Anisotropies of Compton profiles in nickel

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The Compton profiles of nickel are calculated using an *ab initio*, spin-polarized self-consistent linear combination of Gaussian orbitals band-structure method within the local-density-functional theory. The resulting anisotropies are compared with recent experimental results of Rollason *et al.* The present theoretical results, which include the correlation potential, show improved agreement with experiment as compared to an earlier exchange-only calculation, especially in the low- and high-momentum regions. However, the present accurate local-density results still overestimate the anisotropies significantly, indicating the inadequacy of the local-density-functional theory for calculating the ground-state electron momentum density.

During the last few years the technique of Compton-profile measurements has emerged as a powerful tool to probe the ground-state electron momentum density (EMD) of inhomogeneous electron systems. It is possible to test the quality of the wave functions and the adequacy of the approximations involved in a given theory by direct comparison between the theoretical profiles and the experimental ones, especially if experimental results with adequate accuracy are available. This point has been brought out by recent experimental work on single-crystal Ni by Rollason *et al.*[1] and on Cu by Bauer *et al.*[2] Their works have indicated that there are strong correlation effects which the local-density-functional (LDF) theory is unable to account for so far as the EMD is concerned.

The present work on Ni is motivated by a recent experimental report,[1] where a comparison was made with a linear combination of Gaussian orbitals (LCGO) band-structure calculation[3] which was performed with an exchange-only approximation. Moreover, there exists some doubt about the comparison beyond $q = 2.5$ a.u. because of the inadequate theoretical data above that value. Therefore, it was thought desirable to perform an accurate LDF calculation including correlation in order to have a definitive quantitative comparison.

It may be noted that all conventional LDF calculations yield the EMD corresponding to a noninteracting electron gas having the same ground-state charge density as that of an interacting electron system. This problem has been addressed by Lam and Platzman[4] who derived a correction term. However, this term in the local-density approximation (LDA) is isotropic and does not affect the anisotropies. Therefore, in the present report we confine ourselves to the discussion of anisotropies. The full profiles, EMD, spin-momentum density, and other details of our investigation will be reported separately after including the Lam-Platzman (LP) correction and core momentum density.

In what follows we give a brief outline of the computa-tional procedures, the details of which are discussed in Ref. 3 and 5. The Compton profile along direction $\hat{\mathbf{k}}$ is defined as

$$J_{\hat{\mathbf{k}}}(q) = [\Omega/(2\pi)^3] \int d^3p \, \rho(\mathbf{p}) \delta(q - \mathbf{p} \cdot \hat{\mathbf{k}}) , \qquad (1)$$

where $\rho(\mathbf{p})$ is the EMD which is defined as

$$\rho(\mathbf{p}) = \sum_{n,\mathbf{k}} |\psi_n(\mathbf{k},\mathbf{p})|^2 . \qquad (2)$$

The band structure of Ni has been obtained by the self-consistent LCGO method of Wang and Callaway[6] using von Barth and Hedin exchange correlation potential as parametrized by Rajagopal *et al.*[7] The self-consistency was achieved by using 89 **k** points in $\frac{1}{48}$th of the Brillouin zone (BZ). The Gaussian basis set consisted of 13s, 10p, and 5d functions. The resulting self-consistent potential was used to calculate the band structure and wave functions on 505 **k** points in $\frac{1}{48}$th of the BZ. The momentum distribution was calculated up to $p = 10$ a.u. using adequate number of reciprocal-lattice vectors. It may be mentioned that the Gaussian nature of the basis function allows analytic evaluation of the Fourier transforms enabling us to obtain the momentum distribution for large p values without loss of accuracy.

The resulting anisotropies after convoluting the individual profiles with the experimental resolution function (see Ref. 1) are presented in Figs. 1(a)–1(c) along with the corresponding experimental data. The comparison reveals the following features: As noted by Rollason *et al.*[1] the anisotropies are larger than the experimental values. The amplitude difference (maximum minus minimum) for the anisotropic change ($J_{\langle 110 \rangle} - J_{\langle 100 \rangle}$) is of the order 0.16 electrons a.u.$^{-1}$. However, there are significant improvements in the low-momentum and in the high-momentum region. For instance, the sign of the difference profile between $J_{\langle 110 \rangle}$ and $J_{\langle 100 \rangle}$ is in agreement with experiment. The positions of the peaks also coincide well with that of experiment. Especially, the
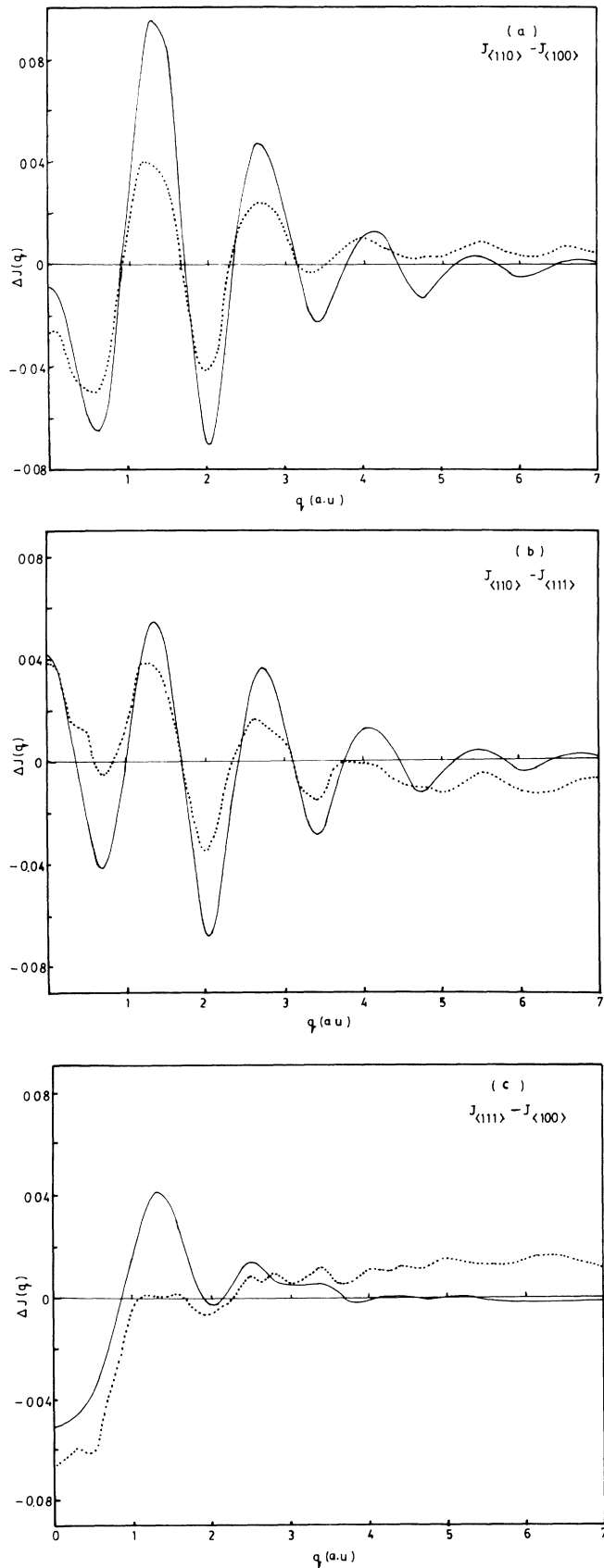
FIG. 1. The comparison of the anisotropies between theory (solid curve) and experiment (dotted curve) for (a) $J_{\langle 110 \rangle}$ $-J_{\langle 100 \rangle}$, (b) $J_{\langle 110 \rangle}-J_{\langle 111 \rangle}$, and (c) $J_{\langle 111 \rangle}-J\langle 100 \rangle$.

peak near 4 a.u. has come closer to the experimental one. It is gratifying to see that the theory reproduces the oscillatory behavior beyond $q = 2.5$ a.u. quite well.

We have also calculated $B(z)$, defined as

$$B(z) = \int \exp(-izp_z)J(p_z)dp_z . \tag{3}$$

In Fig. 2 we present $B(z)$ along the $\langle 110 \rangle$ direction with the experimental one. The improvement over earlier results [Fig. 7(b) in Ref. 1] around 3.6 a.u. is evident. We attribute the improved agreement with the experiments to the inclusion of the correlation potential.

In the present work we have tried to minimize the numerical uncertainties by using full self-consistency, the correlation potential under LDA, a large number of k points in $\frac{1}{48}$th of the BZ, and the long-range part of the momentum density up to 10 a.u. The theory correctly reproduces the oscillatory nature and the peak positions of the experimental anisotropies which indicates that the anisotropies arising out of the nature of the Fermi surface is accounted for quite adequately. Therefore, the origin of the discrepancies between the peak heights must be attributed to the nature of the LDA as pointed out by Bauer et al.[2] Since the momentum density has been calculated by using Kohn-Sham orbitals, it corresponds to the kinetic energy T_0 of noninteracting electron gas having density $n(r)$; i.e.,

$$T_0 = \int \rho(\mathbf{p})(|\mathbf{p}|^2/2m)d^3p . \tag{4}$$

It is evident from the works of Lam and Platzman[4] and Bauer et al.[2] that this discrepancy comes from the
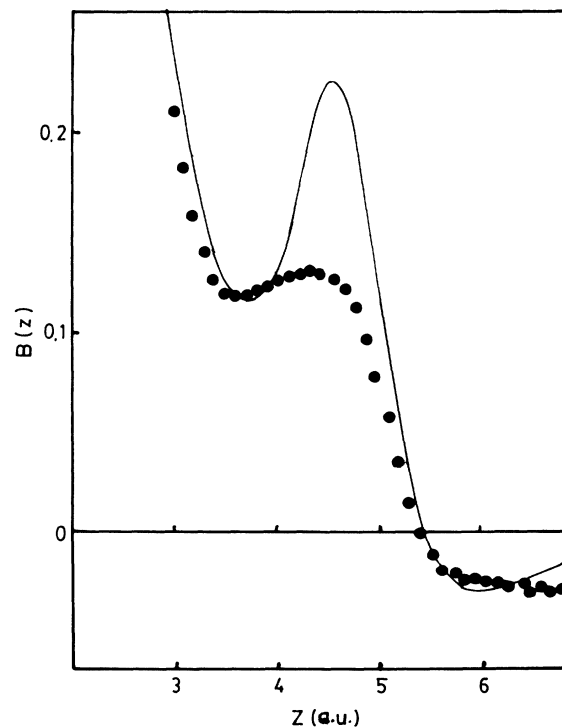

FIG. 2. The comparison of the B function between theory (solid curve) and experiment (solid circles).

difference between the occupancies of the noninteracting and interacting electron gas. The LP correction evaluated under the LDA always yields a spherically symmetric momentum density and cannot account for the anisotropic discrepancies. While this may be adequate for spherically symmetric systems such as atoms and nearly free electron solids having almost spherical Fermi surfaces, it is inadequate for transition metals having complicated anisotropic Fermi surfaces and strong umklapp contributions to the momentum density. In these metals the occupation numbers are anisotropic and this affects not only the momentum density in the first zone, but also that in the higher zones. Indeed, the work on Cu (Ref. 2) and Ni (Ref. 1) does indicate that the spherically symmetric LP correction significantly improves the theoretical profile, bringing it closer to the experimental profile only in the low-momentum region. In order to bring the theoretical anisotropies closer to the experiment, a renormalization of the momentum density near k_F and its umklapp images near $k_F + G$ is desirable. In order to see the effect of this we have carried out an *ad hoc* calculation wherein the momentum density was calculated by rewriting Eq. (2) as

$$\rho(\mathbf{p}) = \sum_{n,\mathbf{k}} f_n(\mathbf{k}) |\psi_n(\mathbf{k},\mathbf{p})|^2 , \qquad (5)$$

where $f_n(\mathbf{k})$'s were taken to be interacting electron gas occupancies calculated by using band-structure charge density and homogeneous electron gas occupancies.[8] After this the difference between the maxima and minima in the anisotropy $(J_{\langle 110 \rangle} - J_{\langle 100 \rangle})$ came down from 0.16 to 0.14 electron a.u.$^{-1}$ as compared to the experimental value of 0.10 electrons a.u.$^{-1}$. Although it is in the right direction, it is still above the experimental value.

In conclusion, we have presented an accurate calculation of the self-consistent local-density anisotropy of the Compton profiles of Ni. The comparison between the present results with the experimental work of Rollason *et al.* reveals the inadequacy of the LDA.

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