Probing the Structure of an Amorphous Solid: Proof from Compton Scattering Measurements that Amorphous Carbon is Predominantly Graphitic

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Even though a thin film of a solid may be 'structureless' using the criteria of *X*-ray and electron diffraction, the nature of the chemical bonding within the film can still be deduced from a profile analysis of Compton scattered electrons; Compton profiles can be measured using a conventional electron microscope fitted with an electron spectrometer of modest resolution.

When a solid film is amorphous, how can one ascertain the nature of the chemical bonding between the constituent atoms? Take, for example, amorphous elemental carbon. Films of this material yield rings in *X*-ray or electron diffraction that are so diffuse that it is impossible to determine whether the atomic structure is graphitic or adamantine so that, in turn, it is not feasible to infer whether sp^2 or sp^3 bonding predominates. We have shown elsewhere, however, that (i) the profile of the Compton scattered electrons, in view of their relationship to the momentum density of the electrons in the scattering material, reflects the ground-state wave-functions of the constituent electrons[1] and (ii) Compton profiles can, subject to certain compliable provisos, be reliably extracted from microregions of thin specimens using a conventional electron microscope fitted with an electron spectrometer.[2]

The Compton profile $J(p_z)$ is defined as the projection of the momentum density $\rho(p)$ onto the scattering vector, p_z [see equation (1)], and this function can be determined

$$J(p_z) = \iint_{-\infty}^{+\infty} \rho(p) \mathrm{d}p_x \mathrm{d}p_y \qquad (1)$$

directly from experimental Compton profiles. The analysis of Compton profile data is greatly facilitated if one utilizes the

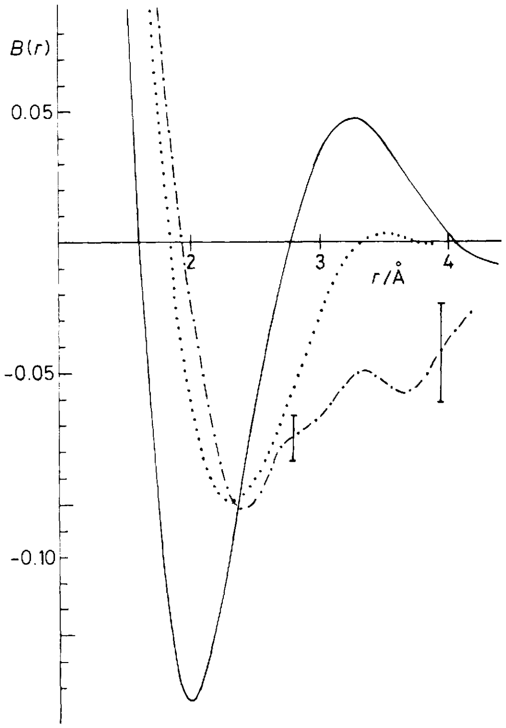

Figure 1. Reciprocal form factors (*B*) for diamond (——), graphite (····), and amorphous carbon (·—·—·—·). The error bars give the standard deviation of the data and the increase with increasing *r* arises from the deconvolution procedure.

so-called reciprocal form factor *B*(z), which is the one dimensional Fourier transform of the Compton profile. It is easy to show[3] that this is the three-dimensional Fourier transform of the momentum density (in the direction p_z) and hence the auto-correlation function of the ground-state wave-function *i.e.* equation (2). The reciprocal form factor is

$$B(z) = \int_{-\infty}^{+\infty} e^{ip_z \cdot z} J(p_z) dp_z$$

$$= \iiint_{-\infty}^{+\infty} \psi(r-z)\, \psi(r) dr \qquad (2)$$

therefore the overlap of the ground state wave-function with itself as a function of distance (*r*). On this basis it has recently been shown[4] that the formation of carbon–carbon bonds from hybrid s and p orbitals produces a negative auto-correlation in the reciprocal form factor at distances related to the bond length and that the amplitude of this correlation is governed by the hybridization state of the atomic orbitals.

To calculate the Compton profile of amorphous carbon *ab intio* is not an easy task but we can make use of the known Compton profiles of powdered diamond and graphite[5] to ascertain the valence state of amorphous carbon. Their reciprocal form factors are compared in Figure 1 with the reciprocal form factor for amorphous carbon obtained from our measurement using an electron microscope. From the reciprocal form factor at about 2 Å, which we have shown depends on the electronic structure of the bond, it is clear that the bonding in amorphous carbon is more like graphite than diamond in its electronic structure. The differences in the reciprocal form factors in the region 3 to 4 Å reflect the interactions between bonds. However, the low resolution thus far attained in both *X*-ray and electron Compton scattering from solids makes the interpretation of the data in this region unreliable. It is clearly necessary to collect data at higher resolution and to model these interactions in the hope of elucidating further the electronic structure of amorphous carbon and other solids.

As in the preceding, related communication,[6] it is helpful to describe the principle involved here in analogical terms. Essentially, what is accomplished is a measurement of the momentum of the valence electrons, and this is done by using high energy electrons as probes to evaluate the magnitude of the momentum which, in turn, tells us the nature of the bonding.

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