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CALCULATED COMPTON PROFILES : EXPERIMENTAL CHECK OF THE DEVIATION FROM THE IMPULSE APPROXIMATION IN GRAPHITE

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RESUME

Pour obtenir les profils Compton de valence du graphite, on soustrait des profils expérimentaux un profil d'électrons de cœur du carbone soit impulsional SCF soit quasi SCF. Les profils de valence ainsi obtenus diffèrent en amplitude d'environ 2% , par contre l'anisotropie reste inchangée.

ABSTRACT

To obtain the valence Compton profiles of graphite, a carbon core electron profile, either impulse SCF or quasi SCF, is subtracted from the experimental profiles. The valence profiles obtained are different by about 2% in amplitude but the anisotropy remains unchanged.

In solid state physics, the measurements of the Doppler broadening of the inelastically scattered X-rays, the Compton profiles (CP), are particularly sensitive to the behaviour of the outer, loosely bound conduction or valence electrons. The experimental CP are easily analysed in the Impulse Approximation (IA) assumption. In this case, the total CP is just the sum of all the mono-electronic CP :

$$(1) \quad J_{\text{tot}}(q_z) = \sum_i J_i = \sum_i \iint |\chi_i(\vec{q})|^2 dq_x dq_y$$

where q_z is the projection of the electron momentum on the scattering vector $\vec{k}$. $\chi_i(\vec{q})$ is the Fourier transform of the wave function of the target electron i ; no care is taken of the final state. Only this total profile is measured; so, in order to compare the experimental directional Compton profile (DCP) to the calculated one, it is necessary to subtract a calculated core profile from the first one, (Fig.1), because most of the solid state calculations lead only to the valence and conduction profiles.

Recent measurements on graphite are performed at LURE-DCI with an incident energy of 12858 eV and a scattering angle of 135°. The transferred energy at the Compton peak is only twice the ionisation threshold of the 1s carbon electron. Under these conditions, although the Impulse Approximation may be valid for the loosely bound valence electrons, this is not the case for the core ones. So, in order to analyse the experimental valence CP, the deviation from the IA has to be taken into account.

Fig.1. Total experimental directional profile ($\vec{k}$ perpendicular to $\vec{c}$) in graphite and the core impulse one that fits the total one at large q_z .

The comparison between the experimental profiles and the calculated ones, leads to a discrepancy that might be decreased by an improvement either in the calculations (1) or in the data treatment. In this comparison, we noticed that the experimental anisotropy (i.e. the difference between two DCP) is slided with respect to the calculated one (2). This might arise either from the core sp^2 hybridization, or from the deviation to the IA. To have an order of magnitude of the sp^2 hybridization effect on graphite, we have calculated the molecular orbitals of the radical CH_3 , and it was found it is similar to the carbon 1s orbital, slightly perturbed by hydrogen sites. Compton profiles calculated within the IA along the C-H axis, and two directions perpendicular, only reveals deviations (in amplitude) less than the statistical accuracy at the center of the profile.

Since the coupling with the electromagnetic field is weak, we may calculate the CP by using the lowest order Born approximation.

Then, in the independent electron and frozen core approximations, the CP - as in the impulse case - is a sum of one electron contributions :

$$(2) \quad J_{\text{tot}} = \sum_i \sum_f \left| \langle \psi_f | \exp(i\vec{k}\cdot\vec{r}) | \psi_i \rangle \right|^2 \delta(e_f - e_i - \Delta E)$$

where i and f refer to initial and final states ; e the energy of the electronic state; ΔE , the energy transferred from the photon to the electron. Different approaches have been presented to explain the deviation to the IA in the case of Compton scattering (3) or electron scattering (4).

The difficulty in calculating the CP using Eq.2 arises from the final state ψ_f of the electron. To compute the core profile of the carbon, we have evaluated numerically ψ_f using the SCF potential of the 1s state without exchange terms. This is an improvement compared to a Coulomb wave final state (5).

This quasi SCF final state leads to a Compton core profile notably different (Fig.2) compared to the impulse one :

- the maximum of the CP is at $q_{\text{max}} = -0.1$ u.a (this deviation is called the "Compton defect") ;
- the principal discrepancy comes from the amplitude of the maximum of CP which is 13% lowest than the impulse one (Fig.3).

These two effects can be due respectively to :

- q_{max} which is related to some mean value $V(\bar{r})$ of the potential experienced by the electron in its final state ;
- the variation of $V(r)$ around $\bar{r}$ is large and goes from $-Z/r$ (Z nuclear charge) for r close to zero, to $-1/r$ for r close to r_{max} ($0, r_{\text{max}}$ are the integration boundaries of the transition matrix element).

Fig.2. Compton core profile in graphite: QSCF(—), IA(---).

Fig.3. Difference between IA and QSCF core profiles.

The experimental valence (or conduction) Compton profile obtained by the subtraction of this QSCF core profile from total one is :

- Symmetric, since the QSCF CP is flat and very slightly asymmetric about $q_z = 0$; the displacement of the valence Compton Peak is negligible (or cannot be evaluated because of the statistics of the experimental results). So the anisotropy of the valence electron is unchanged when subtracting core QSCF profile or impulse one;

- Decreased, in amplitude, near $q_z = 0$ and consequently increased (for normalization reasons) at large q_z as shown on figure 4.

Therefore, the so-corrected valence profile leads to an enhancement of the difference with the calculated one as illustrated on the figure 5. This discrepancy is discussed in term of correlation effects (5).

Fig.4. Experimental valence CP after subtracting :
QSCF core profile (—);
IA core profile (---).
The error bar is the order of magnitude of the thickness of of the line.

Fig.5. Difference between calculated valence profile(1) and experimental one corrected with QSCF core profile (—) and IA core profile(---).

In the case of carbon atom the overlaps between the QSCF ψ_f and ψ_{1s} , ψ_{2s} , ψ_{2p} ; atomic orbitals are respectively $6.8 \cdot 10^{-6}$, $1.2 \cdot 10^{-6}$, $1.6 \cdot 10^{-3}$ at $q_z = q_{\text{max}}$ and decrease with increasing q_z ; so the exchange terms are probably not very important. Although the core^z graphite have the same behaviour than the 1s carbon one, this is not the case for the valence electrons. So, to evaluate the coulombic part of the potential in graphite, one should use Bloch waves describing valence electrons instead of atomic orbital (2s, 2p). A question then arises whether this latter description changes the atomic potential and thus the core profile for graphite.

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