<table>
<thead>
<tr>
<th>Title</th>
<th>Detection of the valence band in buried Co$_2$MnSi–MgO tunnel junctions by means of photoemission spectroscopy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Author(s)</td>
<td>Fecher, Gerhard H.; Balke, Benjamin; Gloskowskii, Andrei; Ouardi, Siham; Felser, Claudia; Ishikawa, Takayuki; Yamamoto, Masafumi; Yamashita, Yoshiyuki; Yoshikawa, Hideki; Ueda, Shigenori; Kobayashi, Keisuke</td>
</tr>
<tr>
<td>Citation</td>
<td>Applied Physics Letters, 92(19): 193513</td>
</tr>
<tr>
<td>Issue Date</td>
<td>2008-5-16</td>
</tr>
<tr>
<td>Doc URL</td>
<td><a href="http://hdl.handle.net/2115/50605">http://hdl.handle.net/2115/50605</a></td>
</tr>
<tr>
<td>Rights</td>
<td>Copyright 2008 American Institute of Physics. This article may be downloaded for personal use only. Any other use requires prior permission of the author and the American Institute of Physics. The following article appeared in Appl. Phys. Lett. 92, 193513 and may be found at <a href="http://link.aip.org/link/?apl/92/193513">http://link.aip.org/link/?apl/92/193513</a></td>
</tr>
<tr>
<td>Type</td>
<td>article</td>
</tr>
<tr>
<td>File Information</td>
<td>APL92_193513.pdf</td>
</tr>
</tbody>
</table>
Detection of the valence band in buried Co$_2$MnSi–MgO tunnel junctions by means of photoemission spectroscopy

Gerhard H. Fecher,1 Benjamin Balke,1 Andrei Gloskowskii,1 Siham Ouardi,1 Claudia Felser,1,8 Takayuki Ishikawa,2 Masafumi Yamamoto,2 Yoshiyuki Yamashita,3 Hideki Yoshikawa,3 Shigenori Ueda,3 and Keisuke Kobayashi3
1Institut für Anorganische und Analytische Chemie, Johannes Gutenberg-Universität, 55099 Mainz, Germany
2Division of Electronics for Informatics, Hokkaido University, Sapporo 060-0814, Japan
3National Institute for Materials Science, SPring-8, Hyogo 679-5148, Japan

(Received 13 February 2008; accepted 15 April 2008; published online 16 May 2008)

This work reports on the detection of the valence band of buried Heusler compounds by means of hard x-ray photoemission spectroscopy. The measurements have been performed on the so-called “half” tunnel junctions that are thin films of Co$_2$MnSi underneath MgO. Starting from the substrate, the structure of the samples is MgO(buffer)–Co$_2$MnSi–MgO($t_{MgO}$)–AlO$_x$ with a thickness $t_{MgO}$ of the upper MgO layer of 2 and 20 nm. The valence band x-ray photoemission spectra have been excited by hard x-rays of about 6 keV energy. The valence band spectra have been used to estimate the mean free path of the electrons through the MgO layer to be 17 nm at kinetic energies of about 6 keV. In particular, it is shown that the buried Co$_2$MnSi films exhibit the same valence density of states as in bulk samples. © 2008 American Institute of Physics. [DOI: 10.1063/1.2931089]

The X$_2$YZ (X = transition metal, Y = main group element) Heusler compounds have attracted scientific and technological interest for their potential use as materials for magneto-electronic devices. The reason for this is the exceptional electronic structure found in particular in the Co$_2$YZ Heusler compounds (see Refs. 1 and 2, and references therein). Kübler et al.3 concluded from ab initio calculations that Co$_2$ based compounds would have peculiar transport properties when they recognized that the minority-spin density at the Fermi energy $\varepsilon_F$ vanishes in Co$_2$MnAl and Co$_2$MnSn. That means that those compounds exhibit a complete spin polarization at $\varepsilon_F$ such that they behave like a metal for electrons of one spin direction and like an insulator for the other one. Therefore, these compounds are called half-metallic ferromagnets.4

One major research area where Co$_2$ based Heusler compounds are used is on tunneling magnetoresistive (TMR) junctions (see Ref. 5 for a review on other materials). For materials with complete spin polarization, an infinite TMR ratio is expected. Structural and magnetic properties of Co$_2$MnSi have been reported for films and single crystals6–8 and the compound was suggested to be suitable for magnetic tunnel junctions (MTJs). For example, the end members of the series Co$_2$Mn$_{1-x}$Fe$_x$Si (x = 0 and 1) have been used for fabrication of MTJs.9–11 Sakuraba et al. measured a high TMR ratio of 570% at 2 K (67% at room temperature) for junctions with Co$_2$MnSi films as both lower and upper electrodes and with an AlO$_x$ tunnel barrier.12 Some of the present authors recently developed fully epitaxial MTJs with a Co$_2$YZ thin film as a lower electrode and a MgO tunnel barrier.13–15 At room temperature (RT), relatively high TMR ratios of 109% (317% at 4.2 K) for Co$_2$Cr$_{0.6}$Fe$_{0.4}$Al/MgO/CoFe MTJs (Ref. 14) and 90% (192% at 4.2 K) for Co$_2$MnSi/MgO/CoFe MTJs (Ref. 13) were reported. Furthermore, high TMR ratios of 179% at RT and 683% at 4.2 K were demonstrated for fully epitaxial MTJs with Co$_2$MnSi films as both lower and upper electrodes and MgO tunnel barrier.16 The high quality of the interface in Co$_2$MnSi films with MgO barrier was proved by x-ray absorption spectroscopy and x-ray magnetic circular dichroism.17

To develop high performance spintronic devices with Co$_2$-based Heusler compound thin films as ferromagnetic electrodes, it is essential to clarify the electronic structures of these films in terms of the half-metallic nature. The purpose of the present study was to investigate the valence band electronic structure of Co$_2$MnSi thin films buried underneath a MgO barrier by means of hard x-ray photoemission spectroscopy (HAXPES).

For the present study, special multicomponent thin film arrangements were produced that correspond to half of a MTJ as used in TMR devices (see Refs. 18–20). In particular, the free electrode was modeled. The fabricated sample layer structure was as follows: MgO(001) substrate/MgO buffer layer (10 nm)/Co$_2$MnSi (50 nm)/MgO barrier ($t_{MgO}$)/AlO$_x$(t$_{AlO_x}$) cap. $t_{MgO}$ = 2 nm, 20 nm and $t_{AlO_x}$ = 1 nm, 2 nm were chosen for the thickness of the MgO and AlO$_x$ cap, respectively. The topmost AlO$_x$ was used for protection of the MgO layer. 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 Co$_2$MnSi and Al, and electron beam evaporation for MgO. The fabrication procedure was the same as in previous work.13–15 The Co$_2$MnSi film was deposited at 300 K and subsequently annealed in situ at 870 K for 15 min. The MgO barrier (2 or 20 nm) was deposited at 300 K. X-ray pole figure measurements confirmed that the fabricated Co$_2$MnSi film was grown epitaxially and was a single crystal with the L2$_1$ structure.18 It was also proved that the MgO barrier was grown epitaxially and was single crystalline from reflection high-energy electron diffraction patterns18 and cross-sectional high-resolution trans-

---

8Electronic mail: fecher@uni-mainz.de.
mission electron microscope lattice images. The AlO₃ cap was prepared by exposing the sputter-deposited Al layer to an O₂ atmosphere of (~1 × 10⁵ Pa for 2 h). Al 1s core level spectra (not shown here) revealed that the 1 nm thick Al layer was completely oxidized to Al₂O₃, whereas the 2 nm layer had a remaining, small contribution of metallic Al. Polycrystalline bulk material was used for comparison. It was produced, prepared, and characterized as described in Ref. 21.

The HAXPES experiments were performed at the beamline BL15XU of SPring-8. The photon energy was fixed at 5.946 77 keV using a double crystal monochromator and a Si(333) postmonochromator. The photoemitted electrons were analyzed for their kinetic energy and detected by means of a hemispherical analyzer (Scien'ta). The overall energy resolution (monochromator plus analyzer) was set to 250 meV, as verified by spectra of the Au valence band at the Fermi energy (εₓ). Additionally, spectra close to the Fermi energy were taken with a resolution of 150 meV. The angle between the electron spectrometer and photon propagation is fixed at 90°. The photons are polarized, that is, the electric field vector is in the plane of incidence and always pointing in the direction of the electron detector. For the thin films, a near normal emission (θ = 2°) detection angle was used. This corresponds to an angle of incidence of α=θ−90°≈88°. (Note that the angles are not that well defined for the cleaved bulk sample due to surface roughness.)

Figure 1 compares the valence band spectra from the 50 nm thick Co₂MnSi layer buried underneath MgO/AlO₃ with different thickness of the MgO interlayer with spectra from bulk material. The valence band spectra of the thin films agree well with those reported from Co₂Mn₁₋ₓFeₓSi bulk samples, even though the emission is strongly suppressed in the film with the 20 nm thick MgO layer. In particular, the peak in the density of states at −1.3 eV below εₓ is clearly resolved (see Fig. 2). The low lying s-band below −8 eV is only seen in Figs. 1(a) and 1(c). This part, in addition to the lower parts of the p and d bands, is covered by the emission from oxygen 2p states in MgO in Fig. 1(b) where the MgO film is 20 nm thick. The similar structure of the valence band close to εₓ in the thin films and the bulk material is obvious from Fig. 2.

The mean electron free path (λ) in MgO is expected to be about 9 nm at 6 keV energy, as calculated from the TPP2M equation (Tanuma–Powell–Penn). In Al₂O₃, one has a mean free path of 11 nm at the same kinetic energy, which should be much smaller in Co₂MnSi (7 nm). From those values, one expects that the electrons emitted from the Fermi energy to easily penetrate the thin MgO and AlO₃ films but not through the 50 nm thick Co₂MnSi layer. The electron mean free path is easily extracted from the experiment by comparing the intensities for different thickness of the MgO layer. Figure 3(b) displays the relative intensities I₂₀⁻/I₂₀ nm. It is seen that the factor I₂₀⁻/I₂₀ nm is constant for energies between −4 eV and εₓ. The scaling factor amounts to I₂₀⁻/I₂₀ nm = 2.878 ± 0.016 on average. The constant intensity ratio is expected as MgO does not contribute to the emission close to the Fermi energy due to its wide band gap. From the exponential decay of the intensity I(z) ∝ exp (−z/λ) with the layer thickness z, one finds the electron
mean free path in the MgO layer from $\lambda = \Delta E / \ln(I_{20\text{ nm}}/I_{20\text{ nm}})$ to be $\lambda_{\text{MgO}} = 17\text{ nm}$.

In Fig. 3(a), the valence band spectra for different MgO thicknesses are directly compared by scaling up the intensity of the 20 nm layer. The identical structure of the spectra, in particular close to the Fermi energy, is easily recognized. The high intensity with a center at about $-1.3\text{ eV}$ is due to emission from bulk samples. 21,22 The Co $t_{2g}$ like states mainly define the upper energy of the minority valence bands, whereas the lower intensity at the Fermi energy arises from deep delocalized majority $d$ bands with $t_{2g}$ character at $\Gamma$.2,24

The value observed here for the electron mean free path in MgO is about twice as large compared to the one calculated from the TPP2M equation. This is explained by the high crystalline order of the MgO film and its large optical gap which prevents inelastic scattering mediated by interband transitions. The large mean free path makes it possible to look through the material of the tunnel barrier as seen in Figs. 2(b) and 3.

In summary, the present study demonstrates the feasibility of HAXPES to explore the valence band electronic structure in deeply buried metallic layers. In particular, it was shown that buried Co$_2$MnSi films exhibit the same valence density of stateslike bulk samples that is typical for half-metallic ferromagnets. This result confirms the promise of an epitaxial, single-crystalline Co$_2$-based Heusler compound film as a ferromagnetic electrode for spintronic devices.

This work was funded by the Deutsche Forschungs-Gemeinschaft (P7-FOR 559). The work at Hokkaido University was partly supported by a Grant-in-Aid for Scientific Research (B) (No. 18360143) and a Grant-in-Aid for Scientific Research on Priority Areas (No. 19048001) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan. The HAXPES measurements were performed under the approval of NIMS Beamline Station (Proposal Nos. 2007A4903, 2007B4907). The authors are grateful to HiSOR, Hiroshima University and JAEA/SPring-8 for the development of HAXPES at BL-15XU.