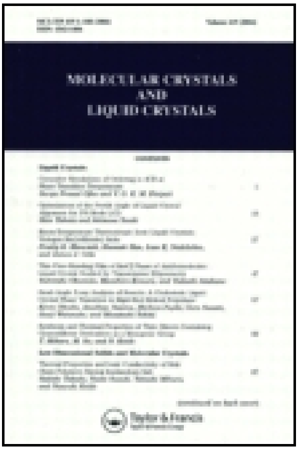

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Momentum density of electrons in $CsRb_2C_{60}$, versus temperature

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Momentum density of electrons in $CsRb_2C_{60}$, versus temperature

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The electronic momentum density in C_{60} and $CsRb_2C_{60}$ are measured as a function of temperature from below T_c to room temperatue. The effect of the superconducting transition on the momentum distribution was found to be minimal and the effect of intercation appears to agree with a rigid band picture.

Keywords: fullerenes; band structure calculations; x-ray Compton scattering

INTRODUCTION

Intercalating C_{60} with electron donors, such as the alkali metals and alloys, leads to type II superconductors with relatively high transition temperatures[1]. For the fullerides that are stable at normal pressure, T_c increases monotonically with the lattice parameter. Thus $CsRb_2C_{60}$, with the largest lattice constant among the fullerides that are stable at normal pressure, is found to have the highest T_c (>31 K).

Compton scattering measurements have been shown to provide an accurate test of the conduction and valence electron densities, particularly in the case of graphite based compounds[2–7]. Furthermore, the insensitivity of inelastic scat-

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* Correspondence Author

tering to crystalline defects makes this method well-suited for understanding of bonds in solids with the poor order such as in synthetic materials.

COMPTON SCATTERING

Compton scattering involves the inelastic scattering of photons by electrons (Fig. 1). Conservation of energy and momentum leads to the following relationship for the shift of the wavelength of photons as a result of the scattering process:

$$\Delta\lambda = \frac{2h}{mc}\sin^2\left(\frac{\phi}{2}\right) + \frac{2\lambda_1}{mc}\sin\left(\frac{\phi}{2}\right)\mathrm{pz}$$

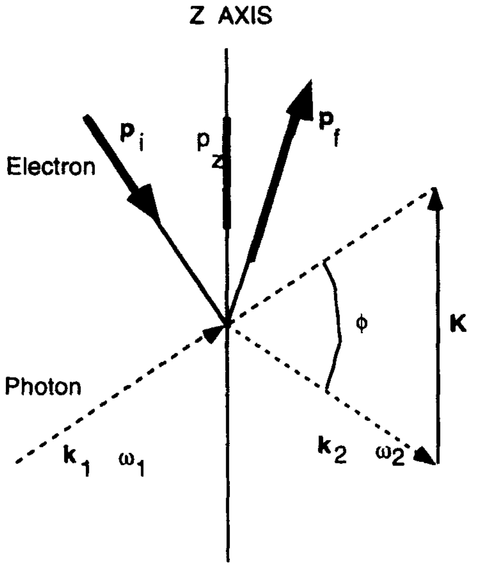

FIGURE 1 Compton scattering of a photon by an electron; subscripts i, 1 and f, 2 refer to the initial and final states respectively

Within the impulse approximation, the directional Compton profile is defined as follows [8,9]:

$$J(q,\mathbf{e}) = \int n(\mathbf{p})\delta(\mathbf{p}.\mathbf{e}-q)d\mathbf{p} = \int \chi(\mathbf{p})\chi^*(\mathbf{p})\delta(\mathbf{p}.\mathbf{e}-q)d\mathbf{p}$$

where **e** is the unit vector along the scattering vector and q is the projection of the initial electron momentum along **e**. n(**p**) is the electron momentum density and

$\chi(\mathbf{p})$ is the wave function of the electron in momentum space, i.e., the Fourier transform of the wave function in real space.

THEORETICAL APPROACH

Since the *ab initio* wave functions for $CsRb_2C_{60}$ are not available at this time and since, to the first order, the population of the π-like bands of C_{60} by the s electrons of the alkali atom follow a rigid-band model, we used the theoretical profile of K_3C_{60} for the purpose of comparison. *Ab initio* self-consistent energy band calculations were carried out for K_3C_{60}, within local-density functional theory 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 calculations of Directional Compton Profiles (DCP). If the wave functions are represented by their plane-wave expansion,

$$\Psi_{n,\mathbf{k}}(r) = \sum_{\mathbf{G}} C_{n,\mathbf{k}}(\mathbf{G}).e^{i(\mathbf{k}+\mathbf{G}).\mathbf{r}}$$

where **G**'s are reciprocal lattice vectors, the DCP can be written as [11]:

$$J(q, e) = \frac{1}{N}\sum_{n}\sum_{\mathbf{k}}\sum_{\mathbf{G}}|C_{n,\mathbf{k}}(\mathbf{G})|^2\delta[(\mathbf{k}+\mathbf{G}).\mathbf{e} - \mathbf{q}]\theta(E_n - E_f).$$

The summation **G** is over all the reciprocal lattice vectors where the $C_{n,\mathbf{k}}(\mathbf{G})$ is non-negligible. The number of G's required to achieve convergence is related to the size of the primitive unit cell. In our experience, it has ranged from 2,000 to 64,000 vectors respectively for graphite and fullerenes. Summation **k** is over the symmetry-reduced sector of the Brillouin Zone (BZ) and is carried out by calculating the wave functions over a grid of **k**'s in the BZ and dividing its volume into tetrahedra. Within each tetrahedra, a linear interpolation is carried out for $|C_{n,\mathbf{k}}(\mathbf{G})|^2$. Summation n is over the occupied states. The function θ cuts off this summation at Fermi energy in the case where the material is a metal or a semi-metal. Since the measurements are made on a powder sample, the comparison is made to the directionally averaged theoretical profile (obtained by averaging four directional profiles).

EXPERIMENTAL PROCEDURE

The experiments on $CsRb_2C_{60}$ were carried out using the Compton spectrometer of the Inelastic X Ray Scattering beamline (ID15) at European Synchrotron Radiation Facility in Grenoble, France. The synchrotron radiation beam was monochromatized to select 57 keV photons and then focused on powder sample of $CsRb_2C_{60}$. The powder was kept in a 1mm diameter Lindemann capillary under Argon atmosphere. The collected Compton spectra were energy analyzed using the 551 germanium Bragg reflection. Measurements were performed at three different temperatures: room temperature, 40K and 10K i.e. below T_c. Each spectrum achieved 10^5 counts at the Compton peak. For the purpose of comparison, the Compton profiles of C_{60} powder were also measured at 40K and room temperature under exactly the same experimental conditions. Raw data were corrected for the full energy-dependent terms such as absorption in the sample and in the analyzer, analyzer reflectivity, and relativistic effects. After the background subtraction, the spectra were normalized to the number of the electrons per carbon atom, i.e. 6 in C_{60} and 8.15 in $CsRb_2C_{60}$.

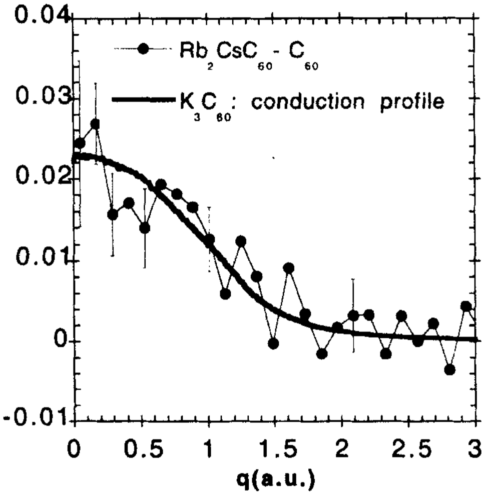

FIGURE 2 Contribution of the conduction electons and the distortion of valence momentum density in $CsRb_2C_{60}$. Measured results are compared to calculatated conduction profile in K_3C_{60}

RESULTS AND DISCUSSION

The difference between the Compton profiles of C_{60} powder at room temperature and at 40K is (for all q values) less that 1% of the value of the profile at q=0. This not only indicates a lack of temperature effect on C_{60} profile but also a lack of systematic errors in the experimental set-up such as changes in the capillary response and the beam position on the sample. The difference between $CsRb_2C_{60}$ profiles measured at 40K and 10K, i.e., above and below the superconducting transition temperature, is very small or zero. This indicates that there is no change in momentum density for electrons above and below T_c. This is expected since the number of electron involved in the conduction process is very small (three out of 489 electrons in $CsRb_2C_{60}$) and very few of them are engaged in the superconductive pair coupling.

In order to isolate the contribution of the conduction electrons and the distortion of filled valence electrons, we subtract the Compton profiles of C_{60} and of the core electrons of Rb and Cs from the total $CsRb_2C_{60}$ profile. The results are presented in figure 2. In the same figure, we show the calculated contribution to the Compton profile of K_3C_{60} from its half-occupied conduction bands.

The good agreement between the theoretical and the experimental results confirms the rigid-band picture of the heavy alkali-intercalated C_{60} compounds which have A_3C_{60} composition. This implies that the distortion of the C_{60} charge distribution is minimal and the alkali atoms simply donate theirs electrons to the relatively undistorted π-like bands of C60.

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