Presence of an Incipient Wigner Electron Lattice in Solid-State Electron Gases

P. M. Platzman and P. Eisenberger
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 23 April 1974)

In x-ray studies of the dynamic structure factor $S(k,\omega)$ in Be, graphite, and Al a new type of excitation spectrum is observed for momentum transfers k between k_F and $2k_F$. The similarity of the observed spectra in these systems, when scaled according to electron-gas parameters, strongly indicates that the new excitation is a property of a solid-state electron gas and not a one-electron band-structure effect.

In previous inelastic x-ray[1,2] and electron[3] scattering experiments it has been observed that the plasmon dispersion in solid-state systems deviates from mean-field theoretical predictions in that the dispersion is much less than predicted for momentum transfers k in the region of the Fermi momentum k_F. Various unsuccessful theoretical attempts have been made to explain these observations by modifying existing mean-field theories for the electron gas by including lifetime effects for the excited electron-hole pair[4] or by including band-structure effects.[5,6]

In this work we extend, for the first time, the region of experimental investigation to the momentum transfer region between k_F and $2k_F$. We discover in Be, graphite, and Al that the observed spectrum deviates qualitatively from mean-field-theory predictions. More precisely we find that, contrary to theoretical predictions, a plasmon-like excitation exists in this region. It is primarily the presence of this excitation which is responsible for the deviation of the observed spectra from mean-field theoretical predictions. To distinguish this rather broad excitation from the well-defined long-wavelength plasmon it will henceforth be refered to as a plasmon band.

The similarity of the observed spectra, when scaled to electron-gas parameters, in the disparate systems Be, graphite, and Al strongly suggests that the observed plasmon band is a general property of the solid-state electron gas and not a property of the one-electron band structure. We will propose that the plasmon band is a manifestation of the spatial inhomogeneity in solid-state electron gases which in turn is a result of the combined effect of the electrons' interaction with other electrons and with the positive background. Such a model can qualitatively be viewed as a soft Wigner electron lattice.[7] In fact, such properties (as we shall see) of the electron lattice as negative dispersion of the elementary excitations are indeed observable in our experimental results.

The measured scattered intensity $I(k, \omega)$ for weak scattering of nonrelativistic x rays is quite generally related to the inverse dielectric function $[\epsilon(k, \omega)]^{-1}$, i.e.,

$$I(k, \omega) = (d\sigma/d\Omega)_0 S(k, \omega) \tag{1}$$

with

$$S(k, \omega) = (k^2/4\pi e^2)\,\mathrm{Im}[1/\epsilon(k, \omega)]. \tag{2}$$

Here k, ω are the momentum and energy transfers involved in the scattering process, and $(d\sigma/d\Omega)_0$ is the Thomson cross section.

The experimental apparatus and procedure is as described previously[2] except that in this study the samples were in a vacuum at nitrogen temperatures and Ge(111), rather than Ge(220), reflections were used in the double-crystal dispersive spectrometer. The low temperature was used to reduce quasielastic scattering [or thermal diffuse scattering (TDS)] and the Ge(111) spectrometer gave 25% worse resolution but three times the scattered intensity. The experiments are performed by fixing k and analyzing the energy distribution of the scattered x rays. In Fig. 1(a) are displayed the spectra obtained in studying Be in the region $1 \leq k/k_F \leq 2$. Each spectrum is taken at a fixed momentum transfer. In Fig. 1(b) we show a series of theoretical curves at corresponding k/k_F values calculated using the conventional random-phase approximation (RPA), i.e., Lindhard dielectric function with no band-structure effects included. The theoretical results have been smeared by the experimental resolution function, a Lorentzian with a full width at half-maximum of 5.0 eV. The experimental data are uncorrected for finite-resolution effects.

Calculations within a mean-field picture have shown that the characteristic loss function (energy transfers of the order of 50 eV) of a metal such as Be is only weakly effected by band-structure effects.[6] The essential validity of this as-

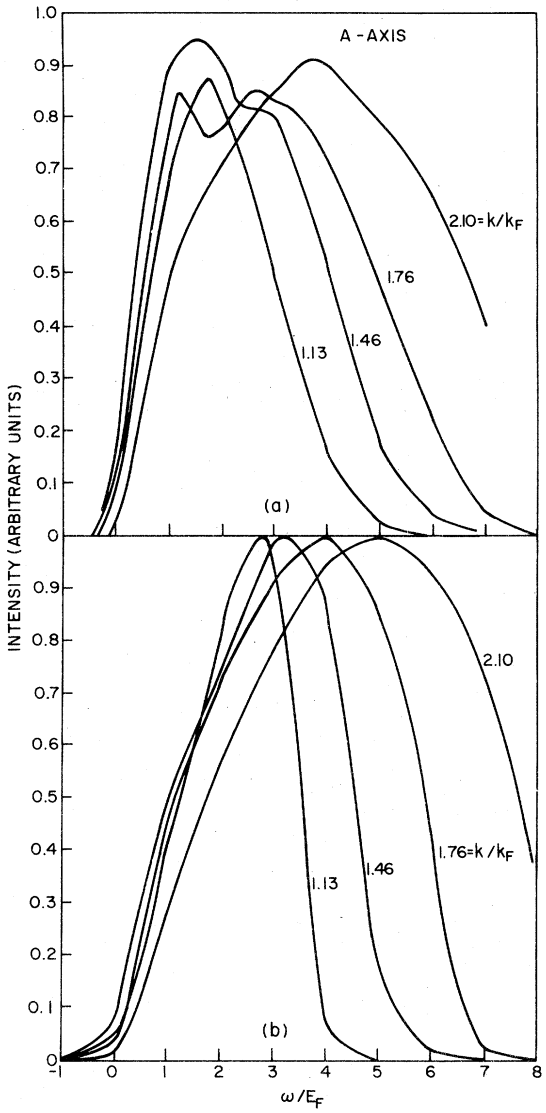

FIG. 1. Plots of $S(k,\omega)$ for fixed k. (a) Experimental results in Be. (b) Theoretical results obtained in RPA.

sumption is strongly supported by the results presented in Fig. 2. Here representative results for Al and graphite as well as Be are displayed. The similarity of the three spectra in these disparate systems is a strong experimental indication that the observed spectra are *not* a result of simple one-electron properties of those systems, since the one-electron band structure of these materials is so different. For other orientations and for other momentum transfers the basic similarity of the spectra is maintained. Indeed in studies of Li, Mg, and Si similar observations have also been made. The results for all sys-

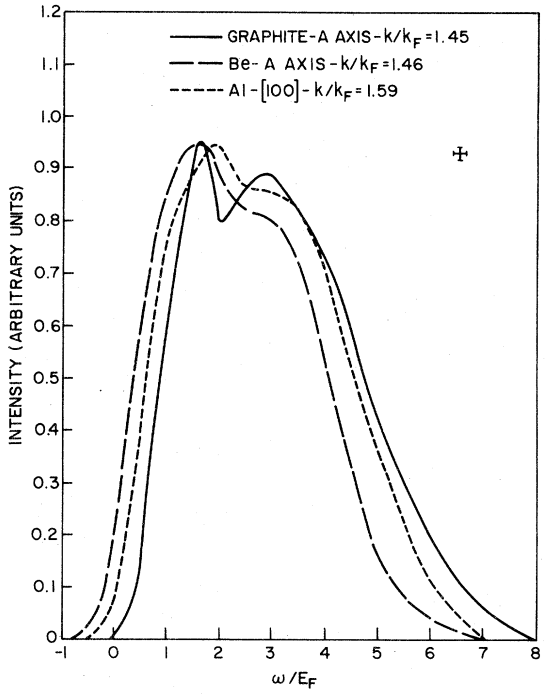

FIG. 2. Plot of $S(k,\omega)$ ($k\cong 1.5k_F$) for Al, Be, and graphite.

tems will be more completely described in a subsequent work. The major thrust of this work is that the observed phenomenon is a general property of solid-state electron gases.

A comparison of Figs. 1(a) and 1(b) for $k/k_F = 1.13$ together with the plasmon-dispersion studies in Be in the region $0 \leq k/k_F = 1$ would lead to the conclusion made by ourselves[8] and others[1] that the experimentally observed dispersion is simply smaller than theoretically predicted. In Ref. 8 we attempted to explain this lagging behind by use of the so-called f-sum rule to relate the position of the peak $[\omega(k)]$ in the spectrum to the Fourier transform of the pair correlation function $[S(k)]$, i.e.,

$$\omega(k) = \frac{k^2/2m}{S(k)}. \tag{3}$$

The slowing down of the dispersion was attributed to the enhancement of $S(k)$ at values of k corresponding to periodicities arising from the short-range order present in the electron liquid. This kind of argument works well in liquid He where the collective phonon-roton peak does indeed dominate the spectrum.[9] In this case, however, the sequence of spectra taken at higher momentum transfers tells us that the situation is

somewhat more complicated. The data [see Fig. 1(a)] at $k/k_F = 1.4$, 1.76, and 2.10 lead to the conclusion that the spectra are more accurately described as being comprised of two portions, one broad RPA-like component and another sharper, relatively local excitation, the plasmon band superimposed upon it. As k increases the relative intensity in the plasmon band decreases until it is swallowed by the broad single-particle-like piece at $(k/k_F) \approx 2$.

The presence of negative dispersion for the plasmon-band peak in the region $\omega/E_F = 1.5$ is also observable from Fig. 1(a). That figure together with Fig. 2 of a previous work[2] give a rather complete picture of the "plasmon" dispersion in Be. Starting at $k = 0$ with a sharp plasmon peak at $\omega/E_F = 1.3$, the plasmon originally has a positive nearly k^2 dispersion. In the region $0.5 \leq k/k_F \leq 1$, the plasmon is broad; the dispersion has slowed but is still positive with peak reaching a value $\omega/E_F = 1.7$. In the region $1 \leq k/k_F \leq 2$ the plasmon-band central frequency decreases with increasing k possibly reaching a value as low as $\omega/E_F = 1.2$ before it becomes very weak in intensity and poorly defined.

It seems clear to us that qualitatively the behavior of the spectrum in this intermediate-momentum-transfer regime is roughly characteristic of an ordered electron (Wigner) solid.[7] Of course this ordering is only incipient, i.e., present on a very short time scale, and probably arises from a combination of electron-electron correlations and the coupling of the electron to the periodically distributed ion cores.

To see qualitatively how such a spectrum arises we must think of an electron as localized and the scattering (for these momentum transfers) as roughly coming from one electron at a time. The electron doing the scattering finds itself in a potential well created self-consistently by the other electrons and ion cores. The scattering from a single electron in such a well roughly consists of two parts, so-called Raman and Compton scattering.

(1) Raman scattering is discrete. It consists of the excitation of the electron to the next excited state in the well. It is, we believe, the analog of the plasmon band. In this simple picture, of course, this feature of the spectrum has no dispersion, i.e., it is strictly a local excitation. Dispersion arises from coupling between wells; in fact, it is well known that the dispersion of such a longitudinal mode in a real Wigner crystal is negative. As k increases the mode decreases in energy.

(2) Compton scattering consists of excitation of the electron to the continuum. The spectrum in this case is a broad smear whose exact shape and width is characterized by the Doppler shift associated with the motion of the electron in its ground state. This type of spectrum is roughly characteristic of the broad RPA-like portion of the spectrum.

The relative intensities in these two types of scattering qualitatively behave in the observed manner. As the momentum transfer increases it becomes more probable to excite transitions to the continuum. The Compton part, as in the experiment, soaks up the intensity. The characteristic crossover occurs when the recoil energy $k^2/2m$ exceeds the local-mode frequency ω_p. As in Fig. 1(a), this is roughly what is observed.

While this picture gives us a rather physically reasonable and qualitatively accurate picture of the experimental results, no real quantitative theory exists. The importance of these experimental results lies in the fact that they graphically point up the inadequacies of an RPA type of calculation. They clearly focus on features of the electron liquid which we knew must be present but which had not been probed before.

We would like to thank W. A. Marra for technical assistance and H. Fukuyama and W. A. Brinkman for informative discussions.

[1]D. M. Militosis, Phys. Rev. B 3, 701 (1971), N. G. Alexandropoulos, J. Phys. Soc. Jpn. 31, 1790 (1971).

[2]P. Eisenberger, P. M. Platzman, and K. C. Pandey, Phys. Rev. Lett. 31, 311 (1973).

[3]H. Raether, in *Springer Tracts in Modern Physics, Ergebnisse der exakten Naturwissenschaften,* edited by G. Höhler (Springer, Berlin, 1965), Vol. 38, p. 85; P. Zacharias, to be published.

[4]K. L. Kliewer and H. Raether, Phys. Rev. Lett. 30, 971 (1973).

[5]M. S. Haque and K. L. Kliewer, Phys. Rev. B 7, 2416 (1972).

[6]K. C. Pandey, P. M. Platzman and P. Eisenberger, to be published.

[7]See the discussion and references in C. Herring, in *Magnetism,* edited by G. T. Rado and H. Suhl (Academic, New York, 1966), Vol. IV.

[8]P. M. Platzman and P. Eisenberger, Solid State Commun. 14, 1 (1973).

[9]A. Miller, and D. Pines, Phys. Rev. 127 1452 (1962).