Magnetic Compton profile of nickel

1991 J. Phys.: Condens. Matter 3 3311

(http://iopscience.iop.org/0953-8984/3/19/009)

J. Phys.: Condens. Matter 3 (1991) 3311–3315. Printed in the UK

Magnetic Compton profile of nickel

V Sundararajan and D G Kanhere
Department of Physics, University of Poona, Pune 411007, India

Received 4 September 1990, in final form 21 January 1991

Abstract. Spin-dependent momentum densities in ferromagnetic nickel have been calculated by using the self-consistent spin-polarized linear combination of Gaussian orbitals (LCGO) method. The resulting magnetic Compton profiles have been compared with the recently measured experimental profiles reported by Timms *et al*. The LCGO magnetic profile along ⟨111⟩ direction disagrees with the experimental profile near the origin ($p_z = 0$), in that the central dip seen in the experimental profile is not reproduced, a feature shared by APW profile. It is argued that the overestimation of the profiles near the origin is mainly due to electron–electron correlation effects inadequately treated in the local-density approximation.

The technique of measurement of Compton profiles (CPs) has emerged as a powerful probe for investigating the electron momentum density (EMD) in transition metals. Apart from providing useful information about Fermi surface sheets, recent comparisons between theory and experiment have revealed that electron–electron correlation effects are not treated adequately in the local-density approximation (LDA) [1–3]. At the time of publication of our earlier paper on nickel [2], experimental results on the magnetic Compton profile (MCP) were not available. The availability of intense synchrotron radiation sources now makes it possible to probe the spin-dependent momentum density by using polarized photon beams [4].

Recently, Timms *et al* [5] reported a measurement of magnetic CPs using circularly polarized synchrotron radiation in single-crystal ferromagnetic nickel along the ⟨100⟩ and ⟨111⟩ directions. These measurements when compared with APW theoretical profiles revealed that, although for the ⟨100⟩ direction the agreement is good, for the ⟨111⟩ direction the central dip at $p_z = 0$ is not reproduced by APW. This feature is very similar to that observed for ferromagnetic Fe [6] where the theoretical profiles calculated by the linear combination of Gaussian orbitals (LCGO) method did not reproduce the central dip at $p_z = 0$, along the ⟨100⟩ direction. In fact, so far as the 3d metals are concerned, the major disagreement between theory and experiment is always near $p_z = 0$. Therefore, it appears that the disagreement is not due to a particular method of band-structure calculation but may be inherent in the LDA. The present work motivated by these observations reports the accurate theoretical MCP of ferromagnetic Ni by employing the self-consistent all-electron LCGO method and compares it with the measurements of Timms *et al* [5].

The LCGO method has been used extensively for calculating the electron momentum density and CPs. It offers the following advantages.

0953-8984/91/193311 + 05 $03.50

(i) Apart from being all electron and self-consistent, it makes no shape-dependent approximation.

(ii) The Gaussian nature of the basis function provides an accurate evaluation of the momentum density analytically for large values of p, giving accurate high-momentum components. The details of the procedure have been given in [6, 7].

The EMD is defined as

$$\rho(\boldsymbol{p}) = \sum_{\text{occ}} |\psi_n(\boldsymbol{k}, \boldsymbol{p})|^2 \tag{1}$$

where $\psi_n(\boldsymbol{k}, \boldsymbol{p})$ is the momentum space wavefunction given by

$$\psi_n(\boldsymbol{k}, \boldsymbol{p}) = (N\Omega)^{-1/2} \int \exp(-i\boldsymbol{p} \cdot \boldsymbol{r})\, \psi_n(\boldsymbol{k}, \boldsymbol{r})\, d^3r \tag{2}$$

with Ω the volume of the unit cell and $\psi_n(\boldsymbol{k}, \boldsymbol{r})$ is the Bloch state for wavevector $\boldsymbol{k}$.

The directional CP is given by

$$J_{\hat{k}}(p_z) = \frac{\Omega}{(2\pi)^3} \int d^3p\, \rho(\boldsymbol{p}) \delta(p_z - \boldsymbol{p} \cdot \hat{\boldsymbol{k}}) \tag{3}$$

and the MCP by

$$J^{\hat{k}}_{\text{mag}}(p_z) = \int_{p_x} \int_{p_y} [\rho_\uparrow(\boldsymbol{p}) - \rho_\downarrow(\boldsymbol{p})]\, dp_x\, dp_y \tag{4}$$

where $\rho_\uparrow(\boldsymbol{p})$ and $\rho_\downarrow(\boldsymbol{p})$ are the up and down EMDs, respectively.

We have made use of our earlier calculation reported in [2], to evaluate the MCP for Ni. A spin-polarized calculation was performed self-consistently on 89 $\boldsymbol{k}$-points and the final band structure sampled at 506 $\boldsymbol{k}$-points in a one-fortyeighth part of the Brillouin zone. The exchange correlation scheme of von Barth and Hedin (VBH) as parametrized by Rajagopal [8] was used. The basis set consisted of 13s, 10p and 5d Gaussian functions. The calculation of momentum density incorporated 1409 $\boldsymbol{G}$-vectors and the integration up to 10 au in $\boldsymbol{p}$-space accounts for 9.936 electrons which is more than 99% of the conduction electrons. The resulting directional CPs as well as their anisotropies agree well with the experimental results of Rollason *et al* [9], which provides a test for the numerical accuracy of the calculation [2]. The magnetic moment per site is 0.596 μ_B as against the experimental value of 0.56 μ_B [10]. The theoretical MCP has been renormalized to an area of 0.7 in the momentum range between -8 and $+8$ au and also convoluted with a Gaussian of FWHM 0.7 au as given in the work of Timms *et al* [5].

The MCPs are shown in figures 1 and 2 for the ⟨100⟩ and ⟨111⟩ directions (full curves), respectively, together with the experimental values of Timms *et al* [5] (histograms). It is quite clear that the agreement between the LCGO theory and experiment is quite good for the ⟨100⟩ direction. For the ⟨111⟩ direction the theory overestimates the experimental result at $p_z = 0$. The disagreement near $p_z = 0$ is similar to that seen with APW results. Thus it is clear that in the cases of both Fe and Ni the overestimation of the profile near $p_z = 0$ is common to both APW and LCGO calculations based on the LDA. It may be mentioned that the EMD and CP calculated for 3d metals by these methods agree with each other, and also with other band-structure calculations having comparable accuracy.

The discrepancy can be attributed to two effects: firstly to the inadequate reproduction of the Fermi surface and secondly to electron–electron correlation effects. From the work of Wang and Callaway [11, 12] it is clear that the LCGO with the VBH potential

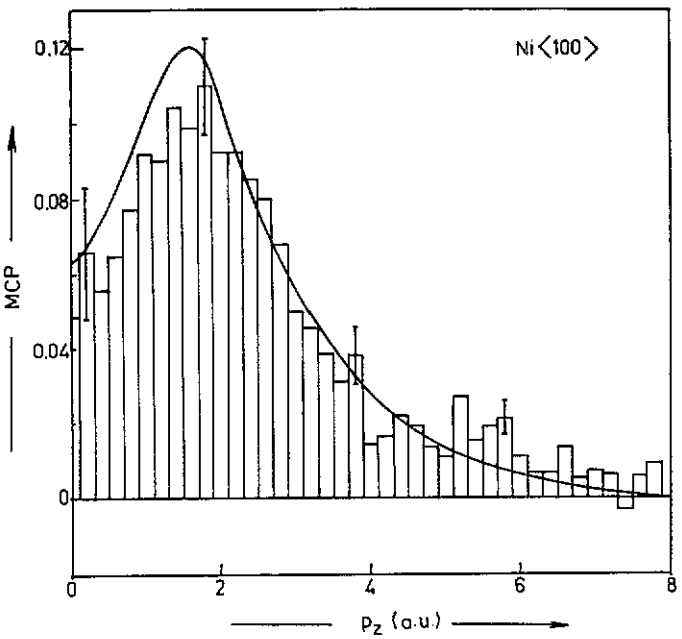

Figure 1. Theoretical (full curve) and experimental (histogram) MCP along the ⟨100⟩ direction.

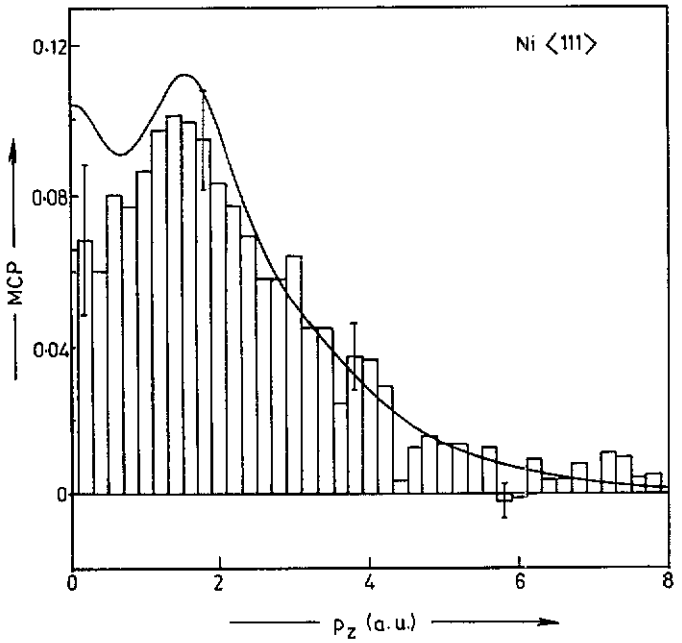

Figure 2. Theoretical (full curve) and experimental (histogram) MCP along the ⟨111⟩ direction.

adequately explains the Fermi surface cross section in Ni, but theory predicts an extra $X_{2\downarrow}$ hole pocket which has not been observed experimentally. However, this does not contribute to the momentum density [13]. Hence, one can attribute the discrepancy to be dominantly due to the electron–electron correlation effect. It is well known that the EMD and CP obtained by the LDA must be corrected by the Lam–Platzman correction. In fact, for all 3d metals, theory when compared with experiment overestimates $J_k(p_z)$ near the origin. Bauer *et al* [1] have discussed this point in detail and have shown that the profiles calculated by the LDA omit the electron–electron correlations. This correction is anisotropic. Since up and down spin densities are different, the correction would be different for up and down electrons.

There is also the possibility that non-local effects contribute to the discrepancy. Such a calculation has not been performed so far. Apart from these corrections, spin–orbit coupling, which is not included in the present work, should also be taken into account. However, in order to make a meaningful comparison, experiments with a better momentum resolution would be required.

In a recent publication of Kubo and Asano [14], the experimental MCP of Fe and Ni has been compared with theoretical results using the FLAPW method. Even though their results show less discrepancy than other theoretical methods, noticeable differences are found for Fe along the ⟨100⟩ and ⟨110⟩ directions and for Ni along ⟨100⟩. It may be mentioned that the best agreement between theory and experiment has been found for the parametrized version of the LAPW method in which the centre of the p band is shifted to obtain a correct Fermi surface. However, it is somewhat surprising to see the linearized version of the APW method giving better MCPs than the APW method does. This may be either due to the muffin-tin corrections incorporated in the LAPW method or a better description of the Fermi surface. In the light of this discussion we feel that it would be very fruitful to make a detailed comparison of CPs (in addition to MCPs) obtained by various theoretical LDA methods and experiments.

The present work demonstrates the inadequacy of the LDA in the theoretical calculations to explain the MCP. We have presented the self-consistent LCGO MCP for the ferromagnetic Ni. Comparison with experiment shows disagreement near the origin, which confirms the results of the earlier APW calculations, indicating a need for a better description of the electron–electron correlation. Moreover, experiments with better resolution would provide a platform for a critical test of theories and to resolve the above discrepancy.

Acknowledgments

The authors would like to thank Professor R M Singru for useful discussions. Financial support for the work from the Department of Science and Technology, India, and Centre for Development of Advanced Computing, Pune, is gratefully acknowledged.

References

[1] Bauer G E W and Schneider J R 1984 *Phys. Rev. Lett.* **52** 675
[2] Sundararajan V and Kanhere D G 1988 *Phys. Rev.* B **38** 12653
[3] Cardwell D A, Cooper M J and Wakoh S 1989 *J. Phys.: Condens. Matter* **1** 541
[4] Cooper M J 1985 *Rep. Prog. Phys.* **48** 415; 1989 *Physica* B **159** 137

[5] Timms D N, Brahmia A, Cooper M J, Collins S P, Hamonda S, Laundy D, Kilbourne C and Saint Lager M C 1990 *J. Phys.: Condens. Matter* **2** 3427
[6] Sundararajan V, Kanhere D G and Singru R M 1991 *J. Phys.: Condens. Matter* **3** 1113
[7] Rath J, Wang C S, Tawil R A and Callaway J 1973 *Phys. Rev.* B **8** 5139
[8] Rajagopal A K 1979 *Advances in Chemistry and Physics* vol 41, ed G I Prigogine and S A Rice (New York: Wiley) p 59
[9] Rollason A J, Schneider J R, Laundy D S, Holt R S and Cooper M J 1987 *J. Phys. F: Met. Phys.* **11** 1105
[10] Danan H, Herr A and Meyer A J H 1968 *J. Appl. Phys.* **39** 669
[11] Wang C S and Callaway J 1977 *Phys. Rev.* B **15** 298
[12] Wang C S and Callaway J 1975 *Phys. Rev.* B **11** 2417
[13] Harthorn R and Mijnarends P E 1978 *J. Phys. F: Met. Phys.* **8** 1147
[14] Kubo Y and Asano S 1990 *Phys. Rev.* B **42** 4431