Minority-spin Dominated Band Structure Near the Fermi Energy of Fe_4N Film Revealed by Spin- And Angle-Resolved Photoemission Spectroscopy

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Fe_4N has attracted a great deal of attention as a strong candidate for spintronic materials because of the inverse tunneling magnetoresistance (TMR) effect [1], the negative anisotropic magnetoresistance [2], and an enhanced spin-pumping efficiency [3,4]. An almost 100% negative spin polarization in the conductivity has been predicted for Fe_4N, where the hybridization between Fe 4*sp* and N 2*sp* orbitals plays an important role [5], while the spin polarization in the density of states at the Fermi level (E_F) is as small as -0.6. The spin polarization for Fe_4N film was measured by the point-contact Andreev reflection experiment, while its sign remained undetermined [6]. The TMR was often used to estimate the spin-polarization using Jullier's model [7], which was significantly influenced by the film-substrate interface. It is reminded that the unexpectedly high magnetoresistance at room temperature was reported for Fe/MgO/Fe magnetic tunneling junction, where the band-selective electron conductivity takes place in 'non-half-metallic' ferromagnetic layers [8]. It tells us that an experimental determination of spin- and wavenumber-dependent band structures are quite important.

Spin- and angle-resolved photoemission spectroscopy (spin-ARPES) is one of the most powerful experimental tools to clarify the spin-dependent electronic band structures. It requires a single crystal with remanent magnetization. In fact, there have been no direct experimental observations of the electronic band structures for the bulk Fe_4N due to the lack of cleavage plane and the non-saturating magnetization in the absence of an external magnetic field. The previous spin- resolved photoemission study on the Fe_4N film reported the negative spin polarization near E_F. However, the ion-sputtering of *ex-situ* grown film caused the deficiency of N atoms and the measured spin polarization remained inaccurate [9].

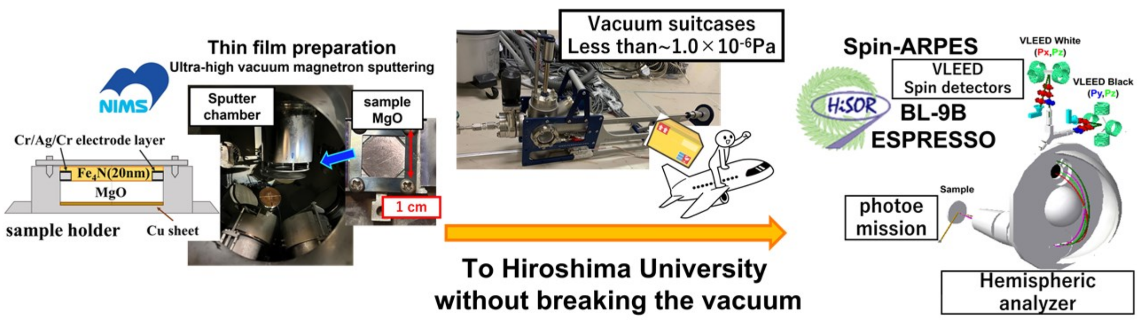

FIGURE 1. Growth of Fe_4N thin film and in-vacuum transportation to HiSOR.

To overcome this problem, we have grown the Fe_4N thin film and *in situ* performed spin-ARPES. The thin film samples were grown on MgO substrates by an ultrahigh-vacuum magnetron sputtering method at the National Institute for Materials Science (NIMS). The degree of order for nitrogen atoms in the film was evaluated to be 0.95 by the in-plane X-ray diffraction. An electrode layer of Cr/Ag/Cr was inserted to increase the electrical conductivity between the thin film and the sample holder, being indispensable for the spin-ARPES measurement.

The film samples were transported from NIMS to the preparation chamber of the spin-ARPES apparatus at HiSOR *via* the vacuum suitcase chamber. The thin films were reheated to 390~400°C to remove the contaminants adsorbed during the transportation. No nitrogen deficiency and no other impurities were detected by the Auger electron spectroscopy and the flat (001) surface was ensured by the sharp LEED spots. Spin-ARPES measurement was performed utilizing the ESPRESSO machine composed of the hemispherical analyzer and the VLEED-type spin detector with *s*- and *p*- polarized undulator radiation at BL-9B of HiSOR [7]. The photoelectron spin polarization (P) was estimated from the measured intensity asymmetry $A = (I^+ - I^-)/(I^+ + I^-)$ with the effective Sherman function ($S_{\text{eff}} = 0.3$) through the relation $P = A/S_{\text{eff}}$, where $I^+(I^-)$represents the reflected electron intensity at the positively (negatively) magnetized Fe(001)-p(1×1)O target of the spin detector.

Figure 2(a) shows the ARPES image of Fe_4N thin film in the ΓM direction of the bulk Brillouin zone (Fig.2(c)). We have observed three band structures that cross E_F at $k_{\|}$ = -1.0, -0.5 and +0.5 Å^{-1}. Figure 2(b) shows the spin-resolved energy distribution curves deduced by $I_{\uparrow\downarrow} = 0.5I(1 \pm P)$ at the fixed wavenumber of the red line in Fig.2(a), where $I_\uparrow$ ($I_\downarrow$) corresponds to the photoelectron intensity in the majority (minority) spin channel and $I = I_\uparrow + I_\downarrow$. We find that $I_\uparrow$ is dominant in the energy (E-E_F) from -1.6 eV to -0.5 eV, while $I_\downarrow$ gradually increases with increasing energy and get much higher than $I_\uparrow$ in the vicinity of E_F.

In summary, the minority-spin dominated character of the band structures in the vicinity of E_F has been unveiled experimentally for Fe_4N thin film. Our finding provides deep insights into the mechanism of the inverse tunneling magnetoresistance effect, the negative anisotropic magnetoresistance, and the enhanced spin pumping in Fe_4N, which will help the development of spintronics devices.

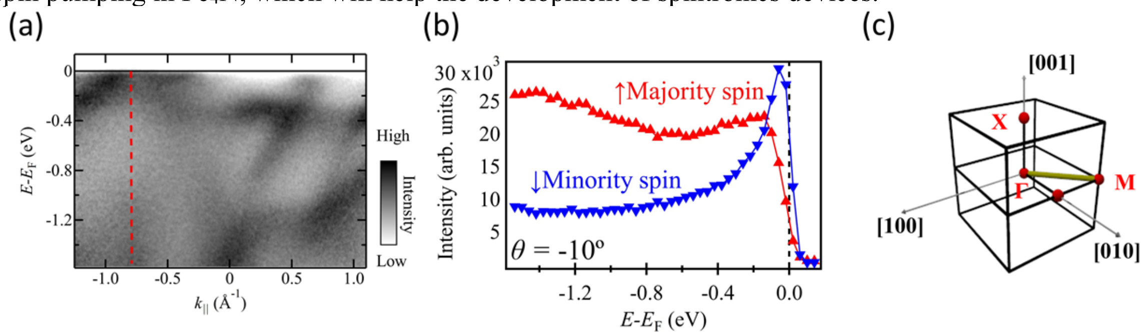

FIGURE 2. (a) ARPES energy dispersions curves along the ΓM line. (b) Spin-resolved energy distribution curves acquired at a wavenumber position denoted with red dashed line in panel (a). (c)Bulk Brillouin zone of Fe_4N.

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