

Direct evidence for minority spin gap in the Co_2MnSi Heusler alloy

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Supplementary information

Calculation Method: Calculations are performed using Vienna ab initio simulation package (VASP) [Kre93, Kre96] with generalized gradient approximation [Wan91] and projected augmented wave pseudopotentials [Kre99, Blö94]. We used the kinetic energy cutoff of 550 eV and a Monkhorst-Pack k-point grid of $11 \times 11 \times 1$ for the surface calculations and $9 \times 9 \times 7$ for the bulk. Initially the structures were relaxed until the force acting on each atom falls below $1 \text{ meV}/\text{\AA}$.

Sample preparation: The sample preparation and SR-PES measurements were performed at the CASSIOPEE beamline of the SOLEIL synchrotron radiation facility. The CMS samples are grown in a MBE set-up connected to the SR-PES chamber. We used 2 e-gun sources for Co and Si and a Knudsen cell for Mn. The growth rates were accurately controlled during the growth using quartz microbalances. The epitaxial process was controlled by using electron surface diffraction (RHEED) and the chemical quality of the films was controlled by Auger spectroscopy available in the MBE chamber ([supfig.1](#)). The CMS surface is very stable since no change on the RHEED patterns was observed after 24h PES measurements; the Auger analysis showed very small surface contamination by C and perhaps O (mixed with the Mn-LMM Auger transitions). This small contamination yet decreases the intensity of the

resonant surface state after long measurements (but not the other PES transitions). The $\frac{1}{2}$ streaks on RHEED pattern along [11] azimuth is due to the chemical order [Neg14]. The CMS surface covering was done by Mn, MnSi or MgO evaporations, calibrated using a quartz microbalance. It should be noted that the $\frac{1}{2}$ streaks discussed above are not seen anymore when Mn or MgO are deposited, which is expected according to Mn and MgO lattices. The MgO deposition is performed using an e-gun source. The MgO growth rate was checked using RHEED oscillations.

Photoemission experiments: Photoemission was carried out in a spin-resolved photoemission set-up using a Mott detector (with a Sherman coefficient equal to 0.12) with a 150 meV energy resolution. The spin detector is equipped with 4 channels that allowed us to measure spectra for both in-plane and out-of-plane spin components. The normal of the sample was oriented along the axis of the detector. The detector aperture is fixed at its maximum and is equal to $\pm 8^\circ$. This corresponds to 50% of the Brillouin Zone (BZ) at 37 eV photon energy at normal incidence. To explore the whole BZ, we rotated the sample off-normal by 8° from the detector axis. PES spectra performed at 37 and 43 eV for 0° and 8° are shown in [supfig.2](#). Similar transitions are observed for both measurement angles, meaning that the whole BZ is explored. Only the surface resonant states (A and B contributions) decreased at 8° off-normal meaning that these states are located along Γ .

To eliminate instrumental asymmetry in spin-resolved measurements, two distinct PES spectra were recorded with opposite magnetizations. As the coercive field of our CMS films was less than 20 Oe, the applied magnetic field was set to 200 Oe to saturate the magnetization prior to PES measurements. We systematically observed a zero out-of-plane spin-polarization, confirming that the magnetization of our CMS films was in-plane. The symmetry of the initial states involved in the transitions observed on SR-PES spectra was determined by measuring successively with s or p polarizations of the incoming photons. As

the photon beam was at 45° from the normal to the sample, *s* polarized photons excite only occupied Δ_5 states, whereas *p* polarized photons excite both occupied Δ_1 and Δ_5 states [Bon12]. The SR-PES analysis was performed using incoming photon energy ranging from 25 to 90 eV. The surface resonant states are observed in a very narrow range of photon energy as shown in [supfig.3](#). This was confirmed by Angle-Resolved Photoemission experiments varying the photon energy from 20 to 110eV.

The PES measurements were performed both at 300K or 80K. We do not observe any significant influence of the sample temperature on the PES results. Around 30 samples were grown and measured during 4 runs of 1 week each.

Figure captions

Sup-fig.1: (a) Typical RHEED patterns on CMS and (b) Auger spectra just after the growth and after PES measurements (24h).

Sup-fig.2: top, schematics showing the Brillouin Zone area explored at 37eV depending on the angle between [001] and the detector axis. Bottom, PES spectra dependence with this angle (0 or 8°) for 37 and 43 eV photon energies.

Sup-fig.3: PES spectra dependence with the photon energy, for both majority and minority spins. The B contribution is observed for photon energies between 35 and 40 eV.