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Spin-polarized band structures of Ga-rich Fe_3Ga film as a promising material for high thermoelectric performance

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Thermoelectric materials are very promising for solving environmental problems because they can directly convert waste heat energy into electrical energy. They have so far been based on the so-called Seebeck effect, which generates an electric field in a direction to the heat flow. Since it is a one-dimensional phenomenon, it becomes an obstacle in minimizing the size and manufacturing cost. On the other hand, the anomalous Nernst effect (ANE) generates an electromotive force perpendicular to the magnetization and the heat flow in ferromagnetic materials. This three-dimensional nature makes it possible to apply it to complex curved surfaces. Therefore, the ANE has a great potential for application in energy harvesting technology.

Recently, a giant ANE up to 2.4 μV/K, which is two order of magnitude larger than that of pure Fe, has been experimentally found in the disordered binary ferromagnetic alloy $Fe_{1-x}Ga_x$ consisting only of earth abundant and non-toxic elements [1]. Later, it has reached 4 μV/K in $D0_3$ phase Fe_3Ga thin film [2]. A recent theoretical study suggests that such a remarkable ANE originates from an intrinsic mechanism caused by a peculiar band structure [2]. To elucidate the mechanism of ANE and develop the thermoelectric device with higher performance, a direct observation of the band structures with spin resolution is essential. However, no experimental evidence on the spin resolved band structures have been missing so far.

Motivated by this, we have carried out a spin- and angle-resolved photoelectron spectroscopy (spin-ARPES) measurement on $D0_3$ phase $Fe_{72}Ga_{28}$ thin film. The thin film sample was deposited on MgO(001) substrate *via* magnetron sputtering method at NIMS (Tsukuba) and transferred to the spin-ARPES apparatus (Hiroshima Univ.) using the vacuum suitcase chamber. The experiment was performed at HiSOR BL-9B using the VLEED spin detector attached to the hemispherical analyzer [3].

Figure 1(a) shows the ARPES image acquired along [110] direction with a photon energy of 60 eV. We find steeply dispersive bands that cross the Fermi level (E_F) around $k_{||} = \pm 0.5$ Å^{-1}. No recognizable features are observed for $|k_{||}| < 0.5$ Å^{-1} above E-E_F = 1.0 eV, whereas less dispersive features with the rather strong intensity are observed at E-E_F = -1.0 ~ -1.5 eV. The spin-resolved energy distribution curves (EDC) at $k_{||}$ = 0 Å^{-1} (cut 1), 0.60 Å^{-1}(cut 2), 0.79 Å^{-1}(cut 3) and 1.12 Å^{-1}(cut 4) [see panel (a)] are shown in Fig. 1(b). Minority-spin EDCs for cut 1 and 2 show a peak near E-E_F = 0 eV, which confirms a minority spin character for the steeply dispersive bands crossing E_F near $k_{||} = \pm 0.5$ Å^{-1}.The minority-spin EDC at $k_{||} = 0$ Å^{-1} (cut 1) shows a single peak at E-E_F = -1.1 eV, while the majority-spin counterpart exhibits a broad feature at the similar energy that can be roughly decomposed into two intensity maxima at -1.0 and -1.2 eV. It tells us that the less dispersive feature shown in Fig. 1(a) near $k_{||} = 0$ Å^{-1} involves three distinctive bands possessing majority, minority and majority spin characteristics at E-E_F = -1.0, -1.1 and -1.2 eV, respectively. All of the experimental results shown here are reasonably explained by the theoretical band structures using the first principles calculation (*not shown*) when the theoretical E_F is shifted towards the lower energy (higher binding energy) by about 100 meV. Since the predicted nodal webs, which should be responsible for the higher Nernst coefficient, are still located above E_F, the further electron doping would be necessary to maximize the transverse thermoelectric performance.

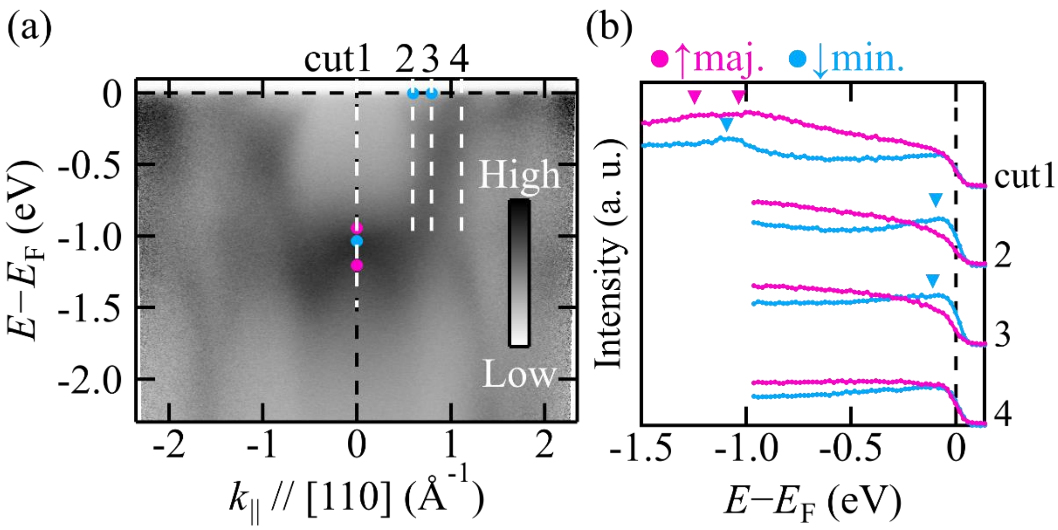

Figure 1 (a) ARPES image of $Fe_{72}Ga_{28}$ thin film along [110] direction acquired at a photon energy of 60 eV. (b) Spin-resolved EDC in majority (red) and minority (blue) spin channels at $k = 0$ Å^{-1} (cut 1), 0.60 Å^{-1} (cut 2), 0.79 Å^{-1} (cut 3) and 1.12 Å^{-1} (cut 4).

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