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Electronic and magnetic properties of Heusler alloy Co$_2$MnSi epitaxial ultrathin films facing a MgO barrier studied by x-ray magnetic circular dichroism

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The electronic and magnetic states of an epitaxially grown full-Heusler alloy Co$_2$MnSi (CMS) 1.1 nm (4 ML) thick ultrathin film and a CMS 50-nm-thick film both facing an epitaxial MgO(001) tunnel barrier were element-specifically studied by means of x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). The observed XAS and XMCD spectra revealed that both the CMS films were not oxidized. The Co and Mn spin magnetic moments for the 4 ML thick CMS film obtained by applying the sum rules were close to those for the 50-nm-thick one and the theoretical values for bulk CMS with the L$_2^1$ structure. These results indicate that 4 ML thick CMS ultrathin films facing a MgO barrier still retain the electronic and magnetic states for the L$_2^1$ structure. © 2008 American Institute of Physics. [DOI: 10.1063/1.2836676]

Co-based Heusler alloys (Co$_2$YZ) are promising ferromagnetic electrode materials for spintronic devices. This is because of their potentially high spin polarization arising from the half-metallic ferromagnetic nature theoretically predicted for some of these alloys, and because of their high Curie temperatures, which are well above room temperature (RT). We have recently developed fully epitaxial magnetic tunnel junctions (MTJs) with a Co$_2$YZ thin film and a MgO(001) tunnel barrier, and demonstrated a relatively high tunnel magnetoresistance ratio at RT. The spin-dependent tunneling characteristics in MTJs are very sensitive to the electronic and magnetic states of the interfacial region of ferromagnetic electrodes with a tunnel barrier. Recently, x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) have proved to be effective techniques for obtaining microscopic information about the element-specific electronic and magnetic states in the interfacial region of MTJs. In this work, we fabricated an epitaxial Co$_2$MnSi (CMS) 1.1 nm (4 ML)-thick ultrathin film and a CMS 50-nm-thick film with an epitaxial MgO barrier in order to investigate the interfacial region selectively by XAS and XMCD measurements at the Mn-L$_{2,3}$ and Co-L$_{2,3}$ edges.

The sample layer structure was grown on a MgO(001) single-crystal substrate and, from the substrate side, consisted of MgO buffer (10 nm)/Fe underlayer (50 nm)/CMS ultrathin film (4 ML)/MgO barrier (2 nm)/Ru cap (2 nm). A monolayer of Co$_2$MnSi consists of a Co plane and a Mn–Si plane, and a unit cell of CMS, whose lattice parameter is 0.5654 nm (Ref. 13), corresponds to 2 ML of CMS. We grew the 4-ML-thick CMS ultrathin film on the Fe underlayer to stabilize its ferromagnetism at RT. Each layer in the sample layer structure was successively deposited in an ultrahigh vacuum chamber (base pressure: about 6 × 10$^{-8}$ Pa) through the combined use of magnetron sputtering (for Fe, CMS, and Ru) and electron beam evaporation (for MgO); the fabrication procedure was the same as in previous work. The CMS film deposited at RT were annealed in situ at 325 °C for 15 min. The composition of the CMS film used in this study was determined as Co$_2$Mn$_{0.97}$Si$_{0.93}$ through inductively coupled plasma analysis with an accuracy of 2%–3% for the composition of each element. In situ reflection high-energy electron-diffraction (RHEED) observations indicated that the CMS film had the L$_2^1$ structure. As a reference sample, we prepared the same layer structure except that a 50-nm-thick CMS film was deposited instead of a 4-ML-thick CMS film.

XAS and XMCD spectra were measured at RT with the total electron yield method using circularly polarized synchrotron radiation at the KEK Photon Factory (BL-11A). The degree of circular polarization of incident light was set to 87% (±4%). XAS spectra for opposite magnetic-field directions were acquired consecutively with the photon helicity fixed. XMCD is defined as the difference between the two spectra with the photon helicity parallel ($\mu_+$) and antiparallel ($\mu_-$) to the 3d majority-spin directions. A magnetic field $B$ strong enough to saturate the sample’s magnetization was applied perpendicular to the film surface using a superconducting magnet ($B = \pm 2$ T for the 50-nm-thick CMS film and $\pm 3$ T for the 4-ML-thick film).

Figures 1(a)–1(d) show XAS (left panel) and XMCD (right panel) spectra at the Co-L$_{2,3}$ edges for Fe/CMS film (x nm)/MgO, where x=50 nm [(a) and (c)] and 1.1 nm (4 ML) [(b) and (d)]. The XAS and XMCD intensities were normalized by the Co-L$_3$ XAS peak intensity with a linear
background subtracted in the XAS. In the XAS spectrum for the 50-nm-thick CMS film, there is a small distinct shoulder at the higher photon energy (about 3 eV) side of the Co-$L_3$ peak. This shoulder at the Co-$L_3$ peak is characteristic of full-Heusler alloys (Co$_2$YZ) with the $L_2_1$ structure and has been commonly observed for bulk samples$^{15}$ or thin films.$^{9,12}$ The XAS and XMCD spectra for the 4-ML-thick CMS film are similar to those for the 50-nm-thick film in terms of the structure and magnitude. The shoulder at the Co-$L_3$ peak which is characteristic of Heusler alloys with the $L_2_1$ structure is also observed in the XAS spectrum for the 4-ML-thick film, though it is not as clear as the shoulder for the 50-nm-thick film. In addition, Co atoms in both CMS films were not oxidized, because the XAS spectrum of CoO exhibits a characteristic multiplet structure$^{16–18}$ and such a multiplet structure was not observed in either film. These results indicate that there is hardly any difference in the electronic band structure of the two films.

Figures 2(a)–2(d) show XAS (left panel) and XMCD (right panel) spectra at the Mn-$L_{2,3}$ edges for Fe/CMS film ($x$ nm)/MgO, where $x=50$ nm [(a) and (c)] and 1.1 nm (4 ML) [(b) and (d)]. The XAS and XMCD intensities were normalized by the Mn-$L_3$ XAS peak intensity. The XAS and XMCD spectra for the 4-ML-thick CMS film are similar to those for 50-nm-thick film in terms of the structure and magnitude. In Figs. 2(a) and 2(b), there is a doublet structure in the Mn $L_2$ region in the XAS spectra, which is characteristic of Co$_2$Mn$Z$ ($Z$=Si,Ge). In Figs. 2(a)–2(d), there is a broad structure in the XAS and XMCD spectra at the higher photon energy (about 2–10 eV) region of the Mn-$L_3$ XAS peak, which is also commonly observed for Co$_2$Mn$Z$. The XAS and XMCD spectra in Figs. 2(a)–2(d) indicate that no MnO existed in either film, because the formation of MnO in CMS films causes a characteristic multiplet structure in an XAS spectrum$^{19}$ and a serious reduction of the XMCD signals at the Mn edges, neither of which are observed. The XAS and XMCD spectra at the Mn edges indicate again that there is hardly any difference in the electronic band structure of the two films.

By applying the sum rules$^{20}$ to the XAS and XMCD spectra, we obtained the magnetic moments of Mn and Co.

![FIG. 1.](image1.png)

![FIG. 2.](image2.png)
atoms. The orbital ($m_{\text{orb}}$) and spin ($m_{\text{spin}}$) magnetic moments are given as

$$m_{\text{orb}} = \frac{4}{3} q r n_p \mu_B,$$

(1)

$$m_{\text{spin}} + 7 m_{\text{orb}} = -\frac{6 p - 4 q}{r} n_p \mu_B,$$

(2)

where $r$ is the XAS energy integral, $r = \int_{L_3}^{L_2} (\mu_+ + \mu_-) d\omega$, $q$ is the XMCD energy integral over the $L_2,3$ edges, and $q = \int_{L_2}^{L_3} (\mu_+ - \mu_-) d\omega$. The $p$ is the XMCD energy integral over the $L_1$ edge, expressed as $p = \int_{L_1} (\mu_+ - \mu_-) d\omega$, and $m_{\text{orb}} = \langle T_z \rangle \mu_B / h$ with $\langle T_z \rangle$ being the expectation value of the intratomic magnetic dipole operator.\(^\text{20}\) For the evaluation of $m_{\text{spin}}$ and $m_{\text{orb}}$ for the CMS films facing a MgO barrier, we assumed, on the basis of a band structure calculation,\(^\text{21}\) a 3d hole number $n_H$ of 4.52 for Mn atoms and 2.24 for Co atoms and ignored the $\langle T_z \rangle$ term, assuming the term to be very small. For Mn atoms, the spin moment obtained should be corrected for the effect of the $jj$ mixing arising from $2p$-$3d$ electrostatic interaction.\(^\text{9,22}\) However, we estimated $m_{\text{spin}}$ of Mn atoms in the CMS films using the bare spin sum rule because a correction factor of Mn atoms in CMS with the $L_2^1$ structure is not accurately known.

The obtained $m_{\text{spin}}$ and $m_{\text{orb}}$ values for Co and Mn atoms for CMS films are listed in Table I. The $m_{\text{spin}}$ values of 1.16 $\pm$ 0.1 $\mu_B$ (Co) and 2.95 $\pm$ 0.1 $\mu_B$ (Mn) for the 50-nm-thick CMS film are very close to the respective theoretical values\(^\text{23}\) of 1.06 $\mu_B$ and 2.92 $\mu_B$ for bulk CMS with the $L_2^1$ structure. On the other hand, the $m_{\text{orb}}$ values for the 50-nm-thick CMS film are very small [0.02 $\mu_B$ (Co) and 0.08 $\mu_B$ (Mn)]. These small $m_{\text{orb}}$ values indicate a quenching of the orbital magnetic moment due to the cubic symmetry in CMS with the $L_2^1$ structure.\(^\text{23}\) For the 4-ML-thick CMS ultrathin film facing a MgO barrier, the $m_{\text{spin}}$ values are 1.25 $\pm$ 0.1 $\mu_B$ (Co) and 3.35 $\pm$ 0.1 $\mu_B$ (Mn). These $m_{\text{spin}}$ values are close to those for the 50-nm-thick CMS film and to the theoretical values for bulk CMS with the $L_2^1$ structure, indicating that 4-ML-thick (=2 unit cell) CMS ultrathin films still retain the electronic band structure in CMS with the $L_2^1$ structure.\(^\text{24}\)

In summary, we element-specifically studied the electronic and magnetic states of an epitaxially grown full-Heusler alloy CMS 4-ML-thick ultrathin film and a CMS 50-nm-thick film both facing an epitaxial MgO(001) tunnel barrier by means of x-ray absorption spectroscopy and x-ray magnetic circular dichroism. The observed XAS and XMCD spectra revealed that both CMS films were not oxidized. The Co and Mn spin magnetic moments for the 4-ML-thick CMS film obtained by applying the sum rules were close to those for the 50-nm-thick one and the theoretical values for bulk CMS with the $L_2^1$ structure. These results indicate that 4-ML-thick CMS ultrathin films facing a MgO barrier still retain the electronic and magnetic states for the $L_2^1$ structure.


\(^{21}\)M. Galanakis, Ref. 29 in Ref. 9.

