

Nesting driven spin spiral in ultrathin Fe/Cu(001) films

Iron is one of the most useful and familiar metals in our daily lives. However, its common physical properties are actually based on special features of its magnetism and crystal structure. Iron is delicately perched in terms of magneto-structural instability, with antiferromagnetic Cr and Mn to its left and ferromagnetic Co and Ni on its right in the periodic table. Its crystal structure is body-centered cubic (bcc) at room temperature, and it undergoes a structural transformation from bcc to face-centered cubic (fcc) at 1184 K. Since fcc is the highest density close-packed structure and expected to be more stable than the bcc form, the bcc phase at room temperature is "exceptional" from the viewpoint of structure. Thus, many studies have been performed to elucidate the correlation between structure and magnetism. Theoretical studies have predicted that, in spite of its ferromagnetism in the bcc form, fcc Fe can become nonmagnetic, ferromagnetic, antiferromagnetic or exhibit a spin spiral, sensitively depending on its lattice parameter. Experimentally, fcc Fe nanoparticle precipitates stabilized in a Cu matrix were found to exhibit a spin spiral with an ordering vector of $\mathbf{q}=(2\pi/a)(0.12, 0, 1.0)$ [1], and remarkable progress has been achieved in understanding the interplay of crystal structure and electronic instabilities in realizing spin order. Interestingly, despite extensive effort, the spin spiral associated with the bcc-fcc ($\alpha - \gamma$) transition in elemental Fe and its momentum-resolved electronic structure have remained challenging unsolved problems for over 40 years.

As an alternative to experiments at high temperatures or using nanoparticles confined in a Cu matrix, a suitable system was developed to study structure-property correlations in Fe: epitaxial ultrathin Fe films on fcc Cu(001). Ultrathin Fe/Cu(001) films have a complex magnetic and structural phase diagram. It is known that (i) below a thickness of four monolayers (MLs), an Fe/Cu(001) film is ferromagnetic and has the face-centered tetragonal (fct) structure (Region I). (ii) Between 5 and 11 ML, it has the fcc structure and a spin spiral, with a top ferromagnetic bilayer (Region II, Fig. 1). The spin spiral ordering temperature is $T_{SS}\sim 200$ K. (iii) Above 12 ML, it transforms to the bulk ferromagnetic bcc structure (Region III). In this work, we focused our interest on the spin spiral phase of Region II [2]. The spin spiral in ultrathin Fe/Cu(001) films was determined by the magneto-optic Kerr effect (MOKE) to have an ordering vector of $\mathbf{q} = (2\pi/a)(0, 0, \sim 0.75)$ [3]. According to theoretical calculations, the ordering vectors

$\mathbf{q}_1 = (2\pi/a)(0, 0, 0.6)$ and $\mathbf{q}_2 = (2\pi/a)(0.5, 0, 1.0)$ may be stable [4]; however, \mathbf{q}_1 and \mathbf{q}_2 do not match the \mathbf{q} obtained by MOKE measurements. The most serious limitation to date, however, is the absence of experimental results relating the momentum (k)-resolved electronic structure to the spin spiral in epitaxial ultrathin Fe/Cu(001) films.

To discuss this relation, it is essential to obtain the in-plane and out-of-plane Fermi surfaces of Fe/Cu(001) films. Although in-plane Fermi surfaces are routinely measured by angle-resolved photoelectron spectroscopy (ARPES) at a fixed photon energy, to extract the q_z component of a spin spiral from out-of-plane Fermi surfaces, we require bulk-sensitive ARPES with a tunable energy. Thus, we chose soft-X-ray (SX) ARPES as our technique: its larger probing depth is typically 10-20 Å.

SX-ARPES was performed with a Gammadata-Scientia SES2002 electron energy analyzer at the undulator beamline **BL17SU** using a spectrometer with a grazing incidence geometry ($<5^\circ$). Such a spectrometer makes SX-ARPES highly efficient and also ensures that the X-ray photon momentum imparted to the electrons is accounted for easily. Circularly polarized X-rays were used to avoid the symmetry selectivity of linearly polarized X-rays. The measurement was carried out at 50 K, which is considerably below $T_{SS}\sim 200$ K, to minimize indirect transition losses.

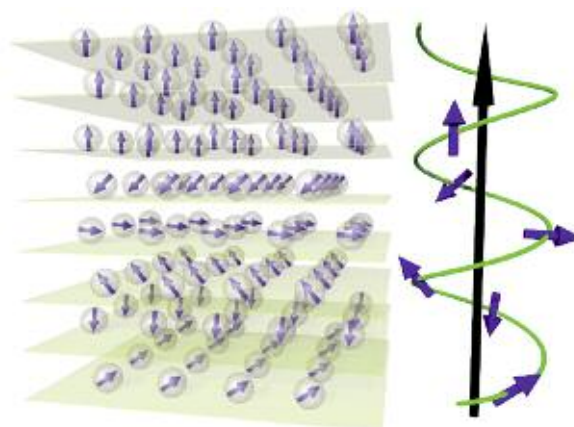


Fig. 1. Schematic representation of 8 ML Fe/Cu(001) thin films with spin spiral order (left). A Cu substrate lies just beneath the Fe thin films, but only Fe thin films are shown for simplicity. The gray balls represent Fe atoms in the thin films and the blue arrows depict spin moments of the Fe atoms. The periodic modulation of the angle of the spins in each Fe layer (blue arrows) is highlighted (right). The black arrow indicates the direction of propagation of the spin spiral.

Fe thin films were grown on Cu(001) using electron beam deposition at room temperature. The thickness of the Fe films was controlled to 8 ML by monitoring reflection high-energy electron diffraction (RHEED) oscillations. The crystallinity and crystal orientation of the Cu(001) substrate and the deposited Fe thin films were also verified by low-energy electron diffraction (LEED).

Figure 2(a) shows the Brillouin zone of fcc Fe. We measured in-plane and out-of-plane band maps along the directions indicated by blue and red lines, respectively. First, we compared experimental band maps with calculated band dispersions for fcc Fe with ordering vectors of both \mathbf{q}_1 and \mathbf{q}_2 [4] but they did not give a satisfactory match with our results. We then tried to obtain the in-plane and out-of-plane Fermi surfaces of the Fe thin films in order to determine the spin spiral vector. The in-plane and out-of-plane intensity maps at E_F were obtained from polar angle scans at $h\nu = 430$ eV and as a function of energy at $h\nu = 385\text{--}595$ eV, respectively. The raw data was measured over one-quarter of the Brillouin zone and symmetrized to obtain the experimental in-plane (Fig. 2(b)) and out-of-plane (Fig. 2(c)) Fermi surfaces for the full Brillouin zone. From peaks in MDCs, we have determined the Fermi surface crossings (Figs. 2(d) and 2(e)). Since the k_x , k_y , and k_z directions are equivalent in the fcc structure, the in-plane and out-

of-plane Fermi surfaces should be the same in the absence of electronic/magnetic anisotropies. However, the results show distinct differences between the in-plane and out-of-plane Fermi surfaces. The in-plane Fermi surfaces exhibit fourfold symmetry, whereas the out-of-plane Fermi surfaces exhibit twofold symmetry. Although the in-plane Fermi surfaces show no evidence of nesting, the out-of-plane Fermi surfaces show clear nesting. From the distance between the nestings in the out-of-plane Fermi surfaces, we identified the q_z component of the nesting vector to be $(2\pi/a) \times (0.86 \pm 0.1)$. This identification is based on precise reciprocal space maps along k_z in successive Brillouin zones. Subsequently, we identified an associated real space compressive strain of $1.5 \pm 0.5\%$ along the c -axis. To determine the in-plane component of the nesting vector, we carried out an autocorrelation analysis. Since the autocorrelation analysis using a full Brillouin zone did not give a meaningful result, we applied the autocorrelation analysis to a restricted area of the Brillouin zone (red dashed boxes in Fig. 2(e)) and obtained $q_x = 0.0$. Thus, the spin spiral vector was experimentally quantified as $\mathbf{q} = (2\pi/a)(0, 0, 0.86 \pm 0.1)$. These results are consistent with MOKE [2] and surface X-ray diffraction [5] results for Fe/Cu(001) films, and suggest the importance of in-plane and out-of-plane Fermi surface mappings for ultrathin films.

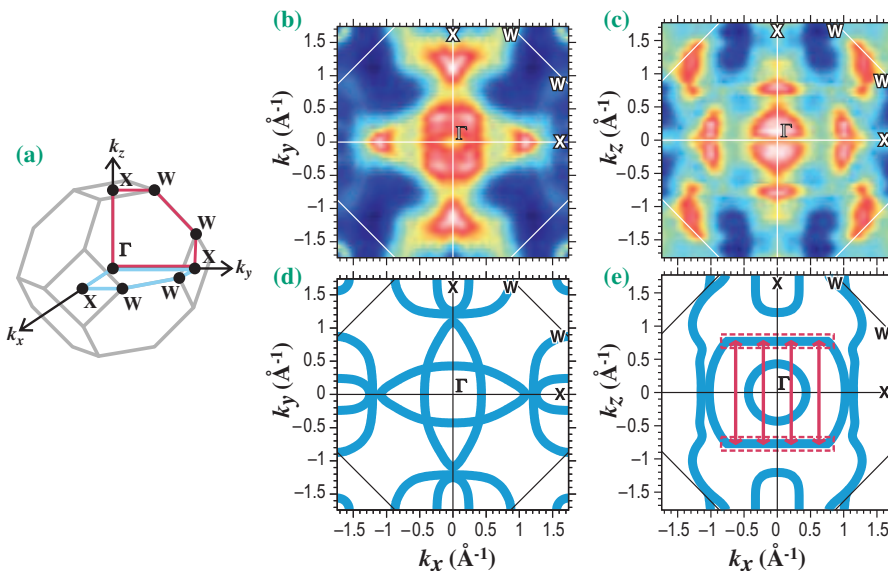


Fig. 2. In-plane and out-of-plane Fermi surfaces of Fe(8ML)/Cu(001). (a) Volume Brillouin zone of fcc Fe. (b) and (c) In-plane and out-of-plane Fermi surfaces for the full Brillouin zone, respectively, which were obtained by symmetrizing data from 1/4 of the Brillouin zone. (d) and (e) In-plane and out-of-plane Fermi surface crossings extracted from MDCs, respectively. The regions indicated by the red dashed lines are nesting parts and were used for the autocorrelation analysis.

Jun Miyawaki^{a,*}, Ashish Chainani^a and Shik Shin^{a,b}

^aSPRING-8/RIKEN

^bInstitute for Solid State Physics,
The University of Tokyo

*E-mail: miyawaki@spring8.or.jp

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