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Electron correlation effects in Ni from Compton spectroscopy

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Abstract

We present Compton profile measurements on Ni single crystal [1 1 0] from room temperature (ferromagnetic state) up to temperatures above the Curie point of 358°C (paramagnetic state). The autocorrelation function $B(\vec{r})$, the Fourier transform of the momentum density, is used to interpret the results. A localized peak in $B(\vec{r})$, appearing in a characteristic lattice position in this direction, is diminished with increasing temperature indicating that correlation effects are gradually diminished with the transition of the material to the paramagnetic state. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Compton scattering is a very useful technique for the investigation of the electronic structure of materials, and provides sensitive test to the accuracy of band structure calculations [1,2]. A Compton profile (CP), is the projection of the electron momentum density (EMD), $n(\vec{p})$, on a direction which is defined in a Compton experiment by the scattering vector $\vec{k}$. Choosing $\vec{k}$ as the z-axis the CP is

$$J(p_z) = \int_{P_x}\int_{P_y} n(p_x, p_y, p_z)\,\mathrm{d}p_x\,\mathrm{d}p_y,$$

where $\vec{p}$ is the electron momentum. A CP is symmetric around its peak, $J(0)$, and normalized so that

$$\int_{-\infty}^{+\infty} J(p_z)\,\mathrm{d}p_z = \int_{-\infty}^{+\infty} J(q)\,\mathrm{d}q = Z,$$

where Z is the total number of scattering electrons while q is related to $\vec{p}$ and $\vec{k}$ by $q = \vec{k}\cdot\vec{p}/|\vec{k}|$.

In the case of free electrons the momentum distribution in momentum space is a sphere of radius p_{F} (Fermi momentum). The corresponding CP in this case is an inverted parabola truncated at $p_z = p_{\mathrm{F}}$. Exchange and correlation effects change slightly the inverted parabola near its edges at p_{F}.. By adjusting a characteristic crystallographic direction of a sample parallel to the scattering vector, we can investigate the EMD in this direction obtaining the so-called Directional Compton profile (DCP). This experimental CP can display directional dependence, which is usually about one percent of the peak height and can be revealed by taking the difference between two DCPs.

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In the present work, we report DCP measurements on Ni along the [1 1 0] direction for different temperatures, aiming to investigate the EMD of Ni in order to get information for correlation effects related to ferromagnetic as well as to paramagnetic state. One way of analysing the results is to take the DCPs at different temperatures. Alternatively, we can use the Fourier transformation of the CP, which provides the function $B(\vec{r})$ along the z-direction:

$$B(z) = \int J(p_z)\mathrm{e}^{-\mathrm{i}p_z z}\,\mathrm{d}p_z.$$

In other words, $B(\vec{r})$ is the three-dimensional Fourier transform of the momentum space density $n(\vec{p})$:

$$B(\vec{r}) = \int n(\vec{p})\mathrm{e}^{-\mathrm{i}\vec{p}\vec{r}}\,\mathrm{d}\vec{p}.$$

If we perform the transformation we obtain:

$$B(\vec{r}) = \sum_j B_j(\vec{r}) = \sum_j \iiint \psi_j(\vec{s})\cdot\psi_j^*(\vec{s}+\vec{r})\,\mathrm{d}\vec{s},$$

where the summation is over all occupied j-states. So, $B(\vec{r})$ is described in terms of the autocorrelation of electronic wave functions in position space and is therefore an ideal probe for the study of overlap effects in solids. This relation between the electronic wave functions and the Compton scattering data can facilitate the interpretation of experimental results even in the absence of a theoretical model.

Such a representation of CP has also a number of experimental advantages. One experimental advantage is that any residual base line error (a function of long periodicity in momentum space), becomes confined to a narrow region around the origin ($z = 0$) in position space. Data at higher z-values are therefore largely free of systematic errors. The second advantage of such a representation is that the experimental resolution appears as a straightforward multiplication with a Gaussian that suppresses the data at high values of z. The remaining data, at intermediate values of z, are relatively simpler to interpret.

2. Experimental procedure

In the present work, DCPs measurements of high statistical accuracy have been obtained on Ni [1 1 0] single crystal for different temperatures. The experimental apparatus is based on a 300 mCi ^{241}Am γ-ray spectrometer determining a scattering angle of 160° [3]. An incident beam of 59.54 keV from the ^{241}Am disc source scattered on Ni crystal was measured with a HPGe detector. The total resolution of the spectrometer is 0.54 a.u. of momentum near the peak of the CP. Sample of the form of disc with 1.2 mm thickness and 2 cm in diameter was positioned in the centre of a cylindrical furnace made of alumina where the temperature continuously monitored, was always stable within $\pm 1^\circ$C to the set point. In order to have high statistical accuracy more than 5×10^5 counts were collected at the Compton peak. The signal-to-noise ratio was 500:1. CPs were measured at four different temperatures: room temperature, 250°C, 320°C, 380°C where Curie temperature is 358°C. Data processing includes background subtraction, correction for Compton cross-section and absorption in the sample as well as correction for multiple scattering. A correction for the asymmetry introduced in the CPs by the detector response function, and source self-absorption have been applied [4].

Each CP was normalized to 24.42 electrons between -7.0 and 7.0 a.u. of momentum corresponding to the number of the electrons of the sample that can be excited with the incident photons. The electrons not excited are the 1s^2 electrons of Ni in the region from 4.1 to 7.0 a.u. since their binding energy is greater than the transferred energy.

It should be mentioned that the effect of experimental resolution on $B(z)$, the Fourier transform of the CP, is a product of the information in position space and a Gaussian damping function. It is the long-range autocorrelation of the wave functions, which is most strongly affected by the finite resolution of the experiment. The autocorrelation function at the origin, $B(0)$, simply expresses the normalization of the wave functions, i.e. for Ni one obtains $B(0) = 24.42$ electrons.

3. Results and discussion

Fig. 1 shows CPs differences for various temperatures on Nickel single crystal [1 1 0]. The differences 250–25°C and 320–25°C are essentially the same so only the second one is shown for simplicity. It gives oscillations between 1.0 and 3.0 a.u. of momentum which is just above the experimental error. The difference 380–320°C is related clearly with the passing through the Curie point (358°C) and it shows oscillations only for momentum components smaller than 2.0 a.u. The total difference 380–25°C, which is the summation of the previous two, shows more pronounced oscillations with equal amplitudes between 0.0 and 3.0 a.u. of momentum. These amplitudes are smaller than 0.01 electrons/a.u. and it is obvious that is about three times smaller than the amplitudes attributed to the anisotropy of Nickel [4]. That means that much smaller electronic changes are responsible for the transition to paramagnetic state compared with electronic differences observed between different directions.

In interpreting the results with $B(\vec{r})$ function we must have in mind the following considerations. The interesting point in the real space representation arises from localized peak in $B(\vec{r})$ centered on the sites of the translational lattice. Fig. 2 shows $B(z)$ for [1 1 0] direction around the lattice translation 4.70 a.u. where it appears a peak in $B(z)$, localized just below the lattice translation at 4.40 a.u. In the independent-particle approximation, where the electrons are described by Bloch states $\psi_{\vec{k}}(\vec{r}) = e^{i\vec{k}\vec{r}}u_{\vec{k}}(\vec{r})$, Schulke [5] has shown that, for a given band, the values of $B(\vec{r})$ at the translation lattice vector, R, is equal to the three-dimensional transform of the Fermi surface:

$$B(\vec{R}) = \frac{1}{\Omega_{BZ}} \int n(\vec{k}) \exp(i\vec{k}\cdot\vec{R})\, d\vec{k}.$$

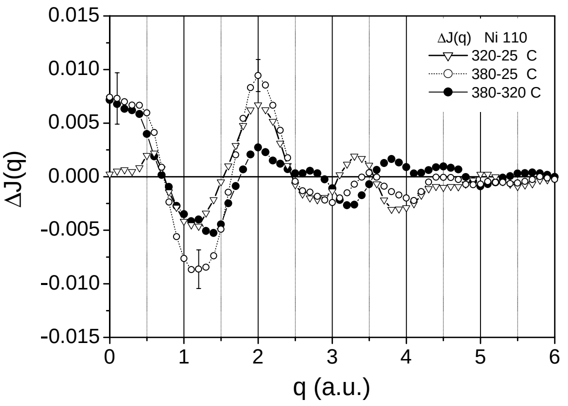

Fig. 1. Experimental CP differences for nickel single crystal in the [1 1 0] direction for various temperatures. CPs have been folded around $p_z = 0$ in order to increase the statistical accuracy. There are no any features for momentum components > 3.0 a.u.

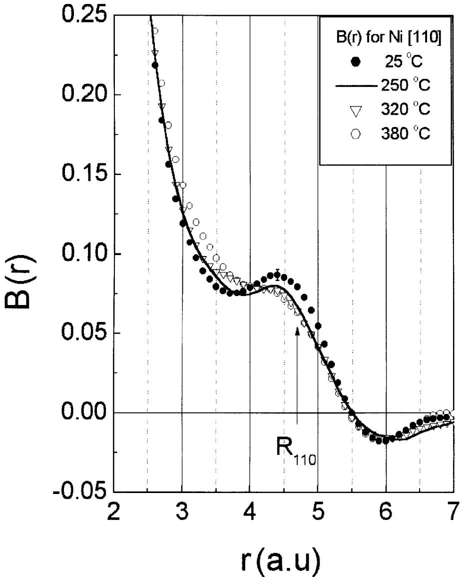

Fig. 2. $B(\vec{r})$ for nickel in the [1 1 0] direction with increasing temperature values. The position of the [1 1 0] lattice translation is indicated by an arrow. As temperature increases the localized peak is smoothed and gradually disappears.

All filled bands give zero contribution at $B(R)$ except at $R = 0$ so, in insulators for example, $B(\vec{r})$ must have a value of zero at all lattice translational vectors. This has been confirmed experimentally for Si and Ge [6]. In the case of a metal such as Nickel, an examination of $B(\vec{r})$ in the region of the lattice vector focuses attention only on the behaviour of those electrons which have an energy close to the Fermi level, i.e. the uppermost partly occupied band, because these are the electrons whose k-dependence is responsible for the shape of the Fermi surface, while filled bands give zero contribution to $B(R)$. So the above maximum is exclusively caused by the presence of localized

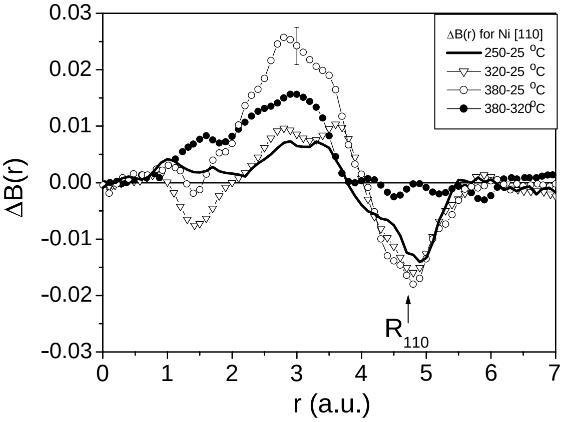

Fig. 3. Results of Fig. 2 are better represented by the difference of $\Delta B(\vec{r})$ functions. The position of the [1 1 0] lattice translation is indicated.

d-electrons in the highest energy partly occupied band.

The peak in question has been found experimentally by others, in Nickel as well as in Copper, but in Nickel it is more pronounced [7]. Here the behaviour of this peak is studied in a temperature range from ambient up to and above the transition to the paramagnetic phase. For room temperature (solid circles) this peak is very pronounced, in a height of 0.09 electrons. This value is generally the same with that found by Rollason et al. [7], taking into account that due to the different resolution functions, 0.42 a.u. of Ref. [7] and 0.54 a.u. in present experiment, our $B(z)$ is more suppressed by a factor of 1.24 in the region of lattice translation. As the temperature is raised, the peak gradually diminishes and it is a dim feature in paramagnetic state (open circles). As these secondary maxima are caused by the presence of localized d-electrons in the highest partly occupied band [1,8] we anticipate that these electrons are not so strongly correlated in paramagnetic state.

The above is shown more clearly in Fig. 3, where the differences of $B(z)$ between the various temperatures are displayed. The 250–25°C and 320–25°C differences (solid line and open triangles) are essentially the same having values only in region of real space <5.5 a.u. where $B(z)$ gives a positive overlap between the wavefunctions. The negative peak, precisely at translation lattice at 4.7 a.u., shows that the overlap at this point is diminished immediately by increasing temperature. Below 4.0 a.u. these differences take positive values, which indicates an increase of the overlap. Beyond 380°C no more changes to the overlap is observed at the region of translational lattice points, as indicated by 380–320°C difference (solid circles). On the contrary, in the range 0.0–4.0 a.u. an important increase in overlap occurs, an increase that is twice that observed in 320–25°C difference. This increase in overlap can be facilitated by the reduction of correlation effects.

4. Conclusions

CP data have been collected in various temperature ranges, including the paramagnetic state, for Nickel in the [1 1 0] direction. The results have been analyzed in real space with the aid of autocorrelation function $B(\vec{r})$. The disappearance of a localized peak at the site of the translational lattice with the transition to paramagnetic states strongly indicates the reduction of correlation effects due to the presence of localized d-electrons in the highest energy partly occupied band. The work will be completed by taking CPs in other directions for Nickel monocrystals.

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