Calculation of Compton profiles in ferromagnetic iron using LMTO wavefunctions

1989 J. Phys.: Condens. Matter 1 5363

(http://iopscience.iop.org/0953-8984/1/32/005)

J. Phys.: Condens. Matter 1 (1989) 5363–5369. Printed in the UK

Calculation of Compton profiles in ferromagnetic iron using LMTO wavefunctions

P Genoud and A K Singh
Département de Physique de la Matière Condensée, Université de Genève, 24 Quai Ernest Ansermet, CH-1211 Genève 4, Switzerland

Received 19 October 1988

Abstract. We report the calculations of magnetic Compton profiles in ferromagnetic iron. Self-consistent and parametrised band-structure calculations were performed using the linear muffin-tin orbital method. These calculations are consistent with other calculations and show a good agreement with the experimental data. Moreover the parametrised curves describe the measured magnetic Compton profiles in the low-momentum region more precisely than the self-consistent calculations. These results confirm the analysis of positron annihilation data made by Genoud and co-workers which require the use of a parametrised band-structure calculation. The main differences between both band structures concern the point N of the Brillouin zone: the large N-centred hole pocket of the minority third band given by the self-consistent calculation almost vanishes in the parametrised calculation. This trend is also supported by de Haas van Alphen measurements.

1. Introduction

In the last two years, magnetic electron Compton profiles (CP) of polycrystalline iron using x-ray (Timms *et al* 1988) and γ-ray source (Sakai and Sekizawa 1987) have been reported. These results have been compared with calculations obtained from linear combination of atomic orbitals (LCAO) (Rath *et al* 1973) and augmented plane wave (APW) (Wakoh and Kubo 1977) methods. The profiles observed experimentally exhibit a pronounced dip around $p_z = 0$. This decrease in the CP around the origin is not so pronounced in the two calculations. Moreover, experimental values do not vanish at high $\boldsymbol{p}$ ($p_z > 5$ au), when the theoretical ones do. Since both band-structure calculations do not include any self-consistent electron–electron correlation effect, it seems obvious that these many-body effects are responsible for the above discrepancies. However, the APW calculations, done using a state-dependent potential and thus accounting for electron–electron correlation in some parametric form, give an even smaller amount of dipping than the LCAO calculations. A more critical test for the band theories is provided by the recent measurement of directional magnetic CP on single crystals of iron using x-rays by Cooper *et al* (1988). It should also be mentioned that neither the magnetic electron CP, nor the total CP (Rollason *et al* 1983) can be explained on the basis of the above calculations.

Apart from the above discrepancies, some controversy exists about the Fermi surface (FS) topology of ferromagnetic iron. In a recent study of the FS of ferromagnetic iron using two-dimensional angular correlation of (polarised) positron annihilation radiation

0953-8984/89/325363 + 07 $02.50

(2D-ACPAR), Genoud *et al* (1988) have shown that there is only a very small N-centred hole pocket in the third band of minority electrons. This is not the case in fully self-consistent band-structure calculations where electron–electron correlation effects are accounted for in the local density approximation (Hedin and Lundqvist 1971). Furthermore, it was also shown that a parametrised band-structure calculation is able to interpret the experimental 2D-ACPAR data as far as the FS topology is concerned. It seems that the difficulties related to the FS topology are due to the use of local potentials in the calculations. This is confirmed by recent LMTO band-structure calculations (Barbiellini and Jarlborg 1988) making use of non local potentials, which seem to provide self-consistent data in better agreement with the positron experimental data.

The aim of this work is to confirm the results given by the positron annihilation data analysis. The interpretation of 2D-ACPAR distributions is more difficult, because the positron wavefunctions are delocalised and hence difficult to handle. In the case of CP calculations, this difficulty does not arise and the analysis is made simpler. Then we can see whether the CP (both total and magnetic) obtained from the parametrised LMTO band structure mentioned above (2D-ACPAR analysis) are closer to experiment than those obtained from other self-consistent calculations.

2. Calculation procedure

Calculations were obtained using the linear muffin-tin orbital (LMTO) band structure method (Andersen 1975, Jarlborg and Arbman 1976). The self-consistent (scalar–relativistic) LMTO band-structure calculation has been performed in the spin polarised local density approximation scheme of Gunnarsson and Lundqvist (1976) (see also Hedin and Lundqvist 1971). Temperature effects are not accounted for ($T = 0$ K). The value of the lattice constant was taken to be $a = 5.41$ au. Terms up to $l = 3$ were used in the basis set and corrections to the geometry of overlapping spheres are included. Calculation of core states was also performed (unfrozen) to achieve self-consistency. However, the contribution of core states was taken from the Hartree–Fock calculation made by Biggs *et al* (1975). It has been shown (Genoud *et al* 1988) that the self-consistent band structure could not explain some fine details observed in the experimental 2D-ACPAR spectra. These discrepancies were removed by the use of a parametrised band-structure calculation performed in the LMTO frame: the LMTO method gives us the opportunity to compute a band structure in a non-fully-self-consistent way by fixing some parameters at chosen values. The centre of gravity E_ν of each l-state is one such parameter which is normally determined self-consistently throughout the calculation. In a non-self-consistent iteration we have imposed a shifted value of the centre of gravity E_ν of the p-states, keeping the other parameters (D_ν, m_ν, etc . . .) identical to the values obtained in the fully self-consistent case. By lowering the centre of gravity of p-states, 2D-ACPAR data were correctly described as far as the FS topology was concerned. The p-character bands at point N of the Brillouin zone (BZ) are mainly affected by the parametrisation procedure: the relatively large N-centred hole pocket of the third minority band given by the self-consistent calculation has almost completely disappeared in the parametrised calculation. Moreover the majority hole arm linking H–N–H points in the self-consistent case is disconnected at point N in the parametrised band structure.

The electron momentum densities $\rho_b(\boldsymbol{p})$, contributed by the band electrons, were calculated using the procedure developed by Singh and Jarlborg (1985). Using 506 k-points in the $\frac{1}{48}$th irreducible part of the BCC BZ, $\rho_b(\boldsymbol{p})$ were computed at more than

190 000 $\boldsymbol{p}$-points inside a sphere of radius $p_{max} = 7.6$ au in the $\frac{1}{48}$th irreducible part of $\boldsymbol{p}$-space. This procedure was performed both for the majority (+) and minority (−) spins of ferromagnetic Fe. The results for $\rho_b^{\pm}(\boldsymbol{p})$ and $\rho_c^{\pm}(\boldsymbol{p})$ were used to calculate the CP for the band and core electrons, respectively, via:

$$J_{b,c}(p_z) = \int \rho_{b,c}(\boldsymbol{p})\, dp_x\, dp_y.$$

The theoretical CP $J_{th}^{\pm}(p_z)$ were obtained by adding the band and core electron contributions through the relation

$$J_{th}^{\pm}(p_z) = J_b^{\pm}(p_z) + J_c^{\pm}(p_z).$$

Adding or subtracting the total contribution of majority $(J_{th}^{+}(p_z))$ and minority $(J_{th}^{-}(p_z))$ electrons gives the final theoretical CP. These distributions are finally convolved with the experimental resolution.

3. Results and discussion

In a recent study of ferromagnetic iron through the analysis of positron annihilation data (Genoud *et al* 1988), we showed that two-photon momentum densities obtained from a fully self-consistent LMTO band-structure calculation could not describe the experiment correctly. This discrepancy is attributed to the description of the FS topology (especially near point N of the BZ), although the LMTO calculation is in very good agreement with other fully self-consistent calculations (Moruzzi *et al* 1978, Callaway and Wang 1977, Wakoh and Yamashita 1966). To verify this assumption we have performed a parametrised band-structure calculation which supports the experimental data very well, in contrast to the self-consistent calculation. This parametrised band structure is obtained by lowering the centre of gravity E_v of majority p-states by an amount of 35 mRyd and the one of minority p-states by 57 mRyd. These parameters were not chosen to optimise the calculated two-photon momentum densities, but only to remove the large minority N-centred hole pocket, as exhibited by other parametrised band structures (Jansen and Müller 1982, Nautiyal and Auluck 1985, Papaconstantopoulos 1986). The agreement with these later calculations is very good. Moreover, de Haas van Alphen measurements can be interpreted (but not unambiguously) with this kind of parametrised band-structure calculation (Lonzarich 1980). The important difference introduced by the parametrised band structure is that the hole arm in the fifth band of majority electrons linking points H–N–H of the BZ is disconnected at point N and that the N-centred hole pocket in the third band of minority electrons has almost totally disappeared. Other points of the BZ are not so drastically affected.

To confirm our positron data analysis we have performed CP calculations from both band structures and compared them with experiment and with other calculations. The comparison of directional total CP with the experimental data of Rollason *et al* (1983) and with the calculation of Rath *et al* (1973) (LCAO) and of Wakoh and Kubo (1977) (APW) is not a critical test for discriminating between the different calculations. This is shown in figure 1, where the absolute difference between theoretical and experimental CP is depicted in the [111], [110] and [100] directions. Both LMTO calculations describe the experiment quite well over the whole momentum range. At $p_z = 0$ the excess of density is smaller than for the APW calculation and is comparable with the LCAO result. In the 2–5 au momentum range the deviation from experiment lies within the error bar.

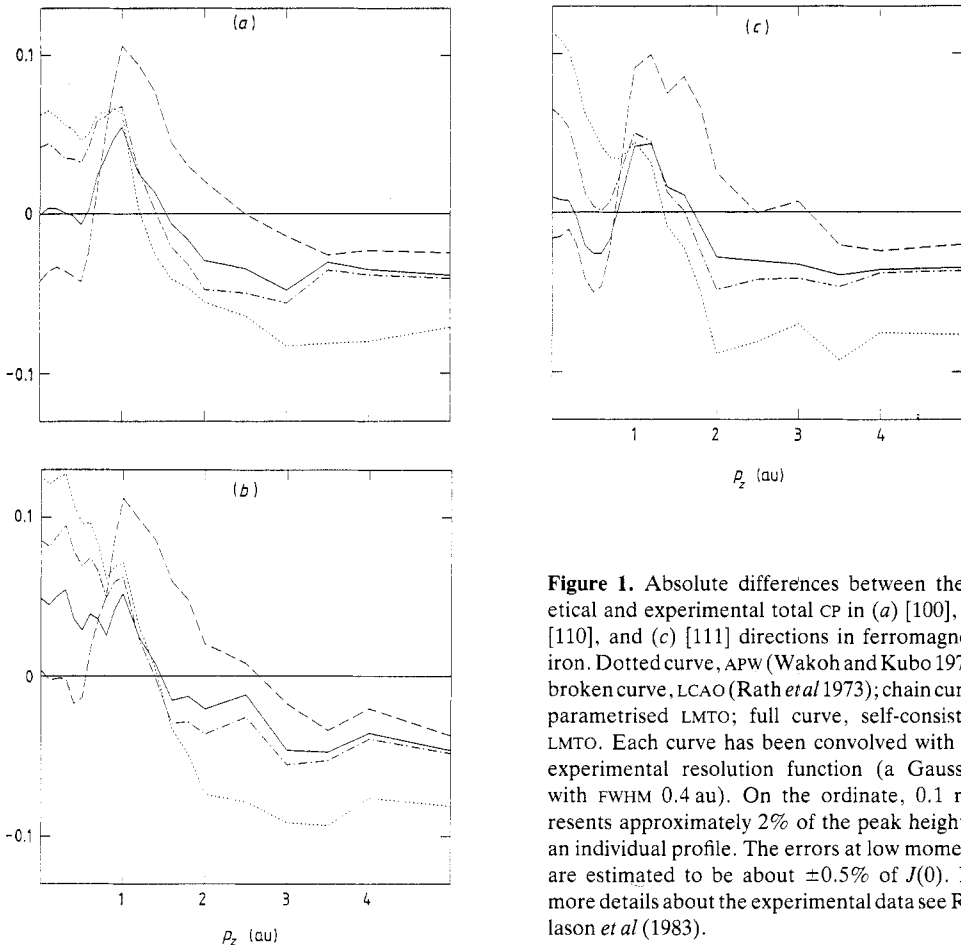

Figure 1. Absolute differences between theoretical and experimental total CP in (*a*) [100], (*b*) [110], and (*c*) [111] directions in ferromagnetic iron. Dotted curve, APW (Wakoh and Kubo 1977); broken curve, LCAO (Rath *et al* 1973); chain curve, parametrised LMTO; full curve, self-consistent LMTO. Each curve has been convolved with the experimental resolution function (a Gaussian with FWHM 0.4 au). On the ordinate, 0.1 represents approximately 2% of the peak height of an individual profile. The errors at low momenta are estimated to be about ±0.5% of $J(0)$. For more details about the experimental data see Rollason *et al* (1983).

Finally around $p_z = 2$ au there is not such a large disagreement as produced by the LCAO calculation. The directional total CP are not sensitive enough definitively to favour the parametrised or the self-consistent LMTO calculation (even if the latter fits the experiment better), because the difference between corresponding curves lies within the error bar. However it confirms, when compared with other calculations, the capability of our method (Singh and Jarlborg 1985) to provide good quality CP results.

The analysis of directional magnetic CP is much more instructive. The calculated curves in the [100], [110] and [111] directions, which have to be compared with the experimental data of Cooper *et al* (1988), are shown in figure 2. Both theoretical magnetic CP have been normalised to the number of electrons with unpaired spins given by the corresponding band-structure calculation. This number is equal to 2.26 μ_B in the self-consistent case and 2.12 μ_B in the parametrised one. For a quantitative comparison, table 1 gives the values of the profiles (experimental (extracted from Cooper *et al* 1988) and calculated (this work)) at $p_z = 0$ au and at $p_z = 1.3$ au in the [100], [110] and [111] directions. The ratio $J(1.3)/J(0)$, which is a measure of the 'crater-like' line shape in the [110] and [111] directions is also given. As in the experiment, this 'crater' (attributed to a negative polarisation of sp-like electrons hybridised with the 3d electrons) is deepest

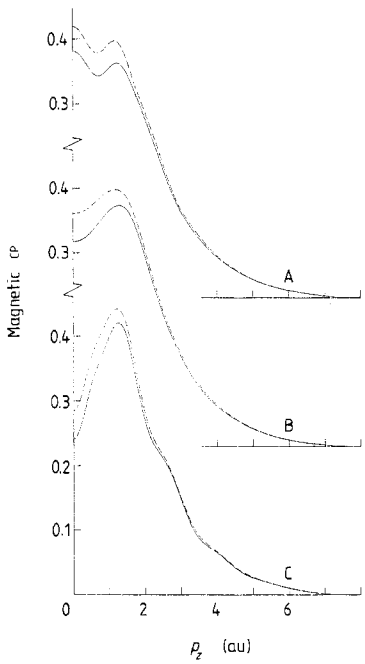

Figure 2. Calculated directional magnetic CP as functions of the electron momentum: curves A, [100] direction; curves B [110] direction; curves C, [111] direction. Broken curves, self-consistent LMTO calculation; full curves, parametrised LMTO calculation. Both curves have been convolved with the resolution function (a Gaussian with FWHM 0.7 au) and normalised to the number of unpaired spins given by the corresponding band-structure calculation (see § 3 of text).

Table 1. Values of the magnetic CP of ferromagnetic iron at $p_z = 0$ au and at $p_z = 1.3$ au in the [100], [110] and [111] directions. The ratio $J(1.3)/J(0)$ is also given. Our calculated values (self-consistent LMTO and parametrised LMTO) are compared with the corresponding experimental data extracted from Cooper *et al* (1988). The momentum $p_z = 1.3$ au corresponds (in the [110] and [111] directions) to the maximum value of the experimental curve.

		Experiment	Self-consistent LMTO	Parametrised LMTO
$J(p_z = 0)$	[100]	0.340	0.419	0.381
	[110]	0.302	0.361	0.318
	[111]	0.234	0.285	0.241
$J(p_z = 1.3)$	[100]	0.355	0.392	0.362
	[110]	0.395	0.397	0.373
	[111]	0.434	0.441	0.420
$J(1.3)/J(0)$	[100]	1.044	0.935	0.950
	[110]	1.308	1.099	1.173
	[111]	1.855	1.547	1.743

in the [111] CP and is completely absent in the [100] curve. Both calculations reproduce this behaviour. But even if the structures remain approximately the same, the self-consistent calculation overestimates the experimental value in the 0–2 au momentum range compared with the parametrised calculation. This is a confirmation of the analysis of positron annihilation data which unambiguously favours the same parametrised band-structure calculation. The directional magnetic CP obtained from the parametrised band structure are very close to experimental results in the [110] and [111] directions, but the magnetic profile is still overestimated near $p_z = 0$, especially in the [100] direction. Referring to the LMTO band results, this means that the contribution of the negatively polarised 4s–p band is underestimated. We mention finally the recent calculations

of Poulter and Staunton (1988) which were performed within the Korringa–Kohn–Rostoker coherent potential approximation (KKR CPA) scheme in order to study temperature effects. The agreement of their results with the experiment of Timms *et al* (1988) is better than those from the APW calculation of Wakoh and Kubo (1977).

4. Conclusion

The CP calculations (both total and magnetic) we have performed from both a self-consistent and a parametrised LMTO band structure match the corresponding experimental data well. The analysis of total directional CP cannot definitively favour one or the other calculation. But it confirms the capability of our method to obtain good quality CP calculations. The comparison with directional magnetic CP confirms the observation made by Genoud *et al* (1988) on positron annihilation data: the momentum densities çalculated from the parametrised LMTO band structure describe the experiment better than those obtained from a fully self-consistent LMTO calculation. This reflects an incorrect description of the FS topology at point N of the BZ. The remaining discrepancy between the experimental data and our parametrised calculation can be attributed to electron–electron correlations since the parametrised calculation models such an effect and in a smaller extent to spin–orbit coupling interactions which are neglected in the calculations.

Acknowledgment

We are extremely grateful to T Jarlborg for providing LMTO band-structure calculation and matrix elements. We also warmly thank A A Manuel and Professor M Peter for their interest in this work, which was supported by the Swiss National Science Foundation.

References

Andersen O K 1975 *Phys. Rev.* B **12** 3060
Barbiellini B and Jarlborg T 1988 private communication
Biggs F, Mendelsohn L B and Mann J B 1975 *At. Data Nucl. Data Tables* **16** 201
Callaway J and Wang C S 1977 *Phys. Rev.* B **16** 2095
Cooper M J, Collins S P, Timms D N, Brahmia A, Kane P, Holt R S and Laundy D 1988 *Nature* **333** 151
Genoud P, Singh A K, Manuel A A, Jarlborg T, Walker E, Peter M and Weller M 1988 *J. Phys. F: Met. Phys.* **18** 1933
Gunnarsson O and Lundqvist B I 1976 *Phys. Rev.* B **13** 4274
Hedin L and Lundqvist B I 1971 *J. Phys. C: Solid State Phys.* **4** 2064
Jansen H J F and Müller F M 1982 *Phys. Rev.* B **26** 2624
Jarlborg T and Arbman G 1976 *J. Phys. F: Met. Phys.* **6** 189
Lonzarich G G 1980 *Electrons at the Fermi Surface* ed. M Springford (Cambridge: Cambridge University Press) ch 6
Moruzzi V L, Janak J F and Williams A R 1978 *Calculated Electronic Properties of Metals* (New York: Pergamon)
Nautiyal T and Auluck S 1985 *Phys. Rev.* B **32** 6424
Papaconstantopoulos D A 1986 *Handbook of the Band Structure of Elemental Solids* (New York: Plenum)
Poulter J and Staunton J B 1988 *J. Phys. F: Met. Phys.* **18** 1877
Rath J, Wang C S, Tawil R A and Callaway J 1973 *Phys. Rev.* B **8** 5139

Rollason A J, Holt R S and Cooper M J 1983 *J. Phys. F: Met. Phys.* **13** 1807
Sakai N and Sekizawa H 1987 *Phys. Rev.* B **36** 2164
Singh A K and Jarlborg T 1985 *J. Phys. F: Met. Phys.* **15** 727
Timms D N, Brahmia A, Collins P, Collins S P, Cooper M J, Holt R S, Kane P P, Clark G and Laundy D 1988 *J. Phys. F: Met. Phys.* **18** L57
Wakoh S and Kubo Y 1977 *J. Magn. Magn. Mater.* **5** 202
Wakoh S and Yamashita J 1966 *J. Phys. Soc. Japan* **21** 1712