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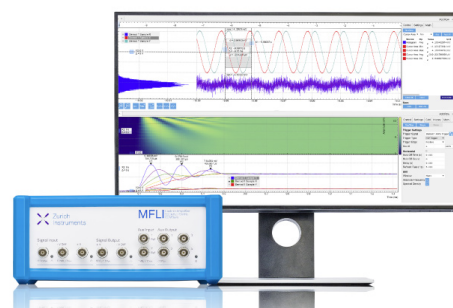
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ABSTRACT

Spin-polarization and magnetic damping are measured for several polycrystalline films with each of them being made of a different single Co₂Mn-based Heusler compound. As several epitaxial Co₂Mn-based Heusler compounds are shown to be half-metal magnetic materials with full spin-polarization and ultralow magnetic damping, we explore here these properties but in polycrystalline films. Co₂MnSi, Co₂MnGe, and Co₂MnGa thin films were grown on glass substrates and analyzed *in situ* by electron diffraction and spin-resolved photoemission and *ex situ* by transmission electron microscopy and ferromagnetic resonance. Despite the polycrystalline state of the films, they still exhibit high spin polarizations and very low magnetic damping coefficients. The latter are at least of the same order as the best epitaxial films using regular ferromagnetic materials. The key point to achieve such properties is to control the Heusler stoichiometry as best as possible.

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About 30 years ago, a new class of magnetic materials was theoretically discovered, called half-metal magnetic (HMM) materials. A HMM material is characterized by a peculiar electronic band structure depending on the electron spin: a gap in the spin projected density of states (DOS) around the Fermi energy E_F is predicted for minority spin (and sometimes for majority spin). Such materials are thus metallic for majority (or minority) spins and insulating for minority (or majority) spins.^{1–3} Moreover, this gap prohibits spin channel exchange, thereby leading to ultralow damping of magnetization precession as predicted theoretically.^{4–6} These two properties together, high spin polarization and low magnetic damping, are very interesting for spintronic applications such as spin-orbit-torque (SOT), spin-transfer-torque (STT), or spin-wave-based (magnonics) devices.^{7–9}

Starting from the theoretical pioneering work by De Groot *et al.*,¹ demonstrating the HMM properties of the Heusler compound NiMnSb, many magnetic Heusler compounds were explored theoretically,^{2,3} and some of them were predicted to be HMM. The experimental evidence of the peculiar electronic properties of HMM Heusler compounds took however a long time to emerge. Although the HMM properties were often claimed to explain the transport properties in Heusler-based devices,^{10–12} the spin gap was only evidenced several

years later—first in Co₂MnSi single-crystalline thin films^{13,14} and very recently in many other Co₂MnZ epitaxial films.¹⁵ The theoretically expected correlation between the values of the magnetic damping coefficient and the width of the spin gap together with the position of E_F in the gap was recently proven,¹⁵ and ultralow magnetic damping coefficients were reported in epitaxial^{14–16} or highly textured¹⁷ Heusler thin films. Record values as low as 4×10^{-4} and 5×10^{-4} were determined for the magnetic damping coefficients of Co₂MnSi and Co₂MnGe, respectively.¹⁵ To this end, the control of the stoichiometry was key to obtaining the best electronic and magnetic properties of these compounds.

The transfer of such properties to devices is not obvious since molecular beam epitaxy (MBE) is not a regular deposition technique used for applications. Sputtering is usually used, and the thin films are often in a polycrystalline state (with some fiberlike texture and, in some very special cases, single-crystalline films can be produced). Nevertheless, the spin dependent gap is predicted in the density of states and should not depend on the crystalline direction. Thus, if the polycrystallinity of the films would not alter the electronic structure of the randomly oriented grains, there would be no fundamental reason for not getting the spin gap with high spin polarization and ultralow damping in sputtered Heusler thin films. Indeed, Shaw and co-workers¹⁷

found small damping in Co_2MnGe films (1.2×10^{-3}) grown by sputtering at room temperature and postannealed. However, the confinement of electrons to the finite size of the grains and the variations of the structural and chemical order across grain boundaries¹⁸ cause spin dependent changes of the electronic structure which could severely affect the spin gap and magnetic damping coefficient of polycrystalline films.

To test this eventuality, we explore in this study the electronic properties of polycrystalline Co_2MnSi , Co_2MnGe , and Co_2MnGa thin films grown by MBE on glass substrates without any buffer layer. The film structure and morphology were characterized *in situ* by using electron diffraction during the growth and *ex situ* by using transmission electron microscopy (TEM). The spin polarization was studied by spin-resolved photoemission spectroscopy (SR-PES), and the magnetic damping coefficients were extracted from ferromagnetic resonance (FMR) experiments.

The Co_2MnSi , Co_2MnGe , and Co_2MnGa Heusler compounds were grown by molecular beam epitaxy (MBE) in exactly the same conditions as used for single-crystalline films,¹⁵ except that the MgO substrate was replaced by a degreased glass substrate. Since glass is amorphous, there is no way to get epitaxy like on MgO(001), and a polycrystalline growth of the Heusler films is expected. We call these films poly-Heusler in the following to distinguish with epitaxial Heusler. Co, Si, and Ge were evaporated using e-guns and Mn and Ga using Knudsen cells. Each compound was obtained by the coevaporation of the three chemical elements. Before the film growth, the atomic fluxes impinging on the substrate were chosen equal to $\phi_{\text{Co}} = 2 \times 10^{14}$ at/cm²/s = $2\phi_{\text{Mn}} = 2\phi_{\text{Si,Ge,Ga}}$ and were accurately calibrated using a quartz microbalance located at the place of the sample. The error on each element concentration is less than 1%. The film thicknesses were fixed to 20 and 40 nm. The growth was processed at around 350 °C. The film structure was observed during the growth by using reflection high energy electron diffraction (RHEED). In the case of a single-crystalline growth, straight truncation rods are observed on the RHEED screen, but the situation is very different if polycrystalline growth takes place. Rings are thus observed exactly like in powder diffraction experiments [Fig. 1(left)]. The observation of such rings is a clear proof of polycrystalline growth, and the indexation of the rings (like in powder diffraction) allows determining the crystallographic structure of the grains constituting the films.¹⁹ The morphology of the films was examined using TEM by extracting a small piece of the substrate covered by the film and deposited on a carbon grid. Using this technique, a top view is possible. An example of the TEM observation performed on the poly- Co_2MnSi film is given in Fig. 1(right). The film is made of single-crystalline grains of 36 nm average size with a dispersion of 10 nm [Fig. 1(a)]. No special texture was observed by TEM [Fig. 1(b)]. The diffraction pattern also shows rings that can be indexed considering the $L2_1$ structure observed in epitaxial films.¹⁵

More information on the chemical ordering can be obtained by looking at grains with the (110) orientation along the e-beam [Fig. 1(c)]. Indeed, the columns for this zone axis come from pure chemical elements (see Ref. 15 for details). Such a High-Angle Annular Dark-Field (HAADF) image clearly shows a mixing of B2 and $L2_1$ chemical ordering into the FCC cell, although only $L2_1$ is observed in epitaxial films.¹⁵ We can go as far as to say that this is not due to some problem of stoichiometry since energy dispersive X-ray spectroscopy on this film leads to 49.9% Co, 25.6% Mn, and 24.7% Si. This phase mixing may rather be attributed to the limited temperature used during the

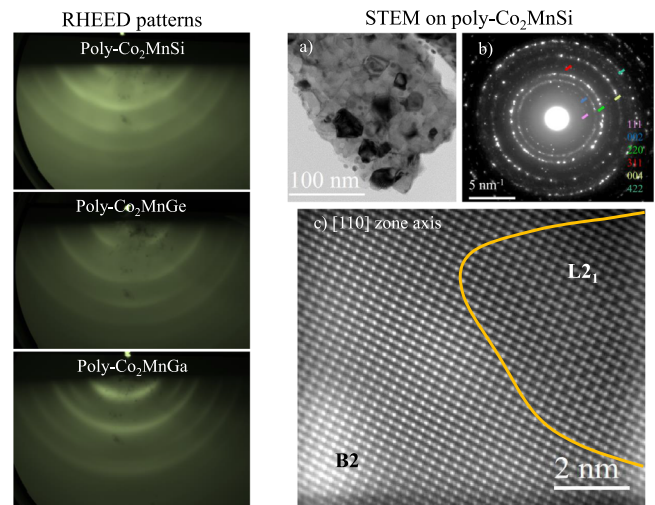


FIG. 1. Left: RHEED patterns at the end of the growth of poly- Co_2MnSi , $-\text{Co}_2\text{MnGe}$, and $-\text{Co}_2\text{MnGa}$ polycrystalline films on glass substrates. Right: TEM observations on the Co_2MnSi film, (a) top view showing the grains, (b) diffraction pattern with the indexation using the $L2_1$ structure, and (c) STEM-HAADF on one grain with the proper alignment to get a [110] zone axis observation: a mixing of B2 and $L2_1$, chemical ordering is clearly observed with the help of the yellow line which is a guide to the eye.

polyfilm growth, insufficient to get the $L2_1$ ordering along the whole film. Note that we did not observe any trace of the A2 phase in this sample since we always observed some columns contrast on HAADF images taken along the [110] zone axis.

The used MBE setup is coupled to the CASSIOPEE beamline at the SOLEIL synchrotron. This beamline is dedicated to photoemission spectroscopy with one end-station for spin-resolved experiments (SR-PES) equipped with a MOTT detector (see Ref. 20 and references therein for details). The SR-PES spectra were obtained at the normal axis of the surface. For epitaxial Heusler already studied on this beamline,^{15,20} only 80% of the Brillouin zone (BZ) was explored due to geometrical limitations (photon energy and detector aperture). This is likely no more the case for polycrystalline films and so the whole BZ and entire electronic band structure is probed. Consequently, a comparison of spin-resolved spectra presented here to those obtained on epitaxial films gives us the opportunity to test the pertinence of PES measurements covering only 80% of the BZ. Another very important point to notice is that PES experiments cannot be performed with an applied magnetic field. Thus, the films were previously magnetized, and the PES spectra were recorded without a magnetic field, i.e., in the remanent state. The remanent magnetization of each film capped with 5 nm Au was thus measured *ex situ* after the PES measurements. In Fig. 2 are shown the SR-PES spectra recorded using photon energy equal to 37 eV and corrected from remanence. The spectra are very similar to the one observed on (001) single-crystalline films¹⁵ except one crucial difference: the transition coming from surface states (note S_1 and S_1 in Fig. 2) usually observed on (001) epitaxial films using this photon energy^{14,15} is much less visible on the polycrystalline films. This can easily be attributed to the low number of properly oriented grains, that is, with one of their (100) crystalline directions parallel to the surface normal of the films. We have shown that these surface states are present in both majority and minority spin channels.^{14,15} The

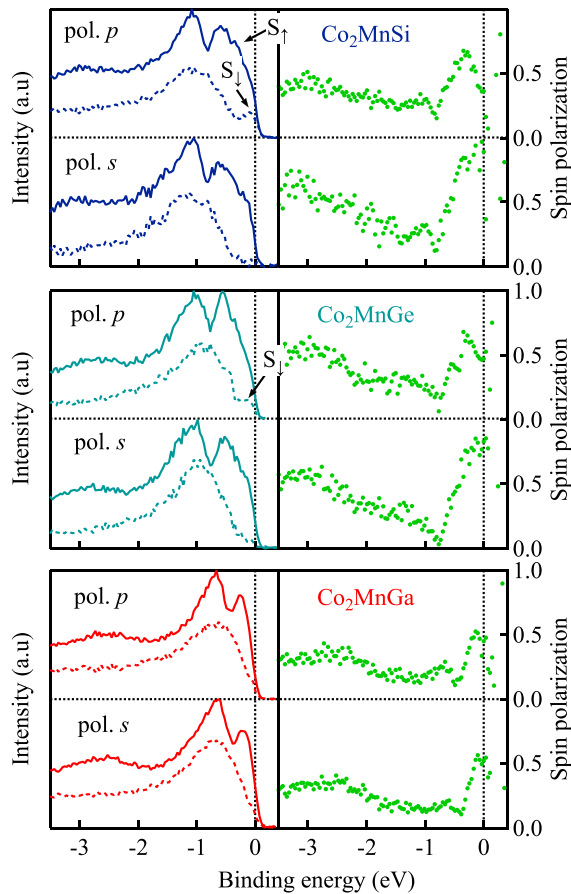


FIG. 2. Spin-resolved photoemission spectra recorded with a 37 eV photon energy using either P or S light polarization on the following Heusler polycrystalline films—top, Co_2MnSi ; middle, Co_2MnGe ; and bottom, Co_2MnGa . The transitions S_1 and S_2 noted [traces of (001) surface states] are not probed using S polarization, revealing the spin gap.

minority spin surface state thus contributed to the decrease in the spin polarization at E_F . To extract the “bulk” spin polarization, one can use the S polarization of the incident photon which kills the surface state transitions due to symmetry reasons.^{14,15} The SR-PES spectra are thus shown in Fig. 2 using both P and S photon polarizations.

Like in epitaxial films, the spin gap is evidenced up to E_F using the S polarized light. The spin polarizations are close to 100% in poly- Co_2MnSi , 90% in poly- Co_2MnGe , and 55% in poly- Co_2MnGa . So, the spin gap previously observed in epitaxial films¹⁵ is still observed in polycrystalline films at least in poly- Co_2MnSi and poly- Co_2MnGe . It should be noted that full spin polarization is now observed for the whole Brillouin zone. This definitely validates our previous results on epitaxial films^{14,15} and the HMM theoretical description of these two compounds. It also demonstrates that grain size and boundary induced changes of the electronic structure are not large enough to close the spin gap of these compounds.

In contrast, the situation is quite different for the third compound studied in the present work, poly- Co_2MnGa . Whereas a full spin polarization is observed in epitaxial Co_2MnGa films,¹⁵ this is not

more the case here for a polycrystalline film. Two hypotheses are proposed to explain this result. First, in contrast to epitaxial Co_2MnSi and Co_2MnGe -films where the upper states of the valence band are far from E_F , these states are almost situated at E_F in epitaxial $\text{Co}_2\text{MnGa}(001)$. Thus, even small spin dependent changes of the electronic structure caused by the finite size of the grains and by the structural disorder at grain boundaries might now be sufficient to get some electronic states around E_F in the minority spin band structure. Another possibility is that there is some minority spin density of state in the 20% part of the BZ not explored in epitaxial Co_2MnGa films, which contributes when exploring poly- Co_2MnGa films. In this case, the spin polarization should be lower for polycrystalline films compared to epitaxial ones. We will show in the following that differences in spin polarization are well correlated with the magnetic damping coefficient values measured on the three films.

The magnetization dynamics were measured by using a vector network analyzer FMR setup used in the perpendicular geometry (see Ref. 15 for experimental details). A static magnetic field, strong enough to saturate the magnetization along the field direction, is applied out of the film plane. This configuration ensures that there is no extrinsic broadening of the linewidth due to the 2-magnon scattering^{21,22} but maximizes the sensitivity of the inhomogeneous linewidth to a random variation of the effective moment.²³ Typical FMR spectra for the three polycrystalline compounds, obtained by fixing the strength of the static field to 1.8 T and varying the frequency of the driving rf field, are shown in Fig. 3(a). Like their epitaxial counterparts, the poly- Co_2MnSi and poly- Co_2MnGe films present very well defined resonance peaks that can be easily fitted. Their linewidths are small, suggesting a strong magnetic homogeneity even in a polycrystalline form. The distorted peak and large linewidth observed for the poly- Co_2MnGa film are again similar to the behavior in the epitaxial case¹⁵ but are now more pronounced. The magnetic damping coefficient α , the effective magnetic moment M_{eff} , and the inhomogeneous

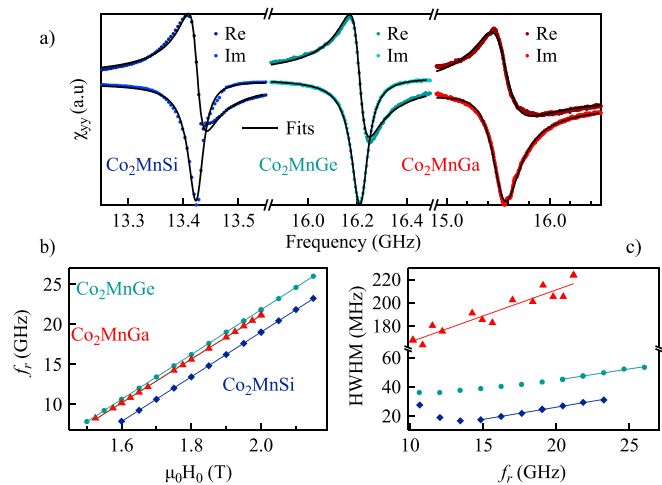


FIG. 3. Results of FMR measurements obtained on the three 40 nm thick polycrystalline films: (a) dynamic susceptibilities vs the frequency of the RF field in the presence of a static, perpendicular magnetic field of 1.8 T. (b) Variation of the resonance frequency f_r with the static applied field used to determine the gyromagnetic ratio g and effective magnetization M_{eff} . (c) Variation of the resonant peak HWHM with f_r used to extract the magnetic damping and inhomogeneous linewidth Δf_c .

linewidth Δf_0 were extracted from fits of numerous spectra (measured for different strengths of the static field) to Kittel's law.¹⁵ Measured variations of the resonance frequency f_r with the applied field and of the linewidth (HWHM) with f_r are shown in comparison to the best fit results in Figs. 3(b) and 3(c), respectively. The magnetic parameters extracted from the fits are summarized in Table I.

The values of magnetic moment obtained for poly-Co₂MnSi and poly-Co₂MnGe are almost the same as in the epitaxial case¹⁵ and in agreement with the Slater-Pauling behavior expected for these Heusler compounds.²⁴ This is a strong indication that Co-Mn antisite disordering,²⁵ if present, is very limited (such disorder strongly affects the magnetic moment which deviates from the Slater Pauling behavior). The effective damping values are 1.15×10^{-3} and 1.65×10^{-3} in the 20 and 40 nm thick poly-Co₂MnSi films, respectively, and 1.5×10^{-3} in the 40 nm thick poly-Co₂MnGe film. They are larger than in single-crystalline films but are still very low and much smaller than in other polycrystalline alloys.²⁶ Moreover, they are of the same order than the best values obtained in single-crystalline films such as epitaxial FeV_x alloys^{27,28} or very recently in epitaxial Fe_{0.75}Co_{0.25}.²⁹ One should also note that our results on poly-Co₂MnGe are very close to the one in Ref. 17 where (001) textured films were grown. Since the measured spin polarizations of 100% for poly-Co₂MnSi and 90% for poly-Co₂MnGe are identical/close to the ones for epitaxial films, the observed damping enhancement must be related to reasons other than a loss of spin polarization. One could assign the higher damping in polyfilms compared to epitaxial ones to the coexistence of B2 and L2₁ observed here. However, this is not so clear since *ab initio* calculation considering such chemical disorder (B2 against L2₁)⁶ did not show a strong impact on the magnetic damping. Finally, the extremely small inhomogeneous linewidth Δf_0 measured for poly-Co₂MnSi and poly-Co₂MnGe is proof of the excellent homogeneity of the magnetic properties in our films, together with the very good control of the stoichiometry.

For poly-Co₂MnGa, the values of the damping coefficient and of its difference to the epitaxial case are 4.5×10^{-3} and 2.5×10^{-3} , respectively. As expected from the severe loss of its spin polarization, both values are much larger than the ones observed for poly-Co₂MnSi and poly-Co₂MnGe. Moreover, they follow the same trend as the epitaxial case. Indeed, even for the almost 100% spin polarization observed in epitaxial Co₂MnGa, its magnetic damping was the strongest (2.0×10^{-3}) among the Co₂Mn-based Heusler series studied in Ref. 15. It was explained by the fact that E_F is too close to the valence band, but the present study highlights another additional possibility: there may be some DOS probed here in the polyfilm in the 20% BZ

not probed in Co₂MnGa epitaxial films. A third explanation relies on the fact that Co₂MnGa has the highest magnetocrystalline anisotropy along Co₂Mn-based Heusler compounds. This implies larger spin-orbit coupling that may affect the magnetic damping.³⁰ Magnetocrystalline anisotropy should affect not only damping but also the effective magnetic moment and the inhomogeneous linewidth. The values of the effective magnetic moment should be smaller, and the inhomogeneous linewidth should be larger for polycrystalline films as for epitaxial films, and both differences should scale with the strength of the magnetocrystalline anisotropy.²³ Indeed, a comparison of the values of the effective magnetic moment and of the inhomogeneous linewidth measured on polycrystalline- and epitaxial-films¹⁵ shows almost no differences for the weakly anisotropic Co₂MnSi- and Co₂MnGe-compounds but a noticeably reduced moment and enhanced linewidth for poly-Co₂MnGa.

To summarize, the HMM behavior demonstrated in some epitaxial Heusler thin films is still observed in polycrystalline films grown under the same conditions, that is, taking great care on the stoichiometry. The spin gap is still observed at least in poly-Co₂MnSi and poly-Co₂MnGe. This is a robust proof of the HMM behavior in these compounds since in such polycrystalline films, the whole Brillouin zone is probed by photoemission experiments. Their magnetic damping coefficients are found to be larger than in epitaxial films, but there are still very low and close to 1×10^{-3} at least for poly-Co₂MnSi and poly-Co₂MnGe. In line with the trend in both the epitaxial case and the observed reduction of its spin polarization, the damping coefficient is the largest in poly-Co₂MnGa. Finally, such small magnetic damping values are of the same order as those measured on the best metallic magnetic films obtained until now but in epitaxial films. Here, no effort is needed to get epitaxy, so one can use any deposition technology and in particular sputtering which is the favorite deposition technique in the spintronic industry. The only key point is to control the stoichiometry. The ease of fabrication in combination with their excellent magnetic dynamics properties makes Heusler-based polycrystalline films very promising for industrial applications in spintronics.

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TABLE I. Magnetic parameters on the three polycrystalline films extracted from FMR measurements (see Fig. 3). The moments in units of μ_B per formula unit (f.u.) are calculated using the lattice constants obtained by x-ray diffraction on those polycrystalline samples ($a = 0.572$ nm for poly-Co₂MnGe and poly-Co₂MnGa and $a = 0.563$ nm for poly-Co₂MnSi).

Compounds	Thickness (nm)	M_{eff} ($\mu_B/\text{f.u.}$)	g (± 0.02)	α ($\times 10^{-3}$)	Δf_0 (MHz)
Poly-Co ₂ MnSi	20	5.09	2.00	1.15 ± 0.03	47
	40	5.03	1.99	1.65 ± 0.10	6.9
Poly-Co ₂ MnGe	40	4.89	2.00	1.5 ± 0.10	13.1
Poly-Co ₂ MnGa	40	4.94	1.95	4.5 ± 1.0	121

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