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Temperature dependent phase transition of EuO on MgO(100)

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We investigated the changes of magnetic properties and phase transition in EuO films grown on a MgO(100) substrate as a function of temperature. As the temperature was varied, we observed a clear phase transition and polarization change at 69 K using spin resolved photoemission spectroscopy and low energy electron diffraction. We will elucidate the temperature driven changes in the electronic and magnetic properties of EuO films on a MgO(100) substrate. © 2007 American Institute of Physics. [DOI: 10.1063/1.2775297]

I. INTRODUCTION

Over the past several decades, the semiconducting material Eu monoxide (EuO) has been intensively studied not only because of its scientific interest but also its many technological applications.¹⁻⁸ Recently, ferromagnetic semiconductors^{9,10} such as EuO and CrO₂ have attracted considerable interest due to their applications, for example, as a source of polarized carriers for injection.¹¹⁻¹³ The localized magnetic moments of the half-filled 4*f* shell of the Eu atoms and the existence of a conduction band make EuO an appropriate system to test the Kondo-lattice model.¹⁴ Interestingly, it also shows a colossal magnetoresistance (CMR) behavior in that an applied field shifts the metal-insulator transition (MIT) temperature, as has been reported for La_{1-x}Sr_xMnO₃.¹⁵⁻¹⁷ However, EuO films are difficult to fabricate because they form only under particular growth conditions (e.g., oxygen partial pressure and substrate temperature), and they are easily changed into Eu₂O₃ films if the growth conditions are not rigorously maintained.^{18,19}

In the present study, we investigate the change in polarization [$P = (I_{\text{spin up}} - I_{\text{spin down}}) / (I_{\text{spin up}} + I_{\text{spin down}})$] and magnetic and electronic characteristics of a EuO film on a MgO(100) surface as a function of temperature using spin resolved photoemission spectroscopy (SRPES), x-ray absorption spectroscopy (XAS), and low energy electron diffraction (LEED).

II. EXPERIMENT

The experiments were performed at the 2A1 (EPU6) undulator beamline at Pohang Accelerator Laboratory. Spin resolved photoemission spectra were measured using a 125 mm hemispherical analyzer equipped with a mini Mott detector. The total energy resolution was set to 300 meV. The soft x-ray absorption spectra of the europium *M*₅ edge (1118–1148 eV) were also measured with a spectral resolution of 0.1 eV. All measurements were performed under

UHV conditions ($P = 7 \times 10^{-11}$ Torr) at room temperature. The MgO(100) surface was cleaned by several cycles of sputtering with 1.5 keV Ar⁺ ions at 300 K for 30 min, followed by annealing at 700 K for 15 min. The cleanness of the surface was checked by photoemission spectroscopy (charging effect) and LEED (1×1 LEED pattern). All magnetic measurements were performed in remanence. Application of a magnetization pulse (full width at half maximum of ~ 0.5 ms and I_{max} of ~ 1600 Oe) in an in-plane direction along the *z* axis was found to be sufficient to saturate the sample.

III. DATA AND RESULTS

Figure 1 shows the Eu 4*d* core level spectra (left panel) and valence band spectra (right panel) for Eu metal, Eu monoxide (EuO), and Eu trivalent oxide (Eu₂O₃). The different stoichiometries were generated by controlling the oxygen partial pressure. Figure 1(a) shows a typical metallic Eu 4*d* core level spectrum with a 127 eV binding energy. As shown in this spectrum, the sharpness of the *D*⁹ states is clearly resolved in the lowest peak structure. Moreover, the exchange-split high binding energy states are broadened by the decay effect, and the low energy spectral features are similar for the spectra convoluted with constant Γ and with term dependent Γ_f plus Gaussian, in accordance with previous results.¹¹

To fabricate a single-phase EuO film, we deposited Eu atoms while maintaining the temperature of MgO(100) surface at 400 °C under an oxygen partial pressure of 2×10^{-8} Torr. Figure 1(b) shows the Eu 4*d* core level spectrum of EuO with a 128 eV binding energy. Compared with the metallic Eu 4*d* core level, the spectral shape is similar (e.g., a decay effect is observed), but the resolved *D*⁹ state has disappeared and the core level spectrum is shifted toward a higher binding energy by 1 eV. Hence, we can clearly distinguish the EuO and Eu metal films based on the spectral data. Increasing the partial pressure of molecular oxygen up to 5×10^{-8} Torr led to the facile formation of a Eu₂O₃ film. The Eu 4*d* core level spectrum of Eu₂O₃, shown in Fig. 1(c),

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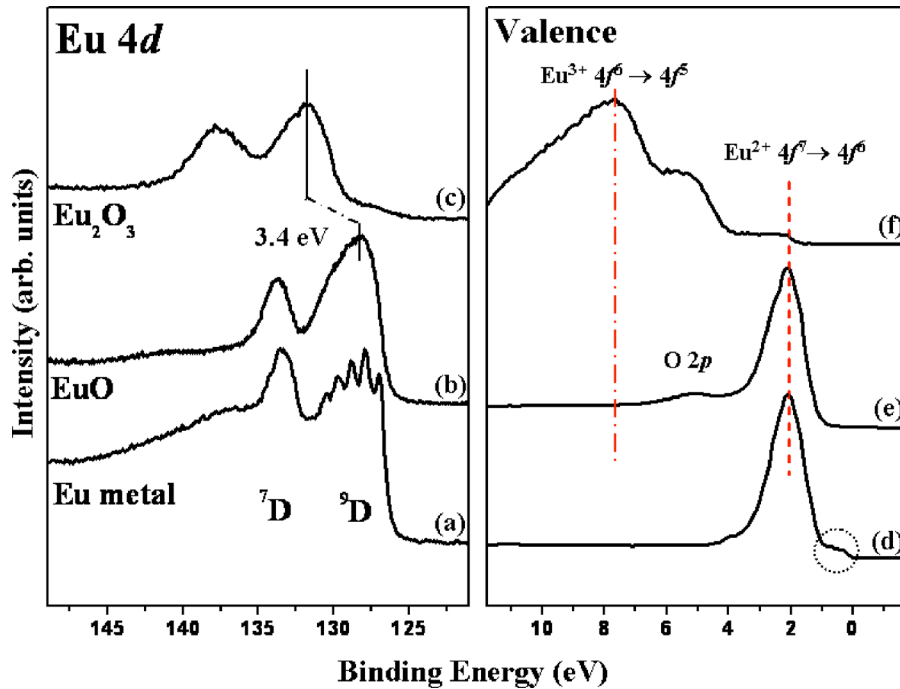


FIG. 1. (Color online) Eu 4d core level spectra (left panel) and valence band spectra (right panel) of Eu oxide films with different stoichiometries. Photoemission spectra of [(a) and (d)] Eu metal, [(b) and (e)] Eu monoxide (EuO), and [(c) and (f)] Eu trivalent oxide (Eu_2O_3).

is clearly different from the spectra for the metallic Eu or EuO film. In particular, it shows a larger binding energy shift (3.4 eV) toward a higher binding energy, indicative of an insulator, and the decay effect of the D^7 state is not observed.

To clarify the variation of the valence band with variations in the Eu oxide stoichiometry, we recorded the valence band spectra at photon energy of 135 eV. Figure 1(d) shows the valence spectrum of clean Eu metal, which exhibits the f^7 transition peak and density of states at the Fermi level (marked with a circle). Figure 1(e) shows a typical valence spectrum of EuO, showing only spectral weight from $\text{Eu}^{2+} 4f$ and from the O 2p levels. Moreover, we observed a zero density of states at the Fermi level in this spectrum. It is noted that EuO is not metallic but semiconducting. On the other hand, the valence spectrum of Eu_2O_3 is remarkably different. As shown in Fig. 1(f), no transition peak from $4f^7$ to $4f^6$ is evident; rather, a transition peak from $4f^6$ to $4f^5$ located at 7.8 eV is observed, which is typical of Eu^{3+} . From these spectra obtained from films prepared under different conditions, we confirmed that a EuO film can be prepared by controlling the oxygen partial pressure.

Figure 2 displays XAS spectra of films with various compositions: EuO, a mixture of EuO and Eu_2O_3 , and Eu_2O_3 . Similar to the photoemission spectra, because EuO has a $\text{Eu}^{2+}4f^7+(spd)^0$ configuration and Eu_2O_3 has a $\text{Eu}^{3+}4f^6+(spd)^1$ configuration, we can easily confirm the formation of a EuO film using XAS. Figure 2(a) shows the XAS spectrum of a EuO film, which we confirmed from the photoemission spectra shown in Figs. 1(b) and 1(e). This spectrum contains two clear peaks: the Eu^{2+} XAS peak (1127.9 eV), which is only observed for EuO, and its satellite peak (1132.0 eV). For comparison, we also measured the adsorption spectrum of single-phase Eu_2O_3 [Fig. 2(c)]. Eu^{3+} induced XAS peaks at 1130.2 and 1133.8 eV, respectively, which are clearly different from the features in the EuO spectrum. Moreover, to clarify the discrepancy between EuO and

Eu_2O_3 , we measured the XAS of a film comprised of a mixture of EuO and Eu_2O_3 . As shown in Fig. 2(b), this spectrum contains a superposition of features characteristic of EuO and Eu_2O_3 and is clearly different from the spectrum of the single-phase EuO film. Thus, we can confirm the formation of a EuO film on the basis of photoemission and XAS spectral data.

After confirming the formation of a EuO film, we performed SRPES to elucidate the polarization change and phase transition according to temperature. Figure 3 displays the SRPE spectra for EuO on MgO(100) surface as a function of temperature. As mentioned in the Introduction, EuO shows MIT behavior and a magnetic property change from ferromagnetic (FM) to paramagnetic (PM) at 69 K.¹³ The spin resolved valence band spectrum obtained at 40 K [Fig.

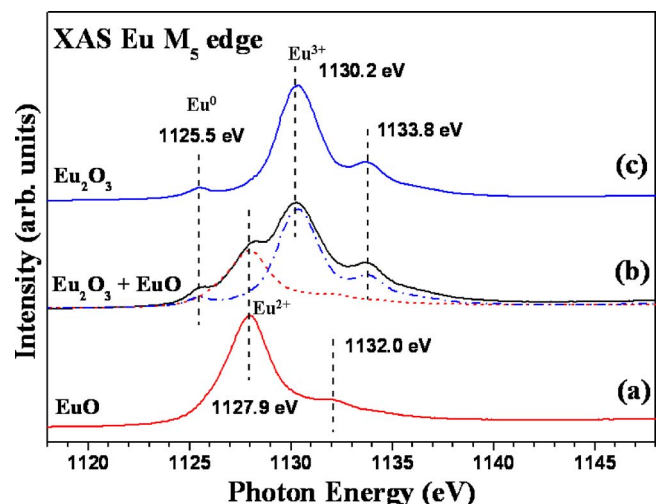


FIG. 2. (Color online) Eu based x-ray adsorption spectra (XAS) with various compositions. (a), (b), and (c) show XAS of EuO, mixed EuO and Eu_2O_3 , and Eu_2O_3 , respectively. The dotted line of Fig. 2(b) shows the result of curve fitting for XAS of mixed Eu oxide.

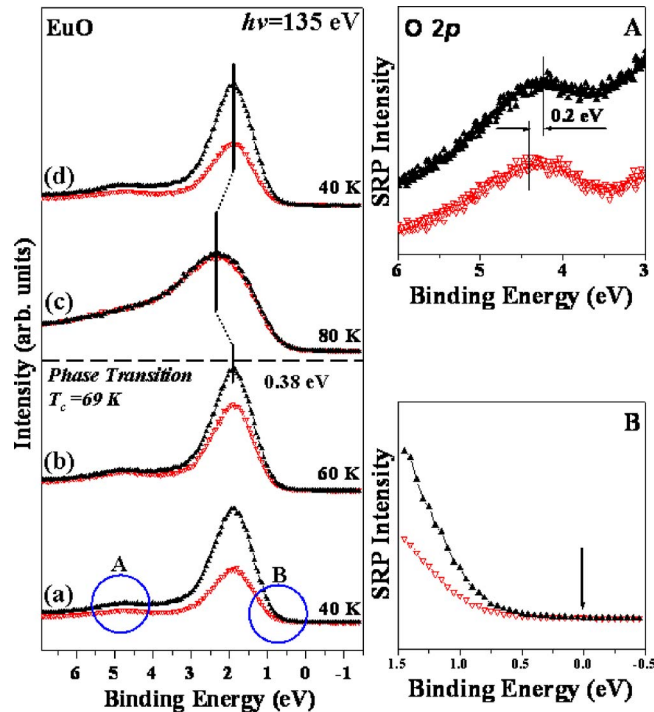


FIG. 3. (Color online) Left panel shows the polarization change for EuO on MgO(100) using spin resolved photoemission (SRPE) spectra as a function of temperature. (a), (b), (c), and (d) show SRPE spectra obtained at 40, 60, 80, and 40 K, respectively. **A** displays the exchange splitting of the O $2p$ region (0.2 eV), and **B** shows the Fermi level region to clarify the half metallic behavior.

3(a)] is a well polarized spectrum, with a 50% polarization value. However, Eu $4f$ shows no exchange splitting in this spectrum. On the other hand, the O $2p$ region induced by EuO shows an exchange splitting of 0.2 eV (see circled area marked as **A** in Fig. 3, upper right panel), which can be explained by hybridization between O $2p$ and Eu $5d/6s$.²⁰

To examine the possibility of half metallic behavior, we scrutinized the Fermi level region (circled area marked as **B** in Fig. 3, bottom right panel). As shown in this zoom in spectrum, the density of states at the Fermi level shows no Fermi level crossing for all spin states, indicating no metallic behavior. To show the half metallic behavior, one of the spin states should be crossing at the Fermi level. However, we did

not find any evidence for half metallic behavior in our EuO film. Hence, we exclude the possibility of half metallic behavior.

On increasing the temperature from 40 to 60 K [Fig. 3(b)], a decrease in the polarization is observed. To see the phase transition as a function of temperature, we further increased the temperature to 80 K [Fig. 3(c)]. At 80 K, we observed a zero polarization (0%) and a binding energy shift toward a higher binding energy of 0.38 eV. These features indicate that a magnetic phase transition from FM to PM occurs between 60 and 80 K, consistent with previous reports that EuO undergoes such a transition at 69 K. Moreover, the 0.38 eV shift toward a higher binding energy clearly shows that the phase transition involves a change in electronic structure. To confirm the reversibility of the transition, we decreased the temperature again to 40 K. As shown in Fig. 3(d), the polarization (50%) reappeared and the binding energy returned after cooling to 40 K.

Figure 4 shows the LEED patterns recorded at 60 and 80 K (either side of the phase transition) to see the correlation between morphological changes and the phase transition as a function of temperature. Interestingly, we observed that the spot size of the LEED pattern decreased when the temperature was decreased from 80 K [Fig. 4(a)] to 60 K [Fig. 4(b)]. The variation in spot size as a function of temperature [Fig. 4(c)] showed a sharp drop off between 80 and 60 K, consistent with a phase transition at 69 K. Thus, the present results show a close correspondence between the phase transition and a change of spot size, indicating that the surface morphology of the EuO film changes as a function of temperature.

Figure 5 displays plots for changes in polarization and binding energy shift as a function of temperature for the EuO film. In this plot, the polarization value is 0 (paramagnetic) and the binding energy shifts toward the higher binding energy region by 0.38 eV at temperatures above 69 K. Below 69 K, in contrast, the polarization increases with decreasing temperature and the binding energy remains at the same value.

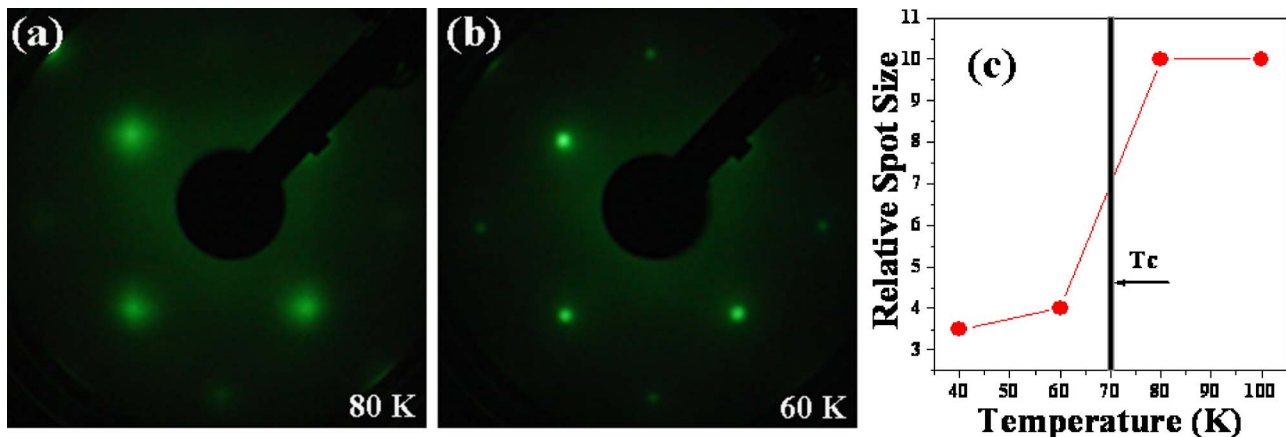


FIG. 4. (Color online) LEED images recorded at two different temperatures: (a) 80 and (b) 60 K. (c) Plot of relative spot size vs temperature.

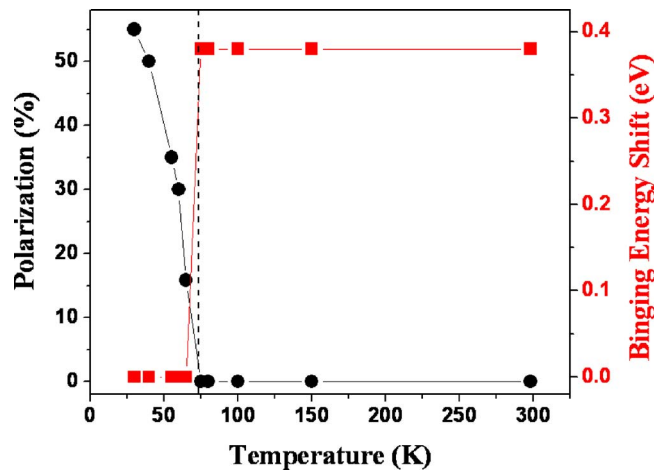


FIG. 5. (Color online) Plots of the polarization (closed circles) and binding energy shift (closed squares) as a function of temperature for a EuO film on a MgO(100) surface.

IV. SUMMARY

In the present study of EuO films on MgO(100) substrate, we obtained three remarkable experimental results. First, in an investigation of the temperature dependent polarization change and phase transition using SRPES, we clearly observed the magnetic transition (from FM to PM) and MIT at 69 K. Second, even though we observed an incomplete spin polarization of the Eu 4*f* peak, we could exclude the possibility that the EuO film exhibits the half metallic behavior on the basis of the change of the Fermi level for the two spin states (majority and minority). Third, in LEED patterns recorded at temperatures spanning the phase transition, we

observed a change in LEED spot size in going through the phase transition, indicating that the transition involves a change in surface morphology.

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