Measurement of the Static Structure Factor for Conduction Electrons with Use of Synchrotron Radiation

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A technique for directly measuring the static structure factor for electrons with use of synchrotron radiation is described. Results for the conduction electrons in beryllium and the basal plane of graphite show a peak at $k \cong 2k_F$ as predicted, but the pair correlation function shows an anomaly at distances considerably shorter than the mean interparticle separation.

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The measurement of the structure factor of gases, liquids, and disordered solids has contributed substantially to our understanding of their atomic structure. The static structure factor for atoms, $S_a(k)$, is obtained by scattering a monochromatic beam from the sample and measuring, for a given momentum transfer k, the intensity integrated over the energy loss due to atomic motions. If $S_a(k)$ is determined at all momentum transfers, then it can be inverted to obtain the pair correlation function for the atoms $g_a(r)$. $S_e(k)$ can be measured by integrating over the losses due to electronic excitation. Since the two types of losses differ by two to three orders of magnitude in energy, they are in principle separable in an x-ray scattering experiment.

Although this idea is certainly not new[1,2] and appears to be conceptually very simple, it has not been pursued until now for a variety of reasons including experimental difficulties. In this work, we describe a novel application of the properties of synchrotron radiation which allows for the direct measurement of $S_e(k)$. While this technique can be applied to the study of arbitrary electronic systems, we have chosen for its initial and possibly most important application, the study of the properties of solid-state electron gases. This problem was chosen because dynamic structure factor, $S_e(k,\omega_e)$, measurements in many materials have revealed an anomalous dispersion of the plasmon and its continued existence well into the single-particle continuum.[3-7] These observations prompted the speculation that the anomaly was a general property of the electron gas.[5,8] The deviation from mean-field theories occurred at momentum transfers around $2k_F$, leading to speculation that it was due to short-range order and that one should find a peak in $S_e(k)$ at $2k_F$.[8] Our results do indeed find a significant peak in $S_e(k)$ for both Be and along the a axis in graphite at roughly $2k_F$, but the resulting electronic pair-correlation function, $g_e(r)$, suggests an alternative interpretation than the one originally proposed.[8]

In a general inelastic x-ray scattering experiment, the cross section[9] can be written when the losses are truly separable as

$$\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d\sigma}{\sigma\Omega}\right)_{\mathrm{Th}} \frac{\omega_1}{\omega_2} \left[S_a(k,\omega_a) + S_e(k,\omega_e)\right], \qquad (1)$$

where $(d\sigma/d\Omega)_{\mathrm{Th}}$ is the Thompson cross section[9] with the energy and momentum transfers given by $\omega_{a,e} = \omega_1 - \omega_2$ and $k = |\vec{k}_1 - \vec{k}_2| \cong 2k_1 \sin\frac{1}{2}\theta$. Here (ω_1, k_1) and (ω_2, k_2) are the energy and wave vector of the incident and scattered waves and θ is the angle between the directions of the incident and scattered waves. $S_a(k,\omega_a)$ and $S_e(k,\omega_e)$ correspond to transitions where the momentum and energy are transferred to the atom (via the bound electrons) and directly to the electrons, respectively. The first involves energy shifts of order 10^{-2} eV, while the other involves shifts of order 100 eV as would be expected on the basis of the relative mass of an electron and of the atom. The fact that the two contributions are separable in the case of the electron gas was shown in previous $S_e(k,\omega_e)$ experiments.[3] In principle, one could perform $S_e(k,\omega_e)$ experiments at various momentum transfers and then integrate the scattering cross section involving losses greater than several electron volts to determine $S_e(k)$. However, this is very time consuming since each $S_e(k,\omega_e)$ takes about eight hours to measure with the energy resolution necessary to separate the two components. It is also not accurate since the integration is very sensitive to the long tails of $S_e(k, \omega_e)$ and to the background subtraction.

To separate $S_a(k)$ and $S_e(k)$, we have employed a method first developed by Ice, Chen, and Crasemann[10] for other purposes. By performing the scattering experiment at an incident x-ray energy just above the absorption threshold of some material, then by filtering the scattered beam with that material, one can selectively reject with high efficiency the $S_a(k, \omega_a)$ contributions which are quasielastically scattered. In this experiment the incident energy was chosen to be 11 546 eV, which is just above the L_{I} absorption threshold of metallic platinum. The polychromatic synchrotron radiation from SPEAR was monochromatized by a pair of Si(220) crystals operating in the parallel mode. An input slit defined the energy resolution to be approximately 1 eV. The sample was oriented and mounted on the θ axis of a four-circle diffractometer, and measurements were performed in the transmission mode. On the detector axis a double-slit system defined the angular acceptance of a NaI scintillation detector to be ± 0.10 deg, which is ± 0.02 Å at $k = 6$ Å^{-1}. Average count rates of 2000 Hz were obtained with an incident flux of 10^{11} Hz. The system was calibrated by measuring the x-ray fluorescent yield from Mn sample in a $\theta - 2\theta$ scan. The resultant distribution agreed to within 1% of that expected from an angularly independent cross section.

The absolute absorption characteristics of the platinum foil used on the detector arm were determined by measuring the transmission with and without the foil in the beam. The results are shown in Fig. 1. $S_e(k)$ was determined by measuring the scattered intensity as a function of momentum transfer with use of a normal $(\theta, 2\theta)$ scan[11] at two different incident energies. It was first measured at the absorption peak (point A in Fig. 1) where the elastic component, $S_a(k)$, was attenuated by a factor $e^{-3.4}$. It was then measured at point B, 10 eV lower in energy, where the attenuation is only $e^{-1.4}$.

The results of the two measurements are shown in Fig. 2(a). Notice the significant reduction of the sharp quasielastic peaks by the filter as well as the roughly sample-independent background in the forward direction which is also mainly quasielastic. The two spectra, taken together with the measured absorption characteristics of the plati-

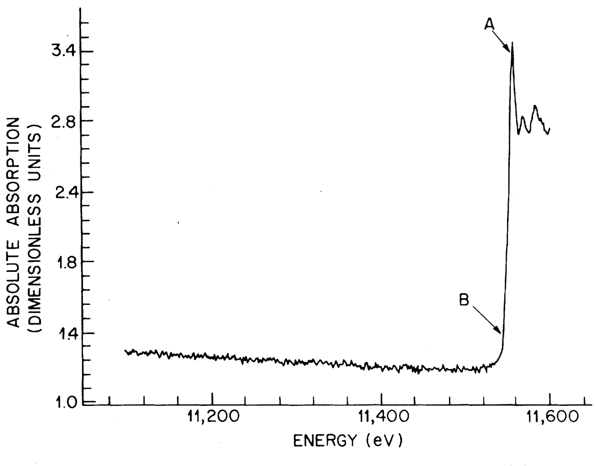

FIG. 1. Absolute absorption of the platinum filter.

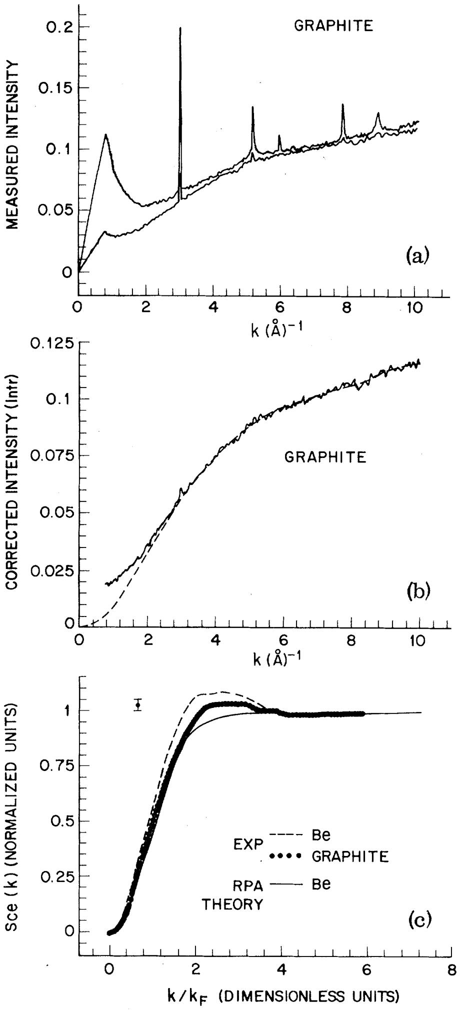

FIG. 2. (a) Total data measured at B (upper curve) and A (lower curve), (b) both $S_a(k)$ and background removed (solid curve) and k^2 continuation to zero (dashed curve), (c) $S_{ce}(k)$ from Eqs. (2)–(4).

num foil, can be used to uniquely determine the contribution of $S_e(k)$. Corrections were also made for the finite absorption effects of the filter on $S_e(k)$. The quantitative evaluation of the accuracy of this removal, which involves no adjustable parameters, can be determined from Fig. 2(b). There the unnormalized, experimentally determined $S_e(k)$ is plotted, and one can see that the sharp spikes are reduced by a factor of 100.

Since it is known that near $k=0$, $S(k)$ should be proportional to k^2, it becomes clear in Fig. 2(b) that the dependence is not manifested by the data. This is explained by the observation that if the sample is removed, one sees an almost identical contribution which is also not removed by the two energy measurement procedures described above. However, direct measurements with and without the filter with no sample, together with the k^2 shape near $k=0$, can be used to uniquely determine $S_e(k)$ in the low-k region with results shown in Fig. 2(b). In any case, this is not the region of interest to this study as the $S_e(k,\omega_e)$ experiments have shown normal random-phase-approximation-like dispersion in that region.[3]

The final curve in Fig. 2(b) represents the unnormalized and sample absorption uncorrected electronic static structure factor which includes contributions from both the core and the conduction electrons. After experimentally measuring the absorption of the sample one can easily correct for sample absorption effects. The absorption corrected curve, $S_e(k)$, can be converted to the normalized contribution of the conduction electrons, $S_{ce}(k)$, by use of Eq. (1) and the following three relationships:

$$aS_e(k) = aS^{\mathrm{core}}(k) + aS_{ce}(k), \tag{2}$$

$$S_{ce}(k \to k_{\max}) = 1, \tag{3}$$

$$S^{\mathrm{core}}(k) = 1 - \{1 + [0.264k/(Z - \tfrac{5}{16})]\}^{-1}. \tag{4}$$

Equation (4) is based on atomic calculations for the two 1s electrons where the use of an effective nuclear charge, $Z-\frac{5}{16}$, was found experimentally to work in a study of $S^{\mathrm{core}}(k)$ for helium.[12] The normalized results for $S_{ce}(k)$ for both beryllium and basal-plane graphite are shown in Fig. 2(c). It is clear that for both beryllium and graphite there is a peak in $S_{ce}(k)$ at roughly $2k_\mathrm{F}$. Note the significant discrepancy from the random-phase–approximation calculation for beryllium (graphite is very similar) which is also shown in Fig. 2(c).

The smaller peak for graphite is consistent with the previous $S_{ce}(k,\omega_e)$ studies[5] where the graphite spectrum was slightly more random-phase-approximation-like than beryllium. Something not previously appreciated from the $S_{ce}(k,\omega_e)$ experiments[4-6] is the broadness of the $S_{ce}(k)$ peak. The breath is a direct consequence of the associated correlations in r space. In scattering studies of liquids which have short-range order, the peaks in $g(r)$ are at the average interparticle separation, $\bar{r}$, or in more disordered systems the distance of closest approach r_e. The two values do not differ significantly in ordinary liquids.

An empirical $g_{ce}(r)$ can be determined from our data by using the general relationship[1,11]

$$g_{ce}(r) = 1 + (1/2\pi^2 N r)\int k \sin(kr)[S_{ce}(k) - 1]\,dk, \tag{5}$$

where N is the particle density. In order to obtain our empirical $g_{ce}(r)$ we had to analytically extend our data to $k=\infty$. This was done by allowing $S_{ce}(k)$ to approach unity with the asymptotic random-phase–approximation form such that $g_{ce}(0)$ was equal to zero. The results of this procedure are shown for beryllium and graphite in Fig. 3. In our data $k[S_{ce}(k)-1]$ is composed of two components. The smooth random-phase-approximation-like component (Fig. 2) will produce a smooth $g_{ce}(r)$ as shown in Fig. 3. However, superimposed on that component is a broad peak centered at $k \cong 2.5k_\mathrm{F}$. This will produce a damped oscillation with a period of $rk_\mathrm{F} \cong \pi/2.5$ with the first peak at $rk_\mathrm{F} \cong \pi/5$ which is in fact what is observed in Fig. 3. The details of height and exact value of the peak should not be taken too seriously because

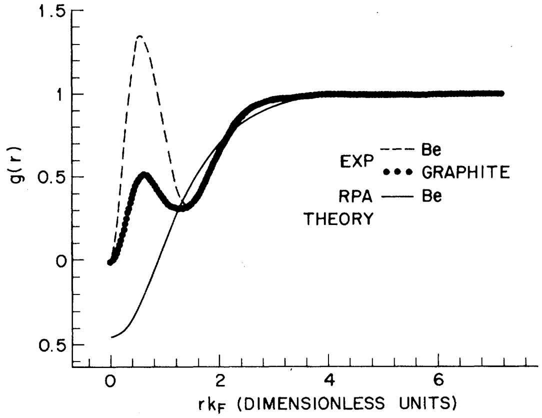

FIG. 3. Experimental and theoretical pair-correlation function.

of the arbitrary extension of $S_{ce}(k)$ to high k and uncertainties in the exact slope of $S(k)$ at high k. Extreme variations of that slope due to possible systematic experimental uncertainties were imposed on the $S(k)$ data. While changes in the exact size, $\pm 25\%$, and position, $\pm 0.3 r k_{\mathrm{F}}$, of $g(r)$ were observed; all had peaks with $r < \bar{r}$. The results, therefore, do strongly indicate the phenomena associated with the anomalous plasmon dispersion and the peaking of $S_{ce}(k)$ at $2k_{\mathrm{F}}$ correspond to features of the electron-electron interaction at distances shorter than $\bar{r}$. It is possible that interaction with the localized, bound core electrons is responsible for the short-range correlation. Arguing against this interpretation is the observations in $S_e(k,\omega)$ studies[5] and in this work that the results look very similar when scaled to electron-gas parameters. However, a possible interpretation is that in real solids, as opposed to the idealized electron gas with its jellium background, the effect of electron correlation is not to condense to a Wigner lattice at $r = \bar{r}$, but rather to make a Mott-type transition to an atomiclike state. However, on the basis of these experiments, we can only make a general observation that the characteristic distance associated with the anomalies is shorter than $\bar{r}$.

It is hoped that this measurement of $S_{ce}(k)$ will stimulate further theoretical interest in the nature of interactions in the electron gas and that the observation in this work that short-range interactions are important will be useful. In a broader sense it is hoped that the general technique described here will be extended to study other Fermi liquids as well as other electron systems. In principle $S_e(k)$ studies can provide a testing ground for our microscopic understanding of electronic interactions.

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Effect of Charging Energy on Kosterlitz-Thouless Transition Temperature in Granular Films

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A relationship between the Kosterlitz-Thouless transition temperature T_{KT} and the sheet resistance, $R_{\square}{}^{N}$, of a granular superconducting film is derived taking into account the electrostatic charging energy. The zero-point phase fluctuations produce a drop of the areal superfluid density at a critical value of the sheet resistance in agreement with the recent observation on granular lead films.

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Recently Beasley, Mooij, and Orlando[1] and Halperin and Nelson[2] considered the possibility of the vortex unbinding transition in superconducting films. A simple relation has been established between the Kosterlitz-Thouless transition temperature T_{KT} and the sheet resistance R^{N} of the film.[1] On comparing this relation with the broadening of the resistive transition, taken from a wide varie-