Temperature Dependent Angle-Resolved Photoemission Spectroscopy on Ferromagnetic EuO Thin Films

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Europium monoxide (EuO) is a ferromagnetic semiconductor with the Curié temperature ($T_{\rm C}$) at around 70 K [1, 2]. In the electron doping case by the Eu excess or substitute Gd³⁺ or La³⁺ from Eu²⁺ ion, the $T_{\rm C}$ increases up to 150 K and the electrical resistivity drops twelve-order of magnitude below the $T_{\rm C}$ originating in a metal-insulator transition (MIT) [2, 3]. To reveal the origin of these physical properties of EuO, it is important to clarify the electronic structure. Three dimensional angle-resolved photoemission spectroscopy (3D-ARPES) using a synchrotron radiation source is the most powerful technique to directly determine the electronic band structure. Using this technique we observed the change in the Eu 4f and O 2p states across $T_{\rm C}$.

Single-crystalline EuO thin films with a thickness of about 50 nm were fabricated by the molecular beam epitaxy (MBE) [4]. Epitaxial growth of the single-crystalline EuO thin films with the 1 x 1 EuO (100) patterns was confirmed with low energy electron diffraction (LEED) and reflection high energy electron diffraction (RHEED) methods. The $T_{\rm C}$ measured with a superconducting quantum interference device (SQUID) magnetometer was 71 K. The 3D-ARPES measurements were performed at the beam line 5U of UVSOR-II combined with the MBE system.

Figures 1(g1) [1(g2)] and 1(x1) [1(x2)] show the temperature dependence of the Eu 4f [O 2p] band at the Γ and X points, respectively. The circles and bright area correspond to the peak positions of the second-derivative energy distribution curves (EDCs). With decreasing temperature across $T_{\rm C}$, all of the bands shift by 0.2–0.3 eV to the lower binding energy side. The overall energy shift is in good agreement with the magnetic red shift estimated from the optical absorption spectra [5]. This indicates that the overall energy shift originates from the changing of the bottom energy of the Eu 5d conduction band at the X point due to the energy gain of the Eu 5dmajority-spin state after band splitting caused by ferromagnetic ordering. In addition, the top of the Eu 4f states is shifted away from the main 4f states at the Γ and X points. In contrast, the O 2p state is spitted into two bands below $T_{\rm C}$. The bands at the higher and lower binding energies are attributed to the majority and minority spin states, respectively. Since the Eu 4fstate is fully polarized, the 4f state mainly hybridizes

with the majority spin state of O 2p and Eu 5d. Therefore, the band shits can be attributed to the hybridization effect between the Eu 4f and O 2p states (superexchange interaction) and between the Eu 4fand 5d sates (indirect exchange interaction). The observed temperature dependent energy shift of the 4fstate is the essential origin of the ferromagnetic phase transition of EuO.



Fig. 1 Temperature dependence of the photoemission peak energies derived from the second-derivative EDCs of the Eu 4*f* [(g1), (x1)] and O 2*p* [(g2), (x2)] states at the Γ and X points. The open circles indicate the peaks of the second derivative EDCs.

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