Three-Dimensional Momentum Density of Magnetic Electrons in Ferromagnetic Iron

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Experimental and theoretical three-dimensional momentum densities of magnetic electrons in ferromagnetic iron are reported for the first time and are found to be in good qualitative agreement. The experimental momentum density is reconstructed from fourteen one-dimensional magnetic Compton profiles, and the theoretical one is calculated by the full-potential linearized augmented-plane-wave method. The experiment indicates that the theory slightly underestimates the negative spin polarization of the s,p-like electrons in the first Brillouin zone and slightly overestimates umklapp processes.

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There are many experiments that aim to investigate electronic structures of magnetic materials. Among them, the measurement of magnetic Compton profiles (MCP), as well as the two-gamma angular correlation of spin-dependent positron annihilation radiation, is a useful technique to study the momentum density of itinerant magnetic electrons in ferromagnetic materials.

In the case of MCP measurements, by utilizing the spin-dependent Compton scattering of circularly polarized x rays, as was suggested by Platzman and Tzoar [1], we can directly measure a one-dimensional (1D) momentum distribution of magnetic electrons, $J_{\text{mag}}(p_z)$, defined as

$$J_{\text{mag}}(p_z)=\int\int \rho_{\text{mag}}(\mathbf{p})dp_x\,dp_y\,,$$
$$\rho_{\text{mag}}(\mathbf{p})=n^{\uparrow}(\mathbf{p})-n^{\downarrow}(\mathbf{p})\,. \quad (1)$$

Here, $n^{\uparrow}(\mathbf{p})$ and $n^{\downarrow}(\mathbf{p})$ are the momentum densities of electrons with up spins and down spins, respectively, and $\rho_{\text{mag}}(\mathbf{p})$ is the three-dimensional (3D) momentum density of magnetic electrons. The z axis is taken to be parallel to the scattering vector of the x rays. The first difficult measurement of MCP was achieved on ferromagnetic iron by one of the authors (N.S.) [2,3] using circularly polarized faint γ rays. The experiment revealed that the theory had insufficiently included the amount of negative spin polarization of itinerant s,p-like electrons. Afterwards, the emergence of synchrotron-radiation (SR) sources made it possible to provide highly intense circularly polarized x rays. The first MCP experiment using off-plane SR was reported by Cooper *et al.* [4]. The interesting crystal-anisotropy effect on MCP of iron was demonstrated by Cooper *et al.* [5], and improved results were reported in Refs. [6] and [7]. MCPs of iron, cobalt, nickel, and gadolinium were also reported by Mills [8] using an x-ray phase plate, which converts linearly polarized x rays into elliptically polarized ones. In addition to these results, many directional MCPs [7,9,10] have been measured using circularly polarized SR. However, no attempt has been made to reconstruct $\rho_{\text{mag}}(\mathbf{p})$ because of insufficient statistical accuracy and limited crystalline directions of these MCPs.

The reconstruction of the 3D momentum density $n(\mathbf{p})$ from 1D Compton profiles has been reported on Si [11–13], on GaAs [14], and very recently on LiH [15]. A corresponding reconstruction of the 3D spin-dependent angular correlation of positron-electron pairs was already achieved for iron by using polarized positrons [16]. The results, however, were strongly affected by the positron wave function, and could not fully reveal the real momentum density of magnetic electrons. This paper reports for the first time the reconstructed 3D spin momentum density of ferromagnetic iron from MCPs. The present success greatly depends on the recent development of SR technology. The preliminary results have been reported in Ref. [17].

The experiment was carried out using a beam line of the accumulation ring in National Laboratory for High Energy Physics (KEK) at Tsukuba, Japan. Circularly polarized intense 60-keV x rays emitted from an elliptical multipole wiggler [18] were used. Energy spectra of Compton scattered x rays were measured by a solid-state detector (SSD) having thirteen Ge elements to achieve high count-rate accumulation. The specimen was a thin plate (200 μm in thickness) of single crystal Fe with 3 wt.% Si. The surface is parallel to the (110) plane. Silicon is added to stabilize the body-centered-cubic structure in growing the single crystal. The details of the experimental apparatus have been reported elsewhere [19]. Two energy spectra I^+ and I^- were measured, where + or − denotes the direction of the sample magnetization. By taking the difference I^+-I^-, we obtain the spin-dependent Compton scattering component; this is because the spin-dependent Compton scattering cross section changes its sign by reversing the spin direction, while the charge scattering cross section does not change sign. The scattering angle was $160° \pm 1.5°$. Fourteen MCPs along the ⟨100⟩, ⟨110⟩, ⟨111⟩, ⟨511⟩, ⟨311⟩, ⟨211⟩, ⟨322⟩, ⟨332⟩, ⟨331⟩, ⟨510⟩, ⟨310⟩, ⟨210⟩, ⟨320⟩, and ⟨321⟩ crystal directions were measured. It took about 8 h on average to measure each MCP under the ring operation of 6.5 GeV with a mean ring current of 25 mA. A total count at the Compton peak together with the charge scattering intensity was about 10^8 per channel, where one channel corresponds to 61 eV. Since the magnetic scattering intensity is only about 1% of the charge scattering, this count gives 1% statistical accuracy of the MCP at the Compton peak. The achieved good statistical accuracy was indispensable in carrying out the reconstruction procedure.

Figure 1 shows the fourteen MCPs obtained. The statistical error is smaller than the size of the dots in the figure. Corrections for the energy-dependent Compton scattering cross section [20] and the sample absorption were made. The correction for the spin-dependent multiple scattering in the sample was estimated to be negligible [21]. The present MCPs along the ⟨100⟩, ⟨110⟩, and ⟨111⟩ directions are consistent with those of Refs. [5], [6], [7], and [10] within their statistical accuracy.

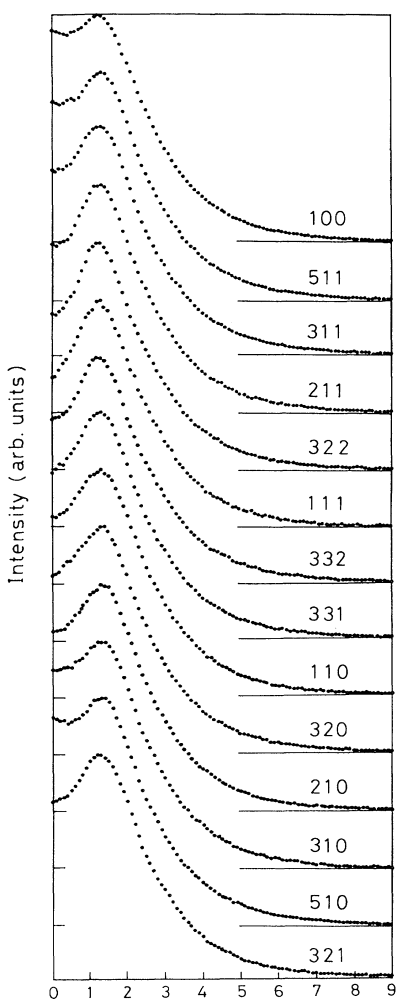

FIG. 1. Magnetic Compton profiles of ferromagnetic Fe +3 wt.% Si along fourteen directions. The evaluated momentum resolution is 0.76 a.u.

A reconstructing technique presented by Suzuki and Tanigawa [22] was used to obtain the 3D momentum density of magnetic electrons from the measured fourteen MCPs. The technique is based on the following Fourier projection theorem:

$$\mathrm{FT}[J_{\mathrm{mag}}(p_n)] \propto \int\int\int \rho_{\mathrm{mag}}(\mathbf{p}) \exp(-i\mathbf{p}\cdot\mathbf{r}) d\mathbf{p}|_n = B_n(r) . \tag{2}$$

The suffix n ($n = 1,2,\ldots,14$) denotes the direction along which the MCP is measured. The value of the 3D function $B(\mathbf{r})$ at an arbitrary position on a cubic grid is interpolated from the experimental fourteen discrete $B_n(r)$ functions. Then $\rho_{\mathrm{mag}}(\mathbf{p})$ is obtained with the inverse 3D Fourier transform of $B(\mathbf{r})$.

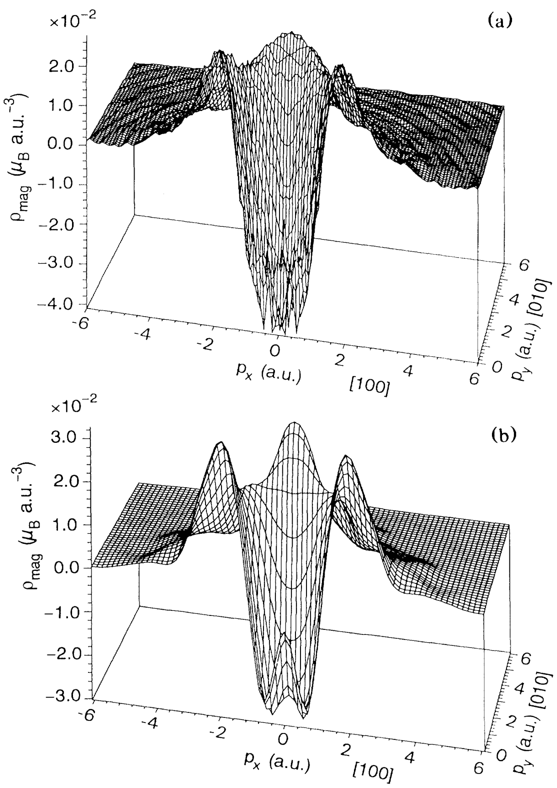

FIG. 2. (a) A cross section of the experimental ρ_{mag} in the (001) plane including the Γ point. (b) A cross section of the theoretical ρ_{mag} in the (001) plane including the Γ point. The density is convoluted with the experimental resolution expressed by a Gaussian of full width at half maximum (FWHM) 0.76 a.u.

A cross section of the experimental ρ_{mag} in the (001) plane including the Γ point is shown in Fig. 2(a). There is a deep minimum around the Γ point and four peaks at $p_x = \pm 1.8$ a.u. and $p_y = \pm 1.8$ a.u., the origins of which will be theoretically explained below. Figure 3 displays contour maps of the ρ_{mag}. The anisotropic momentum density is explicitly shown in the figures. It should be noted that the first Brillouin zone is in the region of negative spin density.

The 3D momentum density of magnetic electrons in iron has been calculated with the full-potential linearized augmented-plane-wave (FLAPW) method by one of the authors (Y.K.) [23]. A cross section of the calculated ρ_{mag} in the (001) plane including the Γ point is shown in Fig. 2(b). The agreement between Figs. 2(a) and 2(b) is very good, demonstrating the reliable calculation with the FLAPW method. However, there are some slight discrepancies between them: The magnitude of the theoretical four peaks is greater than the experimental ones, and the theoretical negative minimum is shallower than the experimental one. According to the theory, the positive peaks are mainly due to umklapp processes in the momentum space. These discrepancies thus suggest that it might be necessary to reduce the effect of umklapp processes in the band theory. A similar slight discrepancy was found in the case of MCP of *pure* nickel along the $\langle 100 \rangle$ direction [7]; a shoulder around 2.5 a.u. in the

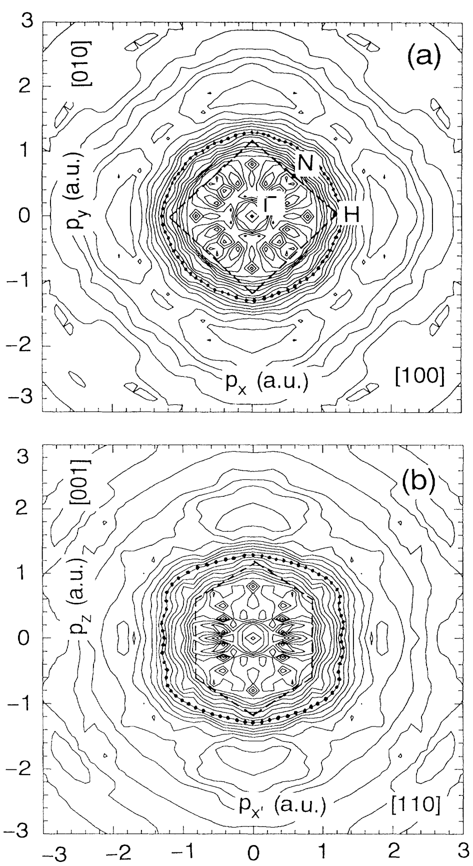

FIG. 3. Contour maps of experimental ρ_{mag}: (a) in the (001) plane including the Γ point and (b) in the $(1\bar{1}0)$ plane including the Γ point. The interval of contour curves is 3.5×10^{-3} (μ_B a.u.$^{-3}$). Dashed lines are the first Brillouin zone boundaries of iron. Dotted curves are boundaries of the negative spin density.

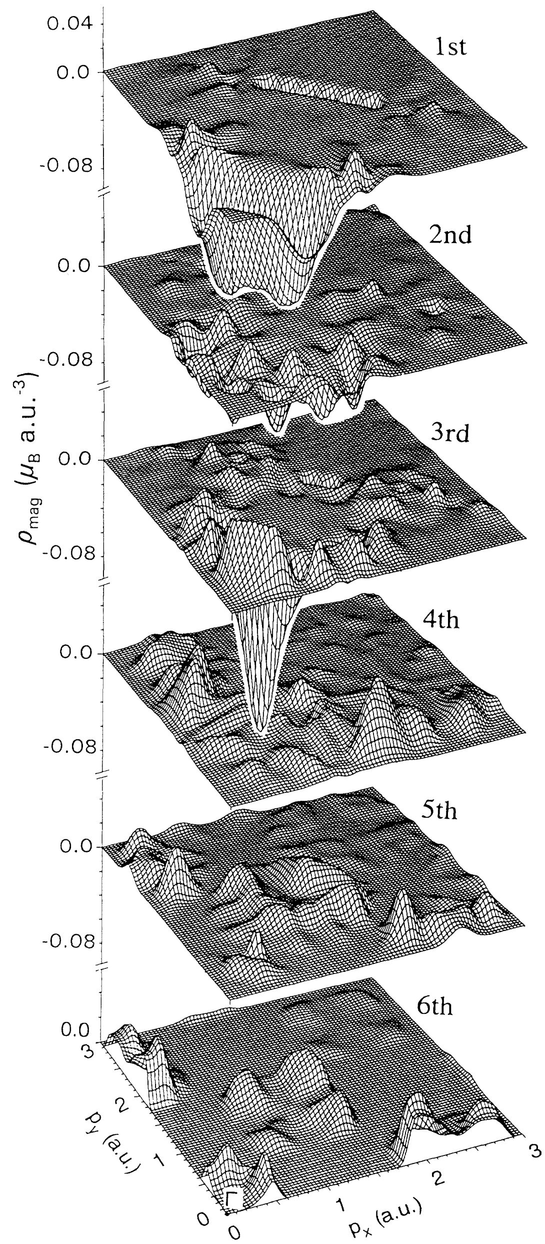

FIG. 4. Partial contributions to the theoretical ρ_{mag} from the first band to the sixth band. Each density is convoluted with a Gaussian of FWHM 0.1 a.u. Each cross section is of the (001) plane including the Γ point.

theoretical MCP, which is induced by umklapp processes, is not observed experimentally. We cannot fully reject the possibility of the magnetic effect of the silicon impurity in the iron matrix, which introduces crystalline disorder and may affect umklapp processes. However, the nonmagnetic silicon impurity does not much affect the magnetic property of iron. This is supported by the fact that the averaged atomic moment of the iron atom is not changed by alloying 3 wt.% Si [24]. The theoretical ρ_{mag} from the first band to the sixth band are shown in Fig. 4. The calculated band structure predicts negative spin polarization of the first band of s-like electrons, negative spin polarization of the second and third bands of p-like electrons, and positive spin polarization of the fourth to the sixth band of d-like electrons. There is no spin polarization at the Γ point. Experimentally, only a very small hump is observed at the Γ point. It should be noted that a sharp spike is likely to appear at the Γ point after the inverse Fourier transform procedure on account of ill treatment of the interpolation of the $B(\mathbf{r})$ function. Thus it is inappropriate at present to discuss quantitatively the discrepancy at the Γ point.

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