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A. L. Ritter

JR Dennison  
*Utah State University*

R. Jones

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## Spectral Momentum Density of Amorphous Carbon from ( $e, 2e$ ) Spectroscopy

A. L. Ritter, J. R. Dennison, and R. Jones

*Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061*

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The spectral momentum density of the valence band of an amorphous carbon film has been measured by ( $e, 2e$ ) spectroscopy. Two "bands," energy as a function of momentum, are resolved. One extends from 23 eV below the Fermi energy to about 10 eV below  $E_F$ . The other,  $\sim 9$  eV below  $E_F$ , does not disperse significantly. Although the existence of diamond bonding in the film cannot be ruled out, the bands are more suggestive of the graphite band structure.

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An important question regarding the electronic structure of amorphous solids is whether vestiges of crystalline band structure exist in the disordered phase. The wave vector is not a good quantum number when translational invariance is destroyed. But Ziman<sup>1</sup> has suggested that the electron momentum is "almost" a good number in amorphous materials in the limit that  $\bar{Q}$  goes to zero. The physical basis for this assertion is that a long-wavelength electron propagates in an effective medium which is not significantly different from the crystal lattice. A wave packet representing the electron can be constructed from a narrow distribution of momentum eigenstates. This distribution broadens as the wavelength of the electron approaches the correlation length for fluctuations in the one-electron potential. The implication of this hypothesis is that the same "band structure" should exist near  $Q=0$  in the crystalline and amorphous state of a particular material, but in the disordered phase the bands will broaden and coalesce as  $\bar{Q}$  approaches the Brillouin zone boundary. Kivelson and Gelatt<sup>2</sup> developed an effective-mass model for the electronic structure of amorphous solids which places Ziman's hypothesis on a more quantitative footing for small momentum. We have measured the spectral momentum density  $P(E, \bar{Q})$  of amorphous carbon by ( $e, 2e$ ) spectroscopy.  $P(E, Q)dE d\bar{Q}$  is the probability of finding an electron in the system with energy between  $E$  and  $E+dE$  and momentum between  $\bar{Q}$  and  $\bar{Q}+d\bar{Q}$ . We find evidence of band structure in qualitative agreement with Ziman's model. This is the first investigation of a solid by ( $e, 2e$ ) spectroscopy with sufficient resolution to observe structure in the valence band.

In constructing models for the electronic structure of amorphous solids, generally it is assumed that the local bonding of an atom to its neighbors is quite similar in the ordered and disordered phase. The theoretical analysis of carbon is complicated by the fact that it has two crystal phases, graphite with

threefold planar bonding and diamond with fourfold tetrahedral bonding. Most models of  $a$ -carbon assume that the structure is a mixture of graphitic and diamond bonding, which can vary in relative concentration depending on details of sample preparation. From resistivity measurements, and other evidence, Hauser<sup>3</sup> concluded that the bonding of carbon atoms in films prepared at low temperatures was predominantly tetrahedral, while films prepared at room temperature contained a mixture of diamond and graphitic bonding. We find that the  $Q=0$  spectrum of an  $a$ -carbon film evaporated at room temperature has features which are similar to graphite, but it is not possible to rule out the presence of diamond structure in the spectrum.

The first ( $e, 2e$ ) measurements on solids were made over a decade ago.<sup>4,5</sup> The energy resolution was insufficient to observe features in the valence band of the sample. By contrast, the application of ( $e, 2e$ ) spectroscopy to the investigation of atomic and molecular states of gaseous targets has been very successful.<sup>6</sup> The technique, in our case, consists of bombarding a thin foil with electrons (25 keV incident energy) and detecting in coincidence at  $\sim 45^\circ$  scattering angles the scattered and recoiling electrons which exit from the back of the film. The binding energy of the target electron can be determined from the measured energies of the scattered and recoiling electrons. The precollision momentum of the target electron can be reconstructed from the scattering angles. We can sweep a range of binding energy from 0 to 80 eV and of momentum from 0 to  $5 \text{ \AA}^{-1}$  (in units of  $\hbar$ ) by varying respectively the transmission window and the position of the two analyzers. The direction of  $\bar{Q}$  is perpendicular to the scattering plane and in the plane of the film. In the Born approximation, the cross section for ( $e, 2e$ ) scattering is proportional to the product of the Mott electron-electron cross section and  $P(E, \bar{Q})$ . We assume that the probability of leaving the sample in an excited state after the

collision is negligible. In our scattering geometry the Mott cross section varies by less than 0.5% over the range of momentum covered in this experiment. The validity of the Born approximation at high energy ( $E \geq 2$  keV) for gas targets has been verified by experiments on simple atomic systems where the spectral density can be calculated with high accuracy.<sup>7,8</sup> In the case of atomic hydrogen,<sup>7</sup>  $P(E, \vec{Q})$  can be calculated exactly and perfect agreement is found, within experimental error, between theory and experiment. More details about  $(e, 2e)$  scattering (including a discussion of sample stability under relatively high beam currents) and a description of our spectrometer are given by Ritter, Denison, and Dunn.<sup>9</sup>

The samples for this experiment are carbon foils, approximately  $100 \text{ \AA}$  thick, prepared by Arizona Carbon Foil. The conductivity of the films is  $\sim 0.5$  mho/cm at room temperature. The coincidence rate at six momenta as a function of binding energy relative to the Fermi energy is shown in Fig. 1. We took data at  $Q = -3 \text{ \AA}^{-1}$  also, but saw no structure in the spectrum. The statistical error bars for the data are within the diameter of the data points. The highest peak ( $E_B = 23$  eV and  $Q = 1 \text{ \AA}^{-1}$ ) corresponds to a coincidence rate of 0.03 Hz. The en-

ergy resolution is  $\sim 6$  eV (full width at half maximum) and the uncertainty in the binding energy relative to the Fermi energy is  $\pm 1$  eV. The momentum resolution is  $0.5 \text{ \AA}^{-1}$  (full width at half maximum). The uncertainty in the absolute magnitude of the momentum is  $\pm 0.5 \text{ \AA}^{-1}$ , while the uncertainty in the momentum increment is  $\pm 0.2 \text{ \AA}^{-1}$ . Data have been taken on another sample at 30 keV. The statistics are worse, but the same band structure is visible.

The mean free path for an electron of energy 25 keV is typically  $\sim 200 \text{ \AA}$ . Therefore, a significant fraction of the electrons contributing to the  $(e, 2e)$  coincidence rate have suffered another collision either before or after the  $(e, 2e)$  event. The most likely multiple events are small-angle, elastic and inelastic collisions. Our measured  $(e, 2e)$  rate, then, is a convolution of the true  $(e, 2e)$  cross section with all allowed small-angle events. It is possible to deconvolute our data, since the small-angle cross section has been measured,<sup>10,11</sup> and obtain the true  $(e, 2e)$  cross section. Details of this procedure will be published later. The strongest influence on the  $(e, 2e)$  spectra in the energy range of our data is the creation of a single plasmon. Figure 2 displays our deconvoluted data. The prominent

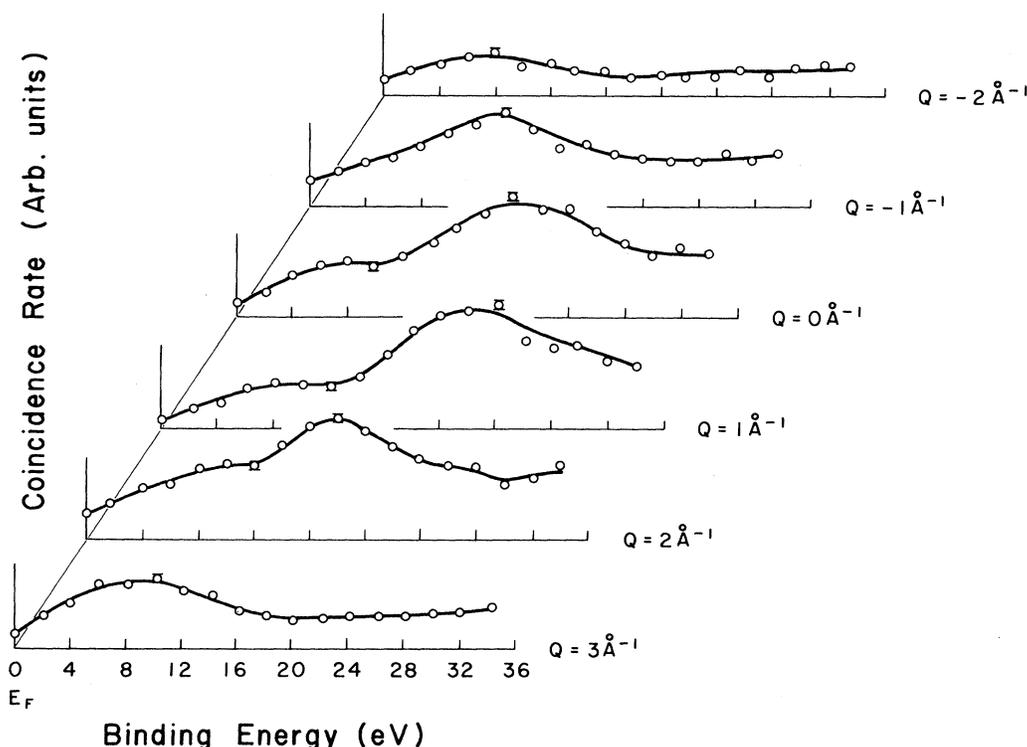


FIG. 1. The spectral momentum density of amorphous carbon as a function of energy and momentum (in units of  $\hbar$ ); uncorrected for multiple scattering.

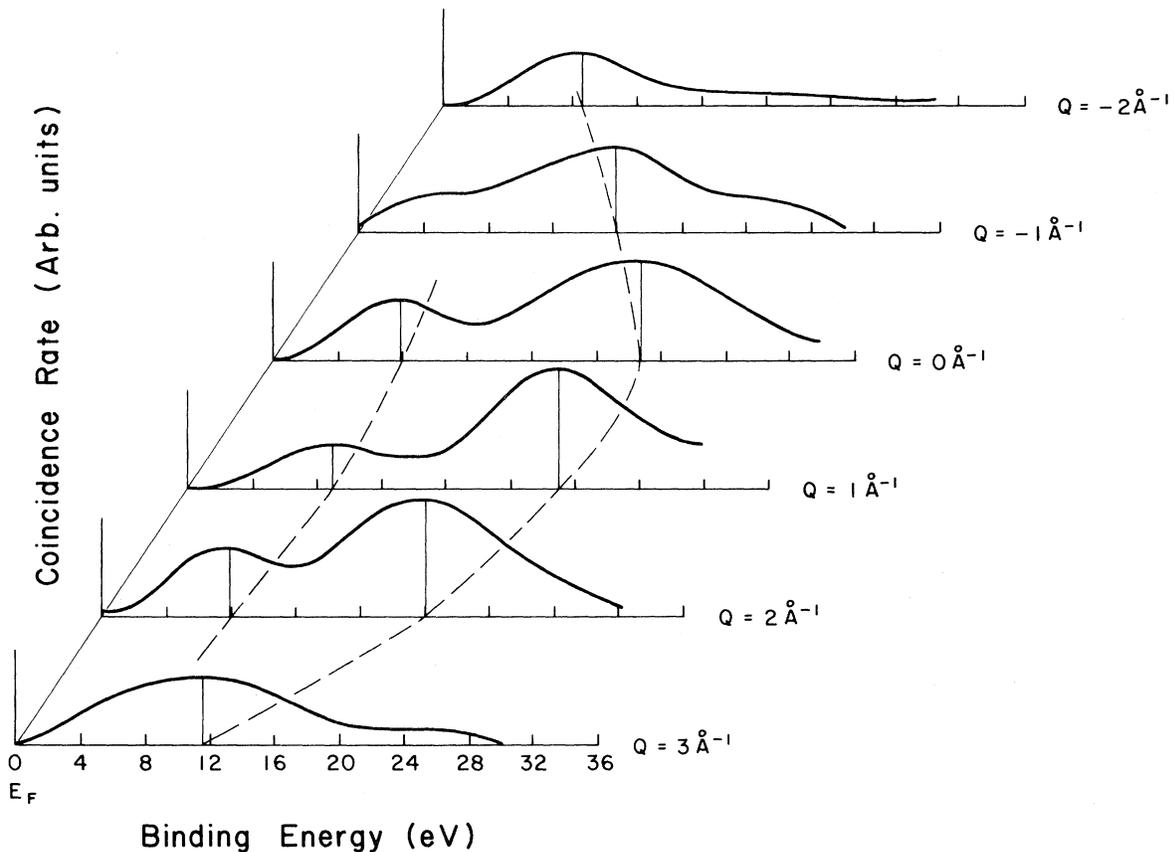


FIG. 2. The spectral momentum density of amorphous carbon as a function of energy and momentum (in units of  $\hbar$ ); corrected for multiple scattering.

band in the spectra disperses from 23 to  $\sim 10$  eV and is symmetrical about a nominal momentum between 0 and  $+1 \text{ \AA}^{-1}$ . The spectral density is maximum at the center of this band and falls off at larger  $\bar{Q}$ , which implies that the band has  $S$ -symmetry character. The width of the dispersing ridge is narrowest at  $Q=0$ . Another band can be seen at  $\sim 9$  eV which does not appear to disperse significantly. The spectral density of this band may have a minimum between nominal momenta of  $Q=0$  and  $Q=1 \text{ \AA}^{-1}$ , which would suggest that it arises from  $P$ -symmetry atomic orbitals. As will be discussed next, this peak in the spectral density may not be a single band.

The band structures of graphite and diamond have been measured by angle-resolved photoemission and good agreement between theory<sup>12,13</sup> and experiment<sup>14,15</sup> is found. The strongly dispersing band in our data is qualitatively similar to the lowest valence band of both the diamond and graphite structures. The interpretation of the upper band in our data is less clear. There are at least three alter-

natives:

(1) The bonding in the film is almost exclusively graphitic. There is an upward dispersing  $\pi$  band in graphite which crosses the  $Q=0$  axis at  $E \approx 8$  eV and two downward dispersing  $\sigma$  bands crossing at  $E \approx 5$  eV. The spectral densities of the two  $\sigma$  bands are small at  $Q=0$  because of the  $P$  symmetry of these states. The band at 9 eV in our data may be the graphite  $\pi$  band. The difference in intensity between this band and the dispersing band may arise because the  $\pi$  orbital is more strongly localized in real space. Moving away from  $Q=0$ , the upper three graphite bands may coalesce, as a result of disorder, and appear as a single band in amorphous carbon.

(2) The bonding in the films is a mixture of graphitic and diamond structure. The lowest band in our data in this case has contributions from both phases. The spectral density of the upper three diamond bands, which should cross the  $Q=0$  axis at  $\sim 3$  eV, is small for low momentum because of the  $P$  character of these states. The band at 9 eV in our

data, then, is the  $\pi$  band of graphite as discussed above. Again, moving away from  $Q=0$ , the diamond bands are smeared together with the upper three graphite bands by disorder, forming a single broad peak.

(3) There is a nondispersing band at  $\sim 9$  eV in  $\alpha$ -carbon which has no analog in the graphite or diamond band structure. Several theoretical studies<sup>16,17</sup> have shown that the  $P$ -symmetry orbitals at the top of the valence band of tetrahedrally bonded semiconductors are more sensitive to disorder than the  $S$ -symmetry orbital at the bottom of the valence band. The density of states is too large in this region of the band for Anderson localization<sup>18</sup> to be plausible; whether quasilocalized states can exist at this energy will require further experimental and theoretical study.

In order to distinguish between these alternatives, we are presently taking data at more closely spaced momentum intervals, improving the energy resolution, and beginning a study of crystalline graphite.

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