17AU010, 18BU008 BL-5

Origins of Thermal Spin Depolarization in Half-Metallic Ferromagnet CrO_2

Hirokazu Fujiwara[a], Kensei Terashima[b], Masanori Sunagawa[a], Yuko Yano[a], Takanobu Nagayama[a], Tetsushi Fukura[a], Fumiya Yoshii[a], Yuka Matsuura[a], Makoto Ogata[a], Takanori Wakita[b], Koichiro Yaji[c], Ayumi Harasawa[c], Kenta Kuroda[c], Shik Shin[c], Koji Horiba[d], Hiroshi Kumigashira[d,e], Yuji Muraoka[a,b], Takayoshi Yokoya[a,b]

[a]*Graduate School of Natural Science and Technology, Okayama University, Okayama 700-8530, Japa*
[b]*Research Institute for Interdisciplinary Science, Okayama University, Okayama 700-8530, Japa*
[c]*Institute for Solid State Physics, The University of Tokyo, Kashiwa, Chiba 277-8581, Japan*
[d]*Photon Factory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba, 305-0801, Japan*
[e]*Department of Physics, Tohoku University, Sendai 980-8577, Japan*

Keywords: Half-metallic ferromagnet, CrO_2, Spin-resolved photoemission spectroscopy

In itinerant electron ferromagnets, interaction between conduction electrons and thermal spin fluctuation is of crucial importance for understanding the physical properties. As a many-body state dominating the transport properties of ferromagnetic metals, the nonquasiparticle (NQP) state, also called the spin-polaron state, was proposed in works on electron-magnon interaction [1,2]. For investigation of the behavior of the NQP state, half-metallic ferromagnets (HMFs), which have metallic electronic structures with an energy gap at the Fermi level (E_F) for one of the electronic spin state in the ground state, are ideal substances. This is because in half-metallic ferromagnets it is predicted that the effect of electron-magnon interaction is not masked by Stoner excitations unlike the usual itinerant ferromagnets, and it makes an observable modification of the electronic structure in the close vicinity of E_F [3,4]. However, only a few experimental studies suggesting the existence of NQP in HMFs have been published [5,6].

Chromium dioxide (CrO_2), which has a rutile-type crystal structure, is the simplest half-metallic oxide without carrier doping. CrO_2 shows almost 100% spin polarization, which is the highest value exhibited by a candidate HMF at low temperature, and this completely spin-polarized feature is suitable for exploring the many-body state. Theoretical studies based on the dynamical mean-field theory (DMFT) for CrO_2 showed that many-body effects broaden the bandwidth of a minority spin state above E_F and that the tail of the state cross E_F, meaning that NQP states are crucial for the occurrence of spin depolarization in CrO_2 [7,8]. Spin-resolved photoemission spectroscopy (SRPES) is a powerful technique to directly observe spin-polarized electronic structures and determine absolute values of spin polarization based on simple analyses. The pioneering works of SRPES on CrO_2, however, did not have sufficient energy resolutions to discuss depolarization near the E_F characteristic of the many-body effects. In this study [9], we have investigated the temperature dependence of the electronic structure and spin polarization of half-metallic CrO_2 (100) epitaxial films, by using laser-based high-resolution SRPES, in order to clarify the origin of the depolarization in CrO_2.

The CrO_2 (100) epitaxial films grown on a rutile-type TiO_2 (100) substrate were prepared by a closed-system chemical vapor deposition method [10]. After the synthesis, the CrO_2 film was removed from the quartz tube and then immediately placed under high vacuum for SRPES measurements. During the procedure, the CrO_2 sample was exposed to the atmosphere for approximately three minutes. High quality of the sample surface prepared with the same procedure was verified by low energy electron diffraction (LEED) and surface-sensitive PES with a synchrotron radiation of 70 eV at HiSOR BL-5 [14]. Spin-integrated and spin-resolved photoemission spectroscopy data were acquired by the laser-based spin-resolved angle-resolved photoemission spectroscopy (SARPES) apparatus in the Institute for Solid State Physics of the University of Tokyo [12]. We used a vacuum ultraviolet (VUV) laser ($h\nu$ = 6.994 eV) with p-light-polarization as an

excitation beam. During the measurement, the instrumental energy resolution was set to 20 meV and the base pressure was kept below 1 x 10^{-8} Pa. Calibration of E_F for the sample was achieved using a gold reference. We magnetized the CrO_2 (100) sample along the magnetic easy axis ([001] direction) by bringing the sample close to a magnet at room temperature. The approximate magnitude of the magnetic field at the sample position was 600 Oe.

A minority spin spectrum measured at 20 K (not shown) exhibits an energy gap of 10 meV below E_F, which is much smaller than the previous estimation of about 500 meV [11,13]. This inconsistency can be attributed to a very small but non-negligible intensity from the surface contaminants (most probably Cr_2O_3), which will result in an underestimation of the spin polarization since the signals from Cr_2O_3 comprise unpolarized background. In order to eliminate this unpolarized background, we subtracted smoothed spectra of the minority spin spectra from both of the majority and the minority spin spectra. Figure 1 (a) shows the spin polarizations for various temperatures. The spin polarizations are normalized to their values at E_B = 80 meV. It is clear that energy-dependent depolarization occurs at 80 K near E_F and it grows up at higher temperatures. Figure (c) and (d) are the spin-resolved energy distribution curves (EDCs) divided by the Fermi-Dirac (FD) function, showing that a minority spin state exists at E_F above 80 K (purple shaded region). This state is enhanced in the minority spin gap as temperature increases. This temperature dependence of the minority spin state is consistent with that of the NQP theories, which constitutes spectroscopic evidence for the appearance of a minority tail state attributed to a many-body effect. A thorough investigation of the fine spin-resolved electronic structure of HMFs will be important for understanding the many-body effect in itinerant electron ferromagnets and for realizing complete spin polarization at room temperature, which may accelerate the development of spintronic devices.

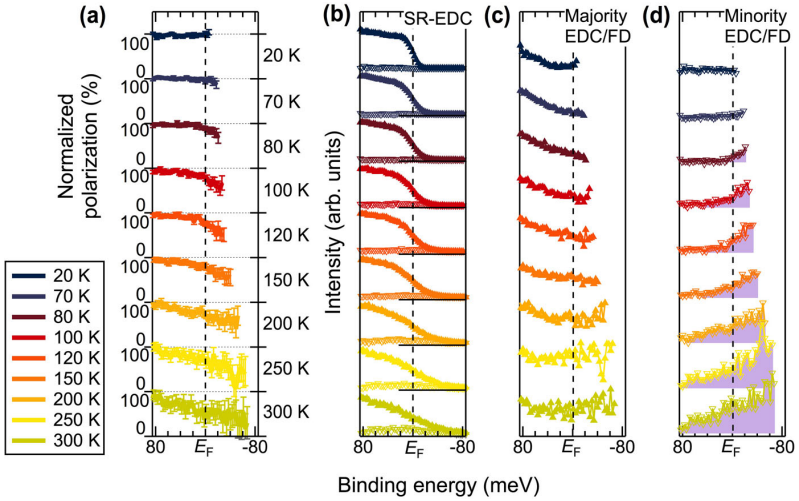

FIGURE 1. Minority tail states at finite temperature. (a) Spin polarizations for various temperatures obtained from majority and minority spin spectra with the background subtraction procedure. They are normalized to their values at EB = 80 meV. The error is indicated by bars. (b) Spin-resolved EDCs calculated from $I_\uparrow = (1 + P_{nor})I_{tot}/2$ (triangle-up) and $I\downarrow = (1 - P_{nor})I_{tot}/2$ (triangle-down), where P_{nor} is the normalized spin polarization shown in panel (a). (c,d) Majority and minority EDCs divided by the FD function at the measured temperature convoluted with the experimental resolution.

REFERENCES

1. J. A. Hertz and D. M. Edwards, *J. Phys. F* **3**, 2174 (1973).
2. J. A. Hertz and D. M. Edwards, *J. Phys. F* **3**, 2191 (1973).
3. V. Yu. Irkhin and M. I. Katsnelson, *J. Phys. C* **18,** 4173 (1985).
4. V. Yu. Irkhin, M. I. Katsnelson, and A. I. Lichtenstein, *J. Phys. :Condens. Matter* **19**, 315201 (2007).
5. L. Chioncel *et al*., *Phys. Rev. Lett.* **100**, 086402 (2008).
6. K. Miyamoto *et al.*, *Phys. Rev. B* **79**, 100405(R) (2009).
7. M. I. Katsnelson *et al*., *Rev. Mod. Phys*. **80**, 315 (2008).
8. L. Chioncel *et al*., *Phys. Rev. B* **75**, 140406(R) (2007).
9. H. Fujiwara *et al*., *Phys. Rev. Lett.* **121**, 257201 (2018).
10. K. Iwai *et al., J. Appl. Phys.* **108**, 043916 (2010).
11. H. Fujiwara *et al., J. Electron Spectrosc. Relat. Phenom.* **220**, 46 (2017).
12. K. Yaji *et al., Rev. Sci. Instrum.* **87**, 053111 (2016).
13. H. Fujiwara *et al., Appl. Phys. Lett.* **106**, 202404 (2015).