Spin-Resolved Photoemission of Heusler-Type Weyl Ferromagnet Films

Kazuki Sumida^a

^aMaterials Sciences Research Center, Japan Atomic Energy Agency, Hyogo 679-5148, Japan

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When electric and thermal currents flow through a ferromagnet, an electric field emerges orthogonally to the current path [Fig. 1(a)]. The two effects are, respectively, called the anomalous Hall (AHE) and Nernst (ANE) effects and are exploited as operating mechanisms in various novel applications. The associated transverse voltage of the electric field is empirically proportional to its spontaneous magnetization. In contrast to the general belief, recent discoveries of both large AHE and ANE, which do not scale with magnetization, have elicited great surprise. In particular, the observed ANE thermopower and AHE conductivity of single crystalline bulk Co_2MnGa and Co_2MnAl are an order of magnitude larger than those of other ferromagnets with similar magnetizations [1,2,3]. These transverse properties are postulated to arise from a Berry curvature emerging within band structures near the Fermi energy (E_F).

Topologically non-trivial Weyl semimetals possessing massless fermions characterized by zero-gap and linear band dispersions are promising candidates featuring a large Berry curvature. Weyl fermions in solids can be realized in materials that break inversion symmetry or time-reversal symmetry. With the breaking of such symmetries, Weyl nodes appear as pairs in momentum space and act as magnetic monopoles with positive and negative chiralities. To date, Weyl fermions have been verified in experiments in non-centrosymmetric (e.g., TaAs-family) and magnetic materials (e.g., Mn₃Sn) through angle-resolved photoelectron spectroscopy (ARPES) and magneto-transport measurements [4,5].



FIGURE 1. Schematic illustrations of (a) the ANE and (b) the ANE thermopower and magnetization of Heusler alloy in bulk and film forms. Black and gray arrows indicate the remanence magnetization. (c,d) Calculated band structures of Co_2MnGa Heusler alloy without and with spin-orbit coupling (SOC). Red and blue colors correspond to the majority and minority spin. (e,f) Three-dimensional view of the Weyl nodal lines of Co_2MnGa without and with SOC. Color corresponds to the location of the Weyl node. The blue arrow in (f) indicates the direction of magnetization.

Recently, Co₂MnGa and Co₂MnAl Heusler alloys have also been theoretically predicted to be a ferromagnetic Weyl semimetal [6] and has been experimentally demonstrated in the bulk form to exhibit large anomalous transport properties under an external magnetic field [1,2,3]. The nature of this highly symmetric crystal creates mirror-symmetry-protected Weyl nodal lines in the band structure as encountered by theory [Figs. 1(c) and 1(e)] and experiments [7]. However, the nodal lines lead to vanishing Berry curvature when integrated over the whole Brillouin zone and cannot explain the observed phenomena. One way to obtain a large Berry curvature is to gap out their nodal lines using remanent magnetization or an external magnetic field (specifically, to break the mirror symmetry) [Figs. 1(d) and 1(f)]. Yet, the experimental evidence for broken mirror symmetry was not provided by the recent ARPES measurement on bulk Co₂MnGa crystal because the remanent magnetization was negligible [Fig. 1(b)] as applying external magnetic fields is not permitted in this measurement. For practical applications in which zero-field operation and gigantic outputs are a requirement, it is thus indispensable to truly understand the band structure responsible for the anomalous transport properties in films with full remanent magnetization.

In this talk, I show the spin-polarized band structure and anomalous transport properties of ferromagnetic Co₂MnGa thin films [8]. Growth of high-quality thin films possessing full remanent magnetization and in situ spin-resolved ARPES (SARPES) measurements permit access to their non-trivial band structures modified by the broken mirror symmetry. We observed spin-polarized Weyl cones located mostly at a Lifshitz quantum critical point and a flat band of surface states [Fig. 2]. Furthermore, when the energy associated with the "massive" Weyl cone approaches E_F , the AHE and ANE conductivities systematically increase as the electron number rises. In particular, the ANE reaches thermopower of ~6.2 μ V/K at room temperature, which is the highest amongst magnetic films to the best of our knowledge. In addition, I also present the spin-polarized band structure of the quaternary Co₂Mn(Al,Si) Heusler film. We succeeded in doping electrons about 350 meV in the Weyl ferromagnet Co₂MnAl by Si substitution and revealed that the spin-polarized multiple Weyl cones and half-metallic gap coexist in the bulk electronic structure [9].



FIGURE 2. (a) Spin-integrated ARPES image of Co-rich Co_2MnGa film along Γ -K-X line recorded at 80 eV with *p*-polarized light. (b,c) SARPES images of the majority and minority spin states. (d) Spin-polarization map. ARPES and SARPES images are symmetrized with respect to the Γ point.

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