

Surface resonance of thin films of the Heusler half-metal Co_2MnSi probed by soft x-ray angular resolved photoemission spectroscopy

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Heusler compounds are promising materials for spintronics with adjustable electronic properties including 100% spin polarization at the Fermi energy. We investigate the electronic states of AlO_x capped epitaxial thin films of the ferromagnetic half-metal Co_2MnSi *ex situ* by soft x-ray angular resolved photoemission spectroscopy (SX-ARPES). Good agreement between the experimental SX-ARPES results and photoemission calculations including surface effects was obtained. In particular, we observed in line with our calculations a large photoemission intensity at the center of the Brillouin zone, which does not originate from bulk states, but from a surface resonance. This provides strong evidence for the validity of the previously proposed model based on this resonance, which was applied to explain the huge spin polarization of Co_2MnSi observed by angular-integrating UV-photoemission spectroscopy.

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I. INTRODUCTION

The design and control of specific electronic properties of metallic thin films is a major requirement for the development of powerful spin based electronics (spintronics). Due to their compositional tunability Heusler compounds represent a prime example for such an optimization of electronic states, which is usually based on band-structure calculations.

Angular resolved photoemission spectroscopy (ARPES) is a well established method providing direct access to the electronic states. However, specifically in the case of Heusler compounds, this proved to be very challenging: These materials do not cleave well, which is the standard method for the preparation of clean sample surfaces for ARPES. Additionally, the reactivity of the elements of Heusler compounds results in the surface degrading faster than the typical time frame for a full ARPES experiment.

The half-metallic ferromagnet Co_2MnSi [1,2] represents a typical example for this class of materials. Photoemission spectroscopy investigations of this and other Heusler compounds focused mainly on measurements of the spin polarization. Whereas the investigation of *ex situ* prepared samples resulted in small polarization values only [3,4], *in situ* spin-resolved and angular integrated UV-photoemission spectroscopy of epitaxial Co_2MnSi thin films demonstrated a large spin polarization close to 100% [5]. This large spin polarization was measured in a range of binding energies much broader than expected by bulk band band-structure calculations, which is consistent with photoemission calculations including surface effects and predicting a highly spin-polarized surface resonance at the Fermi energy. However,

direct experimental evidence for this surface resonance is required, as the angular integrated photoemission data did not show sufficient characteristic features for a detailed comparison with theory.

Without spin analysis the investigation of *ex situ* prepared capped thin-film samples has proven to be possible by less surface sensitive hard x-ray photoemission spectroscopy (HAXPES) [6–8].

Here we demonstrate that by *ex situ* soft x-ray angular resolved photoemission spectroscopy (SX-ARPES) dispersive electronic states are observed investigating epitaxial $\text{Co}_2\text{MnSi}(001)$ thin films capped by 1.8 nm of AlO_x . The experimental data allow for a more reliable test of the band-structure calculations than the comparison with previous angular integrated photoemission experiments [5,9]. Here we provide strong direct evidence for the validity of the calculated photoemission spectra previously used to explain the close to 100% spin polarization observed in angular integrated UV photoemission [5,9]. We show that a surface resonance dominates the ARPES signal in Co_2MnSi in the soft x-ray regime (600–1200 eV excitation energy).

II. TECHNIQUES

Our epitaxial $\text{Co}_2\text{MnSi}(001)$ Heusler thin films (thickness 35 nm) were prepared by rf-sputtering on $\text{MgO}(001)$ substrates and capped by 1.8 nm of polycrystalline or amorphous AlO_x , as described elsewhere [8].

SX-ARPES investigations of these samples were performed at the ADDRESS beamline of the Swiss Light Source at

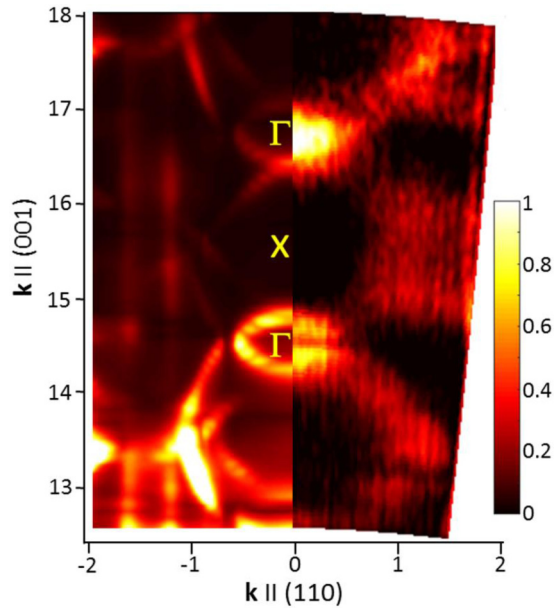


FIG. 1. Photon energy and momentum k_{\parallel} (110) dependence of the photoemission intensity at the Fermi energy obtained investigating an epitaxial $\text{Co}_2\text{MnSi}(001)$ thin film. The left part of the image represents the calculation, the right part the experimental photoemission data. The color scale shows the ARPES intensity in arbitrary units.

the Paul Scherrer Institute [10] using circularly polarized soft x-ray photons. The end station uses an experimental geometry with 20° grazing light incidence angle [10]. With a hemispherical energy analyzer (Specs PHOIBOS 150) the photoemission intensity was recorded as a function of the binding energy and momentum parallel to the thin-film surface. With a photon energy of 1 keV, the energy and momentum resolution amount to 200 meV and $\simeq 0.02 \text{ \AA}^{-1}$. The measurements were carried out at 12 K to reduce thermal broadening effects [11].

The experimental results were compared with photoemission calculations based on *ab initio* spin-density functional theory with local-density approximation. As in our previous work, discussing angular integrated photoemission spectroscopy on Co_2MnSi , the electronic structure of $\text{Co}_2\text{MnSi}(001)$ was computed for a semi-infinite system including surface related effects using the fully relativistic Korringa-Kohn-Rostoker method as implemented in the Munich SPR-KKR package [12,13]. All technical details can be found in Refs. [5,9].

III. SX-ARPES EXPERIMENTS AND CALCULATIONS

For the identification of the center of the Brillouin zone (Γ point), the photon energy was scanned in an energy range from 580 to 1200 eV. The corresponding plot of the photoemission intensity integrating from $E_b = -0.2$ eV to the Fermi energy is shown in Fig. 1 (right panel) in comparison with corresponding one-step model photoemission calculations (left panel) as described above. This corresponds to a cut through the Fermi surface perpendicular to the sample surface, as for a given binding energy the photon energy selects the component k_z of the crystal momentum perpendicular to the

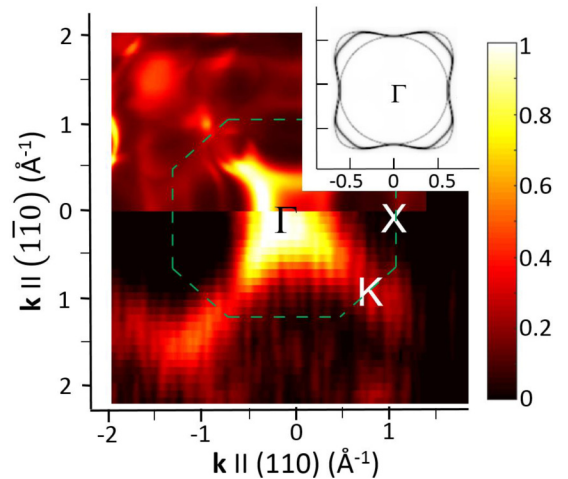


FIG. 2. Cut through the Fermi surface of $\text{Co}_2\text{MnSi}(001)$ in a reciprocal space plane parallel to the sample surface through the Γ point. The bulk Brillouin-zone boundary as well as the X and K points are indicated. The color scale shows the ARPES intensity in arbitrary units. Top panel: calculation including surface effects; bottom panel: experimental photoemission data. The inset (same scale as main figure) shows the corresponding calculated bulk Fermi surface of Co_2MnSi (see discussion in Sec. IV).

sample surface. Assuming a free-electron-like final state of the photoexcitation process, the photon energy can be converted into k_z [14,15]. However, within our one-step model photoemission calculations, no such approximations are required and the photon energy dependence of the ARPES intensity is calculated directly as shown in Fig. 1. Good agreement of the calculated (left panel) and experimental data (right panel) is found allowing the identification of the center of the Brillouin zone (Γ point), which corresponds to a photon energy of 1020 eV.

To add further evidence for the validity of our combined band-structure and photoemission calculations, we additionally show the momentum parallel to the sample surface, i.e., k_{\parallel} , dependence of the SX-ARPES intensity obtained with a photon energy of 1020 eV and integrating from $E_b = -0.2$ eV to the Fermi energy. This corresponds to a cut through the Fermi surface in a reciprocal space plane parallel to the sample surface through the Γ point. In Fig. 2, the experimental data (bottom panel) are shown next to the corresponding calculated intensity distribution (top panel). Again, good agreement concerning the large spectral weight at the Γ point is obtained.

Finally, in Fig. 3, we show dispersing electronic valence states with momentum k_{\parallel} (110), i.e., along the Γ - K direction (upper panel) and for k_{\parallel} (100), i.e., along the Γ - X direction (lower panel). Specifically, close to the Fermi energy for binding energies below -1 eV good agreement between experimental data (right) and calculation (left) is obtained, which is consistent with the good agreement of the calculated and measured Fermi-surface cuts.

For larger binding energies > 1 eV, no comparison of the experimental data with the calculations is possible, as no dispersive features are observed by SX-ARPES. Instead, at a binding energy of 1 eV a k -independent large photoemission

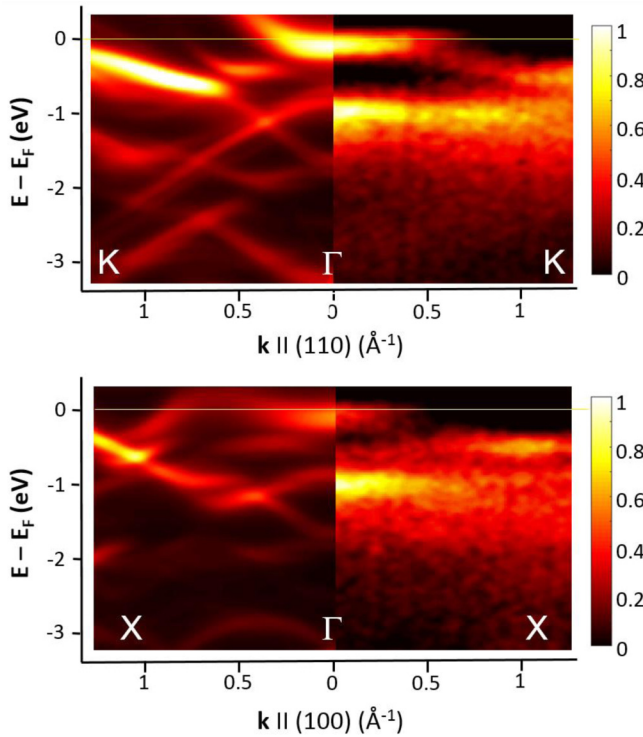


FIG. 3. Dependence of the calculated (left panel) and experimental (right panel) photoemission intensity on the binding energy $E - E_F$ and on the photoelectron momentum parallel to the sample surface. Top panel: $k \parallel (110)$; bottom panel: $k \parallel (100)$. The color scale shows the ARPES intensity in arbitrary units.

intensity is experimentally observed, which could be associated to photoemission from the AlO_x capping layer.

IV. SURFACE RESONANCE VS BULK STATES

At the Fermi energy we consistently observe, in agreement with our photoemission calculations, a large photoemission intensity at the Γ point, which vanishes with increasing values of k for both directions parallel (110) and (100) to energies above the Fermi edge. However, in most published calculations of the band structure of Co_2MnSi no electronic bulk states are present at E_F in the region around the Γ point. Typically, the calculated bulk majority bands cross the Fermi energy along the Γ - X direction, i.e., for $k \parallel (100)$ close to the X point, whereas the minority states show a gap with the Fermi energy either in the center or close to the edge of the band gap [16–20]. This is consistent with our own calculations of the bulk band structure of Co_2MnSi and we obtain a bulk Fermi surface with a radius of $\simeq 0.7 \text{ \AA}^{-1}$ as shown in the inset of Fig. 2. If the bulk states would dominate the ARPES data, the maximum ARPES intensity would be expected around, but not at the Γ point. Thus the observation of maximum spectral weight at the Γ point provides evidence for surface states dominating the ARPES intensity.

Nevertheless, as the minority states at the Γ point in the bulk band structure are close in energy to E_F , they in principle could show up in the experimental SX-ARPES Fermi surface due to energy broadening effects. Also the surface resonance

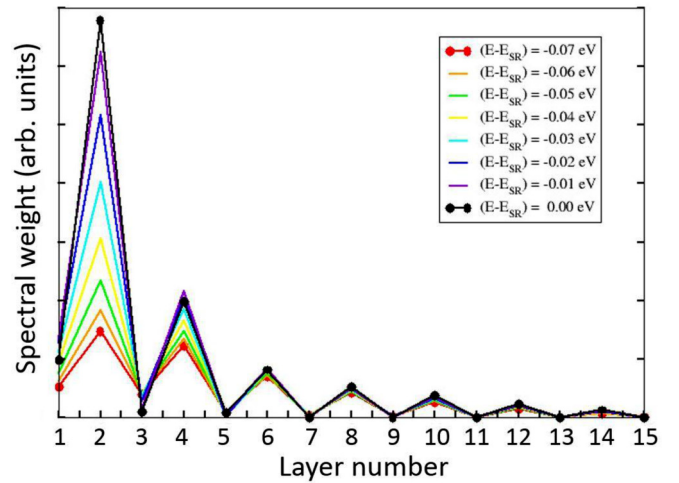


FIG. 4. Spectral weight of different electronic states plotted as a function of the initial-state energy, from the energy $E - E_{\text{SR}} = 0 \text{ eV}$ (black), indicating the maximum in the amplitude of the surface resonance intensity distribution, to $E - E_{\text{SR}} = -0.07 \text{ eV}$ (red), corresponding to bulklike emission.

discussed in our previous work to explain the close to 100% spin polarization observed by spin-resolved angle-integrating UV-photoemission spectroscopy (SRUPS) [5] is situated close to, but above, the Fermi energy of Co_2MnSi [9].

The comparison with the experimental data shown in Fig. 4 serves to determine the Fermi energy. Thus, for our photoemission calculations we have renormalized the Fermi level by a rigid energy shift of 0.2 eV to account for a well-known shortcoming of the local density approximation (LDA). Using the LDA one systematically underestimates self-energy effects in the electronic structure of simple metals [21]. We deduce that the surface resonance is situated about 0.15 eV above the Fermi energy, whereas the bulk minority states are found about 0.25 eV below the Fermi energy.

Additional to being closer in energy to E_F as the bulk minority states, the surface resonance also contributes much stronger to the SX-ARPES intensity than all other electronic states. To elucidate its character we calculated the layer dependence of the SX-ARPES intensity: A prototypical surface resonance splits off in energy from the corresponding bulk bands and disperses in the vicinity of these bulk states. The resonance shows up with a considerable spectral weight within the first three or four atomic layers. Due to its bulk contribution [22], the spectral weight of a resonance is in general much larger than that of a real surface state. Thus, the surface resonance can be observed even with soft x-ray excitation of about 1 keV. At these energies the inelastic mean free path of about $\simeq 25 \text{ \AA}$ [23] allows for a layer-dependent study of the spectral distribution of the various electronic states, in order to estimate the fraction surface and bulklike contributions to the total intensity distribution. Correspondingly, we performed layer-resolved one-step photoemission calculations in normal emission to reveal the inelastic mean free path (IMFP) of the surface resonance.

The result is shown in Fig. 4, where the absolute value of the photoemission intensity is plotted as a function of the

atomic layer number starting from the surface (layer 0). This is shown for different initial-state energies, from the energy of the surface resonance E_{SR} to larger binding energies. It is clearly visible that the spectral weight in the first four layers is considerably enhanced. For initial-state energies close to E_{SR} the surface resonance shows up with the maximum intensity in the photoemission spectrum. For other energies the IMFP is estimated to about 10–12 layers in accordance with [23] and as a consequence indicates bulklike emission. The absolute value of the spectral weight is peaked at layer 2, with a considerable contribution at the fourth layer, where even layer numbers indicate layers containing Co atoms only. This way the surface resonance could be identified as a Tamm-like surface feature that split up in energy from the corresponding Co bulk states.

V. SUMMARY

The Fermi surface of the half-metallic Heusler compound Co_2MnSi , as well as dispersive bands close to the Fermi energy, were investigated by SX-ARPES of epitaxial thin

films capped by a 1.8-nm AlO_x layer. The experiments were compared with photoemission calculations including surface related effects, which results in good agreement. In particular, a large photoemission intensity was obtained at the center of the Brillouin zone, although no electronic state are present around the Γ point in bulk band-structure calculations. Based on our photoemission calculations of the semi-infinite system, this is explained by a surface resonance of $\text{Co}_2\text{MnSi}(001)$ dominating the SX-ARPES intensity. The comparison of theory and experiments, based on angular resolved data, provides direct strong evidence for the existence of this resonance proposed previously to explain the close to 100% spin polarization observed by angular integrated photoemission experiments [5,9].

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