals is a well defined elementary excitation. Moreover in cubic systems such as Al these excitations are isotropic. Ignoring dispersion and lifetime broadening the dynamic form factor reads

$$S(\mathbf{q},E) = \mathbf{q}\cdot\mathbf{q} \quad \delta(E-E_{pl}). \tag{8}$$

In this approximation the Fourier transform equation (6) can be performed immediately

$$\begin{aligned} p_E(r) &= 2\pi\int_{q_e}^{q_c} q^2S(q,E)dq\int_0^\pi d\vartheta\sin\vartheta e^{iqr\cos\vartheta} \\ &= 4\pi\int_{q_e}^{q_c} q^4\frac{\sin(qr)}{qr}dq \quad \delta(E-E_{pl}) \end{aligned} \tag{9}$$

where we have used the fact that no wave number transfer less than the characteristic transfer $q_e = k_0E/(2E_0)$ can occur.

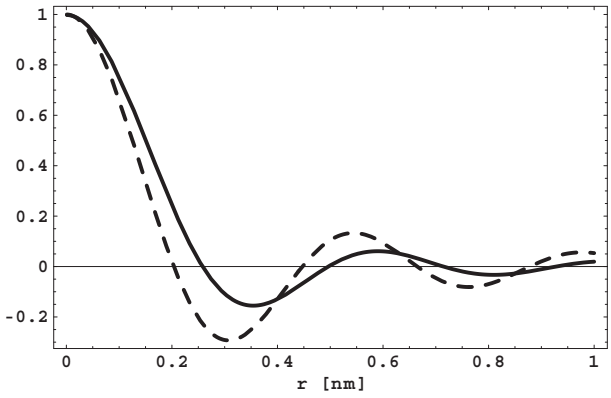

Fig. 1. Density autocorrelation function of the Al plasma excitation. Full line: equation (9), dashed line: projection onto plane of observation, equation (10). Abscissa in nm.

If the density correlation is studied experimentally, e.g. by an electron interferometer or by phase contrast imaging, this will be done in an electron microscope. The spatial information available will then be extracted from a projection onto the image plane with coordinates $\mathbf{x}$ (which by Fourier transform corresponds to the subspace in reciprocal space subtended by the diffraction plane). When comparing predictions with experimental results one is therefore rather interested in the z-projected correlation function $\bar{p} = \int p(\mathbf{r})dz$. This is the two-dimensional Fourier transform with respect to the coordinates $\mathbf{q}_x$ in the diffraction plane

$$\begin{aligned} \bar{p}_E(\mathbf{x}) = FT_{\mathbf{q}_x}[S(\mathbf{q},E)] &= \int_0^{q_c} S(q,E)e^{i\mathbf{q}_x\mathbf{x}}d^2q \\ &= 2\pi\int_0^{q_c} S(q,E)e^{iqx\cos\varphi}qdqd\varphi \\ &= 2\pi\int_0^{q_c}\left(q_e^2+q^2\right)J_0(qx)qdq \quad \delta(E-E_{pl}). \end{aligned} \tag{10}$$

This integral can be performed analytically. Results are shown for Al, compared to the 3-dimensional correlation function, in Figure 1. Parameters were $q_E = 0.0938/\text{nm}$ and $q_c = 15/\text{nm}$.

The first zero is found at 0.2 nm for the projected correlation function. Its FWHM is at 0.12 nm, and the enveloppe has a value of 0.1 at a distance of 0.77 nm. This shows that contrary to intuitive arguments the collective oscillation of the nearly free electrons in aluminium has low coherence. The average distance of conduction electrons in Al in the ground state is 0.22 nm. The electric field of an oscillating charge is so strongly screened that the second nearest neighbours react only slightly. When excited (e.g. by a fast electron) the electrons in the solid state plasma rearrange such that at a distance of 0.35 nm from the excitation a depletion zone is created whereas at 0.55 nm a higher density of electrons is found. We note in passing that a more rigorous description including plasmon dispersion, lifetime broadening and a smooth decay of the cross section instead of a sharp cutoff at q_c will

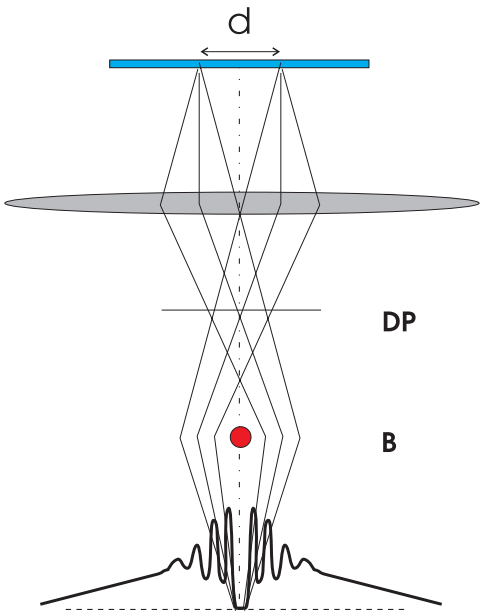

Fig. 2. Schematic of an interference experiment with an electrostatic biprism. [6]. Electrons coherently emerging a distance d apart from the specimen form a damped sinusoidal interference pattern in the observation plane after having passed the biprism (B). DP is the diffraction plane of the objective lens.

slightly alter these results with a tendency to reduce the amplitude of the wiggles.

In principle, these wiggles are well beyond the resolution limit of modern electron microscopes. However, in energy filtered imaging in the transmission electron microscope (EFTEM) with the pass energy set at the plasma frequency, correlations are not visible. What is visible is the localization of excitations. Since the nearly free electrons in the target are delocalized the plasma excitation may take place at any position in the specimen. The EFTEM image is a homogeneously illuminated area.

3 Coherence length of plasmon scattered electrons

In order to make the correlation visible one needs an interferometer. Lichte and coworkers [5,6] reported interference experiments with fast electrons having excited a plasmon in Al. They found a coherence length of almost 10 nm from fringe contrast in the inelastic hologram. The experiment is sketched in Figure 2.

How can this surprisingly high value be reconciled with a correlation length of only 0.2 nm?

In their experiment partial waves emerging from points a distance d apart in the specimen were brought to interference by use of an electrostatic biprism. The distance d could be tuned by the voltage on the biprism. Fringe contrast decreased beyond the 10% level when d approached

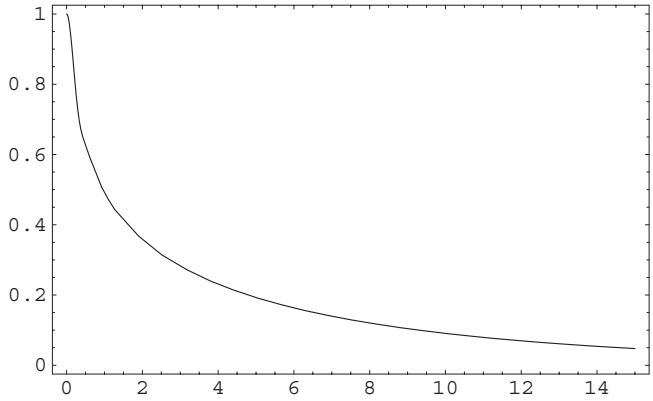

Fig. 3. Mutual coherence function μ_E of electrons emerging from the exit plane of an Al specimen, after excitation of a plasmon. Abscissa in nm.

10 nm[1]. The contrast measured in this type of experiment is the mutual coherence function [11] of electrons having suffered a plasmon loss.

Similar to the previous reasoning, and according to the Wiener-Khinchin theorem, the mutual coherence function of the fast probe electron is the Fourier transform of the energy filtered electron density distribution ρ_E in the diffraction plane of the objective lens:

$$\mu_E(\mathbf{x}) = FT_{\mathbf{q}_x}[\rho_E(\mathbf{q}_x)]. \tag{11}$$

Here we have neglected aberrations of the post-specimen lenses. Since ρ_E is proportional to the experimentally accessible double differential scattering cross section, equation (7) we can assume that this is a Lorentzian angular distribution with cutoff q_c. The Fourier transform can be performed exactly as that of the dynamic form factor, equation (10), and we arrive at

$$\begin{aligned}\mu_E(\mathbf{x}) = FT_{\mathbf{q}_x}\left[\frac{1}{q_E^2+q^2}\right] &= \int_0^{q_c} \frac{1}{q_E^2+q^2} e^{i\mathbf{q}\mathbf{x}} d^2q \\ &= 2\pi \int_0^{q_c} \frac{1}{q_E^2+q^2} J_0(qx) q dq. \end{aligned} \tag{12}$$

This function is shown in Figure 3 for the Al plasmon. The 10% level is at a distance of 9.1 nm, in excellent agreement with the interference experiment of Lichte and coworkers. Figure 4 shows the first 2 nm together with the 2-D correlation function. The mutual coherence function is so broad because of the long range Coulomb interaction between target charges and the probe electron. So it may happen that two partial inelastically scattered waves emerging from the specimen a distance d apart have excited the same plasmon and thus are coherent over that distance although the plasmon *per se* is very localized. The only physical information on the target is contained

[1] A recent biprism experiment of the same group with a field emission machine shows fringe contrast up to more than 20 nm. Preliminary calculations yield good agreement with the present theory [10].

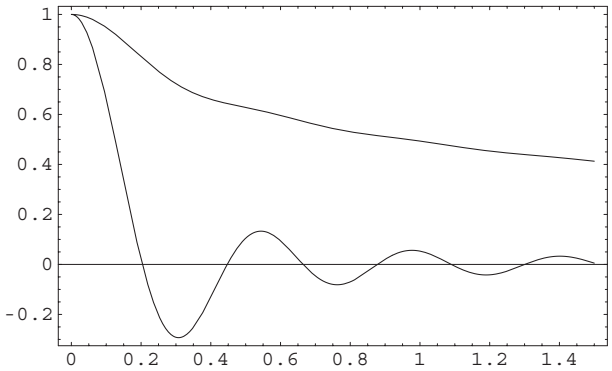

Fig. 4. Full line: Mutual coherence function μ_E of electrons as in (a). Dashed line: 2-dimensional projection of the density correlation function, equation (10). Abscissa an nm.

in the faint wiggles at the first nm which are in fact remnants of the strong oscillations in the correlation function with a period of ≈0.55 nm. It seems difficult to extract these wiggles from the contrast observed in a bisprism experiment.

4 Conclusion

In conclusion, it was shown how the small correlation length in a plasma excitation can be reconciled with the coherence length of almost 10 nm found in inelastic interference experiments. The difference of nearly 2 orders of magnitude can be traced back to the long range Coulomb interaction between probe and target electrons. It is the high coherence of the probe electrons, not the correlation of the target electrons that gives rise to the strong interference fringes. A consequence is that interference experiments in the electron microscope cannot directly reveal correlation lengths, at least not in the low energy loss region. It is possible that the faint wiggles visible in the coherence function can be used to obtain information on the correlation length. Another surprising prediction is that even without spatial correlation in the target, e.g. for core level ionization, the biprism experiment would reveal similar fringes as for the plasma excitation.

This work was supported by the Austrian Science Fund under project nr. P14038-PHY, and by the joint project of the French CNRS and Vienna University of Technology PICS 913. P. S. acknowledges financial assistance of the Hochschuljubiläumsstiftung of the municipality Vienna (project H-175/2001).

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