

ANISOTROPY OF MOMENTUM DENSITY IN CRYSTALLINE C₆₀

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Introduction

C₆₀ molecule is the most symmetric and stable of the many allotropes of carbon and has been subject of intense experimental and theoretical studies in the recent years.¹⁻² Since most of the theoretical studies of electronic structure of C₆₀ have been carried out using the density-functional-theory and the local-density-approximation (LDA),³⁻⁴ it is important to test the applicability of these approaches, as well as the quality of the specific calculation, by a direct experimental test of the ground-state charge density, which within LDA approximation is related to the Fourier expansion of the one-electron wavefunctions. Compton scattering measurements have been shown to provide such a probe, especially for the related graphite based compounds.⁵⁻⁶ Two important reasons lead to choice of Compton scattering for investigation of synthetic materials: first, it is a bulk measurement and second, due to incoherent nature of the process, it is insensitive to defects in solids.

In this work, we have carried out a combined experimental and theoretical investigation of solid C₆₀ as well as for graphite as a reference material. The wave functions have been used to calculate Compton profiles which are compared with measured profiles.

Method

Compton scattering involves the inelastic scattering of photons by electrons. The conservations of energy and momentum lead to a relationship between the energy lost by the photon and the projection of the initial electron momentum along the scattering vector **K**. The component of energy loss which results from scattering by electrons at rest is referred to as the Compton shift. The motion of electrons in the system results in a Doppler broadening of the distribution around the Compton shift and is a direct indicator of their initial momentum distribution.⁷ Directional Compton profile (DCP) for the valence elec-

trons can be written as: $J(p_z) = \int n(\mathbf{p}) dp_x dp_y = J(q)$ where $n(\mathbf{p})$ is the electron density obtained from the wavefunction $\chi(\mathbf{p})$ and $q=p_z$ is the projection of electron momentum on the scattering direction.

Experiment

Compton profile on a sample of C₆₀ powder has been measured using the focusing high resolution spectrometer⁸ at LURE (French Synchrotron Facility, Orsay). Synchrotron radiation beam is monochromatized to select 20 keV photons. The resolution function is 0.16 a.u. of momentum. For comparison, directional Compton profiles have been measured on an highly oriented pyrolytic graphite.

Measurements on C₆₀ single crystal were carried out with a similar spectrometer installed on the main arm of the κ diffractometer, at ESRF (European Synchrotron Radiation Facility, Grenoble) on beamline 11.⁹ Optics are designed in order to obtain a small spot of 0.2x0.2 mm² size on the sample, which is an ideal situation for studying small crystals such as C₆₀ ones. Resolution function is the same as the one described above. Profiles for 8 different directions of scattering vector **K**, with respect of crystallographic directions, and two different temperatures (above and below phase transition temperature) were measured.

Theory

Ab initio, all-electron SCF energy band structure calculations were carried out for solid C₆₀ within the local density approximation using the LCAO method with a Gaussian basis.¹⁰ The ground-state wave functions were then expanded in plane waves to be used in the calculation of directional Compton profiles. Each (DCP) is obtained by the summation over the 120 occupied valence bands of crystalline C₆₀.¹¹ The large size of the unit cell in real space leads to a very small Brillouin zone (BZ), thus a mesh of 11 **k** points in the irreducible sector (1/24) of the

BZ was sufficient for accurate integration. On the other hand, the number of G 's (reciprocal lattice vectors) required to achieve convergence is related to the size of the primitive unit cell and also increases with the hardness of pseudopotentials. Since the magnitudes of the reciprocal lattice vectors were very small here, it was necessary to keep 64 000 components in the sum over G in order to achieve convergence. In contrast, for graphite, 45 k points were used in the IS and it was sufficient to retain 2200 terms in the sum over G .

Results and Discussion

In Fig. 1, experimental and theoretical results for the difference between the C_{60} powder profile and the graphite profile, measured with scattering vector parallel to the c axis are compared. An advantage of this representation is that the effects of the systematic errors are canceled out, leading to an accurate picture of electron density in the solids. It is seen that the agreement between the calculated and measured differences is excellent.¹²

The observed narrowness of the C_{60} powder profile with respect to graphite profile can be interpreted as a larger delocalisation of valence electrons in C_{60} than in graphite. This delocalisation is mainly a molecular effect: this assumption is supported by the low-temperature measurement. At liquid nitrogen temperature, the molecules are no more orientationally disordered, leading to expect stronger interball bonds in the ordered phase. The difference between the two temperatures measurements does not exceed the statistical error bar for any value of q , indicating the weakness of interball bonds.

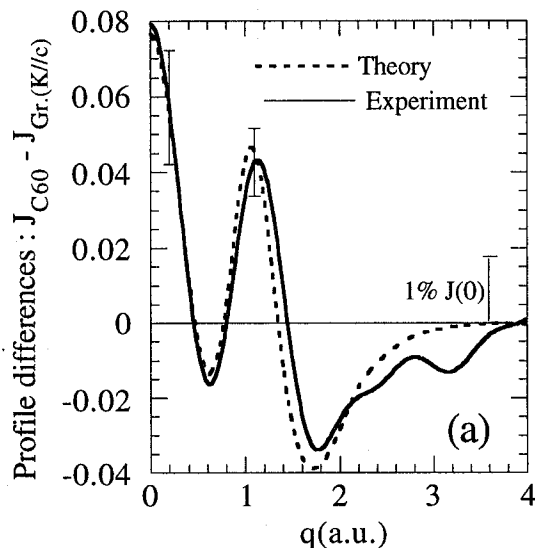


Figure 1: difference between Compton profiles of C_{60} (powered sample) and graphite

Experimental differences between two DCP of C_{60} single crystal, measured at room temperature, *i.e.* Compton

profile anisotropy, is compared with theoretical anisotropy in Fig. 2. Even if the anisotropies are very small, they are evidenced in both theory and experiment, supporting the effect of closest neighbors on the electronic density of each ball. In addition, the measured anisotropies are temperature dependent.

From the 8 measured profiles on C_{60} single crystal, the reconstruction of a 3D electron momentum density map is in progress.

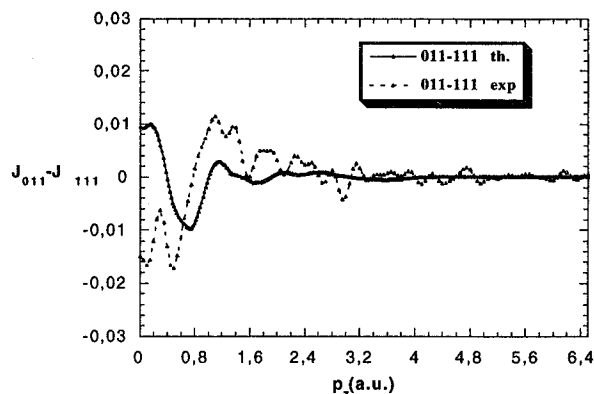


Figure 2: anisotropies between 011 and 111 directions

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