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Compton profiles of aluminium and silicon

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Abstract. Compton profiles of aluminium and silicon have been measured using 29.5 keV synchrotron radiation x-rays with a momentum resolution of 0.084 au. Comparison of the profiles with the corresponding angular correlation curves of positron annihilation radiation gives us for the first time direct information about the effect of the positron on real interacting electron systems.

1. Introduction

It is well known that measurements of energy profiles of Compton-scattered x-rays or γ-rays in matter are in principle complementary to angular correlation measurements of positron annihilation radiation; the former provides information about the true electron momentum density while the latter gives the momentum density sampled by a positron. The positron annihilation technique has been much more widely used than Compton profile measurements for studies of electronic structure of metals and alloys because it has many practical advantages. Examples are better momentum resolution (0.05–0.1 au), two-dimensional information on the momentum density, laboratory sources of adequate intensity, and practically no limitation on the atomic number of samples. However, the positron annihilation technique has an inherent disadvantage in that the effects of the electron–positron many-body interaction and the spatial behaviour of the positron wavefunction are never known exactly in real electron systems. Therefore, measurements of the true electron momentum density, especially those with a momentum resolution equivalent to that achieved in positron annihilation experiments, are indispensable for studying the effects of the presence of the positron on real electron systems.

The differential cross section for Compton scattering can be written using the Compton profile $J(p_z)$, which is defined as the double integral of the electron momentum density $n(\boldsymbol{p})$. Thus

$$d^2\sigma/d\Omega\, d\omega_f = F(\omega_i, \omega_f, \boldsymbol{k}_i, \boldsymbol{k}_f, \theta, p_z)J(p_z) \tag{1}$$

with

$$J(p_z) = \iint n(\boldsymbol{p})\, dp_x\, dp_y \tag{2}$$

where the explicit form of the function F is given in Ribberfors (1975), ω_i and ω_f are respectively the energies of the incident and scattered x-rays, $\boldsymbol{k}_i$ and $\boldsymbol{k}_f$ their momenta, θ the scattering angle, $\boldsymbol{p}$ the electron momentum, and the z axis is taken along the scattering vector $\boldsymbol{k}_f - \boldsymbol{k}_i$. Conservation of momentum and energy give the explicit expression for p_z

$$p_z = mc[\omega_i - \omega_f - \omega_i\omega_f(1-\cos\theta)/mc^2](\omega_i^2+\omega_f^2-2\omega_i\omega_f\cos\theta)^{-1/2}. \tag{3}$$

The Compton profile $J(p_z)$ can be obtained by measuring the differential cross section for a fixed incident x-ray or γ-ray energy as a function of ω_f at a fixed scattering angle θ. A typical set-up to measure Compton profiles consists of a γ-ray source and a solid state detector. The overall momentum resolution of such a set-up is mainly limited by the energy resolution of the solid state detector, which at best yields 0.4 au on the momentum scale (Pattison and Schneider 1979, Eisenberger and Reed 1972). The possibility of high-resolution measurements using monochromatised synchrotron radiation was first explored by Loupias and Petiau (1980). The results obtained by them clearly indicated that synchrotron radiation with enhanced intensity in the high-energy region of the photon spectrum, made available by incorporation of a wiggler, would make Compton-profile measurements truly complementary to the angular correlation measurements.

In this paper, Compton profiles of single crystals of aluminium and silicon measured with a momentum resolution of 0.084 au, which is the highest resolution so far achieved, are compared with the corresponding one-dimensional angular correlation of positron annihilation radiation. The qualitative differences between the Compton profiles and the angular correlation curves are discussed.

2. Experimental procedure

An x-ray spectrometer consists of a bent-crystal monochromator, a Cauchois-type bent-crystal analyser and a position-sensitive detector. Details of the spectrometer and its performance characteristics are described elsewhere (Shiotani *et al* 1989). Further improvements in the momentum resolution and detection efficiency have been made by employing a photo-stimulatable phosphor film, the so-called imaging plate, as a position sensitive detector (Itoh *et al* 1989). Compton profiles of single crystals of aluminium and silicon were measured using 29.5 keV x-rays. The aluminium sample has a thickness of 1.8 mm and the silicon sample has a thickness of 1.5 mm. The profile of aluminium was measured with p_z along the [111] direction and that of silicon with p_z along the [110] direction. From the full width at half maximum (FWHM) of the elastically scattered x-ray peak at 29.5 keV the overall momentum resolution was found to be 0.084 au. Total accumulated Compton events were 1.2×10^7 for aluminium and 9×10^6 for silicon. The raw profiles were corrected for background noise, the energy-dependent efficiencies of the analyser and the detector, absorption in the sample, and the energy-dependent, Compton-scattering cross section of Ribberfors' formula (Ribberfors 1975).

3. Results and discussions

In order to separate the conduction-electron part from the observed profile, we assume that the core-electron contributions are well represented by the core profiles computed from atomic Hartree–Fock wavefunctions by Biggs *et al* (1975). The validity of this assumption has been firmly established by various Compton-profile measurements with

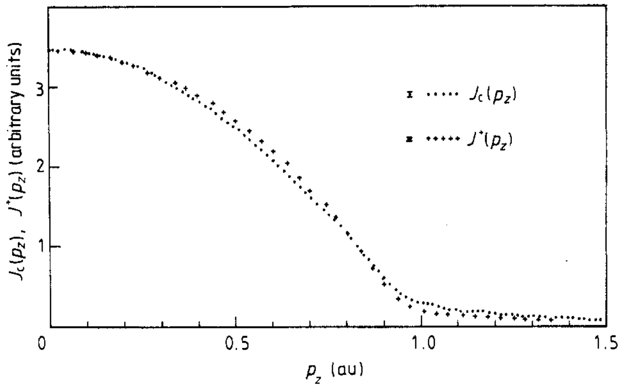

Figure 1. The conduction-electron part of the Compton profile, $J_c(p_z)$, of Al measured with p_z along the [111] direction. Subtraction of the core-electron contributions is described in § 3. This is compared with the one-dimensional angular correlation curve of positron annihilation radiation $J^+(p_z)$ measured by Okada *et al* (1976) in which the core-electron contributions are included. The two curves are normalised at $p_z = 0$. The statistical accuracy at $p_z = 0$ is indicated by an error bar.

conventional set-ups under the condition that the impulse approximation holds. Then, the core-electron contributions can be subtracted by fitting the extended tail part (p_z above a 2 au) of the observed profile to the theory. Figure 1 shows the obtained conduction-electron part of the profile $J_c(p_z)$ of aluminium with p_z along the [111] direction. The main part is *nearly* an inverted parabola. A cut-off at the Fermi momentum P_F is clearly visible around $p_z = 0.94$ au, with a distinct long residual tail above the Fermi momentum. In a free-and-independent-particle model the profile is an inverted parabola. The deviation from this simple shape and appearance of the tail above p_F in the real electron system are caused by three different effects. These are: (i) the many-body interaction between the electrons pushes a proportion of the occupied states normally below p_F to momentum states above p_F (Daniel and Vosko 1960), which produces the 'many-body' tail; (ii) due to orthogonalisation to the core-electron states, the conduction-electron wavefunctions must have high-momentum components; (iii) due to the periodic lattice potential, the electron states couple (particularly the states close to the Brillouin-zone boundaries) and thus contain high-momentum components; hence, (ii) and (iii) produce the 'high-momentum' tail.

The corresponding one-dimensional angular correlation curve of positron annihilation radiation $J^+(p_z)$ measured by Okada *et al* (1976) with a momentum resolution of 0.1 au is shown in figure 1 for comparison. The two curves are normalised at $p_z = 0$. The angular correlation curve contains the core-electron contributions. However, because of the exclusion of the positron wavefunction from the core region, the core-electron contribution is small. There are two distinct differences between the curves. Firstly, the main part of the angular correlation curve bulges, and secondly, despite the inclusion of a small core contribution, the tail above p_F in the angular correlation curve is less pronounced. These differences are the first clear experimental evidence for the combined effects of the electron–positron many-body correlation and the positron wavefunction on a fully interacting electron system. Carbotte and Kahana (1965) studied the effect of the electron–positron many-body correlation in an interacting electron gas. They found that in comparison with the case of a free-electron gas, the momentum-dependent

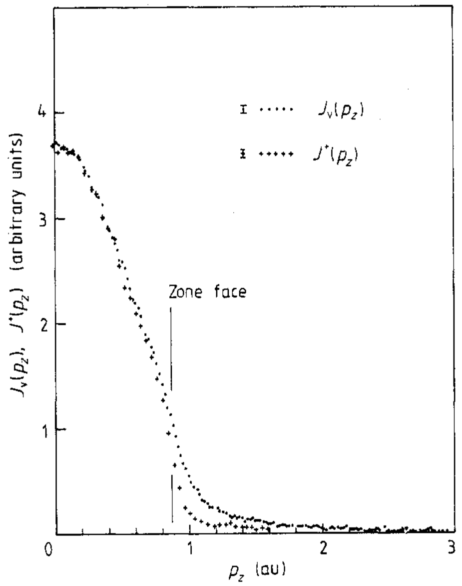

Figure 2. The valence-electron part of the Compton profile $J_v(p_z)$ of Si measured with p_z along the [110] direction. Subtraction of the core-electron contributions is described in § 3. This is compared with the one-dimensional angular correlation curve of positron annihilation radiation $J^+(p_z)$ measured by Fujiwara and Hyodo (1973) in which the core-electron contributions are included. The two curves are normalised at $p_z = 0$. The statistical accuracy at $p_z = 0$ is indicated by an error bar.

annihilation rate increases as the momentum approaches p_F, causing the bulge of the angular correlation curve, and that above p_F the annihilation rate is essentially zero, hence yielding no many-body tail. These theoretical findings are qualitatively in agreement with the observed bulge and reduced tail in the angular correlation curve in comparison with the Compton profile.

Figure 2 shows the valence-electron part of the Compton profile $J_v(p_z)$ of silicon measured along the [110] direction, subtraction of the core-electron contributions having been done in the same way as in the case of aluminium, together with the corresponding angular correlation curve $J^+(p_z)$ measured by Fujiwara and Hyodo (1973) with a momentum resolution of 0.05 au. The two curves are normalised at $p_z = 0$. As in the case of aluminium, the angular correlation curve contains a small core-electron contribution. It is immediately noticeable that the gradient at the [220] Jones-zone face is steeper and the falling-off of the tail is faster in the angular correlation curve than in the Compton profile. Although the momentum resolution of the angular correlation curve (0.05 au) is better than that of the Compton profile (0.08 au), this difference does not affect the gradient by more than 1%. Therefore, the differences shown in figure 2 are definitely due to the presence of the positron. In silicon, eight valence electrons fill the Jones zone completely and there is no Fermi surface. The effect of the electron–positron many-body interaction is naturally different from that in aluminium. Fujiwara *et al* (1972) employed a two-band model where the lower band was filled with nearly free electrons and separated from the upper empty band by an energy gap. Assuming an effective electron–positron interaction potential, they studied the effects of inter- and intraband transitions due to the electron–positron effective potential on the momentum distribution. Their results were that in the presence of the positron the gradient at the zone face becomes steeper and the falling-off of the high-momentum tail is faster than in the absence of the positron. The observed difference between the Compton profile and the angular correlation curve shown in figure 2 is therefore qualitatively in agreement with the prediction of Fujiwara *et al* (1972).

It should be noted again here that the theoretical predictions on the effects of the electron–positron many-body interaction cited above for aluminium and silicon are made in comparison with the independent-particle model in the absence of the positron, while the experimental results shown in figures 1 and 2 compare the differences between the fully interacting electron system with and without the presence of the positron. Therefore, further theoretical elaboration is certainly needed to explain the observed differences between the Compton profiles and the corresponding angular correlation curves.

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