

Spin polarization of the electric current in half-metallic Co_2MnSi Heusler thin films

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Using propagating spin wave spectroscopy we measure the spin wave Doppler shift in patterned $\text{MgO}/\text{Co}_2\text{MnSi}/\text{MgO}$ thin films and determine the degree of spin polarization of the electric current. Our measurements reveal that the current is fully spin polarized in the devices. This shows that the half-metallic character of the electron band structure translates into a fully spin polarized current flowing across the patterned films. Additionally, we measure a current-induced change of the spin-wave attenuation from which we estimate the nonadiabatic spin-transfer-torque parameter.

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I. INTRODUCTION

A spin-polarized electron band structure is the defining property of any ferromagnet, yet only half-metals are predicted to have an actual 100% spin-polarization due to the presence of a gap at the Fermi level of one of its spin bands [1]. A textbook example of such materials is the Heusler compound Co_2MnSi , which has an experimentally confirmed 0.4–0.8 eV spin gap at the Fermi level [2,3]. The fact that there are only majority electron states in such a wide gap should reduce electron-electron interactions resulting in the record low magnetic damping between metallic ferromagnets down to $\alpha = 4.6\text{--}9 \times 10^{-4}$ [3,4].

These properties make Co_2MnSi promising for applications in spintronics and magnonics [3,5]. Indeed, early measurements of large ratios of both giant and tunneling magnetoresistance in heterostructures [6–8] motivated the further exploration of spin-polarized electron transport: in the ballistic regime, a spin-polarization of the current of 54% was obtained by point contact Andreev reflection [9], and from nonlocal spin valve measurements values of 63% spin polarization have been estimated [10]. Most of these investigations attribute the reduced spin polarization to chemical disorder of the crystal [9,11] or to the spin relaxation through material interfaces to which these methods are bounded. [10].

Recent developments in the growth of Co_2MnSi thin films allowed for the reduction of chemical disorder and controlled stoichiometry [3,11]. Indeed, the predicted spin gap has been confirmed to exist even at room temperature through spin-resolved photoemission spectroscopy near the surface of films [2,3,12]. This suggests that mainly majority electrons should transport the electric current with a 100% spin polarization leading to a relatively simple description of electron transport [4]. However, the room-temperature spin polarization of the diffusive electron current has not yet been measured in these epitaxial films. Investigating this transport regime is important because the relationship between the electronic band structure and spin-polarized transport is not straightforward, owing to electron scattering from thermal excitations and other mechanisms [13]. Moreover, diffusive transport is the regime most relevant to practical device applications.

For this purpose, one can resort to the spin wave Doppler shift technique [14] allowing to measure the spin polarization of the bulk electric current in the diffusive regime without the need for current injection through material interfaces, reference materials or restricted temperature regimes. This technique has been used to successfully characterize the diffusive spin transport in thin films of permalloy, Fe and $(\text{CoFe})_{1-x}\text{Ge}_x$ alloys [13–18].

In this work, we apply an electric current in $\text{MgO}/\text{Co}_2\text{MnSi}/\text{MgO}$ thin films where spin waves propagate and measure directly the spin wave Doppler frequency shifts from which we determine the degree of spin-polarization of the electron current. We show that in agreement with the half-metallicity of the films, the diffusive electric current is indeed 100% spin polarized at room temperature, even after patterning. Our technique also determines the current-induced attenuation of the spin waves from which we estimate the nonadiabatic component of the spin-transfer-torque process.

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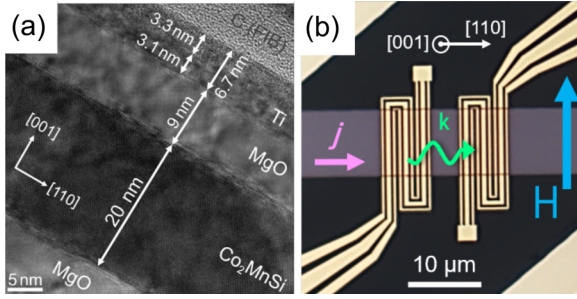


FIG. 1. (a) High-resolution transmission electron microscopy micrograph of the deposited film stack. (b) Microscope picture of a spin wave Doppler shift device. $\mathbf{j}$ is the applied DC current, $\mathbf{k}$ is the wave vector, $\mathbf{H}$ the applied magnetic field.

II. FILMS AND DEVICES

The films are grown on commercial MgO substrates by molecular beam epitaxy following a process similar to that detailed elsewhere [2,3,19] (see also Supplemental Material [20]). The samples for this study consist of the following stack MgO(001)/MgO(10nm)/Co₂MnSi(20nm)/MgO(20nm)/MgO(8nm)/Ti(4nm). The films were characterized *in situ* by reflection high-energy electron diffraction and *ex situ* by X-ray diffraction and transmission electron microscopy (TEM) showing good epitaxy and crystallinity comparable to the ones reported before Ref. [3]. In Fig. 1(a) we present a high-resolution TEM micrograph of the stack obtained from a lamella cut by focused ion beam. We also confirmed that the chemical order predominantly corresponds to the perfectly ordered structure L2₁ [11,20–22]. Additionally, we measured the electric and magnetic properties of the films (at 300 K) before and after the subsequent nanofabrication without noticing any significant change: same resistivity ($\rho = 42.5 \mu\Omega \text{ cm}$) [23], ferromagnetic resonance frequencies, magnetic damping ($\alpha = 1.2 \times 10^{-3}$) and saturation magnetization ($\mu_0 M_s = 1.33 \text{ T}$) (see details in the Supplemental Material [20] and Refs. [24,25] therein).

We patterned the films into strips (along Co₂MnSi's [110] direction) for the current conduction and for the propagation of spin waves. DC contacts are patterned to inject the electric current [20]. The devices are covered with an 85-nm insulating layer of sputtered SiO₂. Finally, a pair of antennas [Ti(5 nm)/Au(140 nm)] are patterned on top of the strip. These are used to excite and measure inductively coherent propagating spin waves [26] [Fig. 1(b)].

III. EXPERIMENT AND RESULTS

The antennas are connected via two RF probes to a vector network analyzer that provides a microwave current to measure the two-port scattering matrix between them. The propagating spin wave spectroscopy experiments are all performed at room temperature. From this we extract the mutual inductance L_{ij} ($i, j = 1, 2$) between the two antennas. For our measurements, the external magnetic field H is applied in-plane perpendicularly to the strip, parallel or antiparallel to the [1–10] crystal direction of Co₂MnSi, (Damon-Eshbach geometry, Fig. 1) while we sweep the microwave frequency.

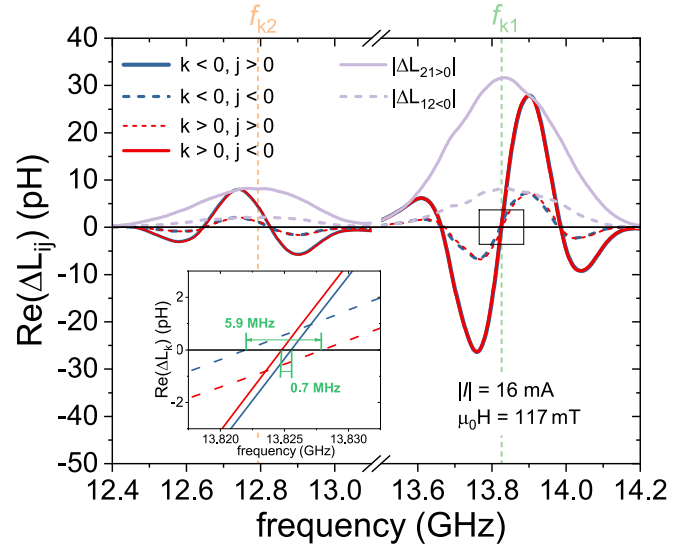


FIG. 2. Mutual inductance as function of frequency for spin waves with $k_1 = 3.90 \text{ rad}/\mu\text{m}$ and $k_2 = 1.86 \text{ rad}/\mu\text{m}$ at an applied current $|I| = 16 \text{ mA}$ and magnetic field $\mu_0 H = 117 \text{ mT}$. The inset shows a closer look of the current-induced frequency shifts at f_{k_1} .

When the resonance condition is matched, the excitation magnetic field produced by one antenna can efficiently excite coherent spin waves that propagate until the region below the second antenna where their stray field is picked up inductively. This is measured as a change in the mutual inductances ΔL_{ij} (Fig. 2). The geometry of the antennas is optimized for the excitation of spin waves with wave vectors $k_1 = 3.90 \text{ rad}/\mu\text{m}$ and $k_2 = 1.86 \text{ rad}/\mu\text{m}$ leading to the observation of two main resonance frequencies f_{k_1} and f_{k_2} , respectively (see details in Ref. [20]).

Now, when we inject a DC electric current $\mathbf{j}$ into the Co₂MnSi strip, the carriers also transport a spin current due to the ferromagnetic nature of the material. The relative motion between this itinerant spin (or magnetic moment) with respect to the propagating spin waves produces the so called spin-transfer-torque (STT). This torque modifies the linearized equation of motion of the magnetization [27]:

$$i\omega \mathbf{m} = \gamma \mu_0 (\mathbf{H}_{\text{eq}} \times \mathbf{m} - \mathbf{M}_s \times \mathbf{h}) + i \frac{\alpha}{M_s} \omega \mathbf{M}_s \times \mathbf{m} + i \delta \omega_{\text{Dop}} \mathbf{m} - i \frac{\beta}{M_s} \delta \omega_{\text{Dop}} \mathbf{M}_s \times \mathbf{m}. \quad (1)$$

Here, ω is the angular frequency, γ is the gyromagnetic ratio and μ_0 is the permeability of vacuum. $\mathbf{M}_{\text{eq}}$ ($\mathbf{H}_{\text{eq}}$) and $\mathbf{m}$ ($\mathbf{h}$) are the static and dynamic components of the magnetization (effective magnetic field), respectively, α is the Gilbert damping parameter representing magnetic losses. The last two terms in Eq. (1) correspond to the adiabatic and nonadiabatic parts of the STT, respectively. The first term proportional to $\mathbf{m}$ represents the current-induced shift of the precession frequency; also known as the spin-wave Doppler shift [14]

$$\delta \omega_{\text{Dop}} = - \frac{\mu_B}{e M_s} P \mathbf{j} \cdot \mathbf{k} = \mathbf{u} \cdot \mathbf{k}. \quad (2)$$

Here, μ_B is the Bohr magneton, e is the electron charge, $\mathbf{u} = - \frac{\mu_B}{e M_s} P \mathbf{j}$ is the effective magnetization velocity and P is the

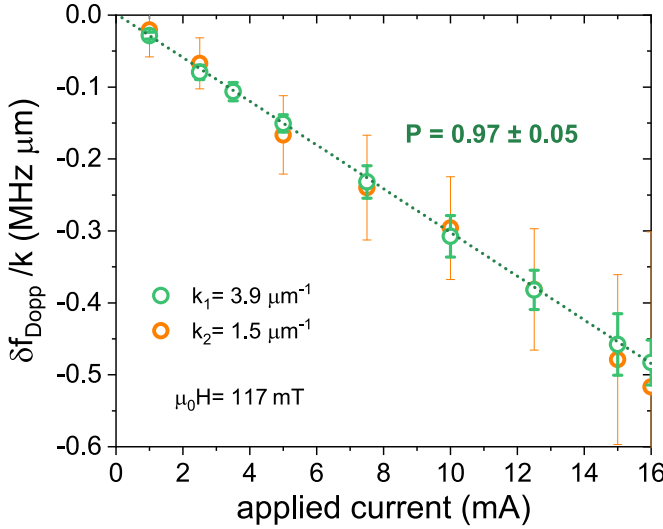


FIG. 3. Extracted spin wave Doppler shift as function of the applied electric current for wave vectors k_1 and k_2 at applied magnetic field $\mu_0 H = 117$ mT for device WA22 in Table I in Ref. [20]. The dotted line is the linear fit of data corresponding to k_1 to Eq. (2) from which we obtain an exemplary $P = 0.97 \pm 0.05$.

degree of spin polarization of the electron current. Under the two current model this is defined as $P = \frac{\rho_\downarrow - \rho_\uparrow}{\rho_\uparrow + \rho_\downarrow}$ [16,28], where $\rho_\uparrow$ is the resistivity of the majority electrons and $\rho_\downarrow$ that of the minority electrons.

The last term in Eq. (1) accounts for the current-induced (electric field-driven) modification to the magnetic losses [29–31]. β is the so-called nonadiabatic STT parameter and quantifies the magnitude of such modifications. Given the formal similarity of this term with the Gilbert one, it can be considered as a current-induced component of damping [32].

A. Current-induced spin-wave Doppler shift and spin-polarization

To measure the spin-wave Doppler shift we can follow the current-induced modification of the phase of ΔL_{ij} . Indeed, in Fig. 2 we see directly how the spectra shift in opposite directions for the two current polarities. The shift's magnitude is different for the two propagation directions since, on top of the Doppler shift, there is an additional frequency shift due to the current-induced Oersted field. These two can be separated by a symmetry analysis [20,33]. Note that there is a clear amplitude nonreciprocity between the spectra of the two propagation directions. This is expected in the Damon-Eshbach geometry and does not affect the frequency shifts [34].

In Fig. 3 we plot the extracted Doppler shift as a function of the applied electric current for the spin waves with the two experimentally accessible wave vectors k_1 and k_2 for one of the fabricated devices [20]. We observe a linear dependence with very similar slope for both wave vectors ($\pm 5\%$). The main difference between the two data sets comes down to the error bars. This difference originates from the lower signal-to-noise ratio for the k_2 peak compared to the k_1 one. By fitting these data sets to Eq. (2) we obtain the slope and having measured

$\mu_0 M_s = 1.33 \pm 0.02$ T [20], we determine an average value for the spin polarization of $P = 0.99 \pm 0.08$. This value is obtained as the average between the values of P obtained for the two measurable wave vectors, different magnetic fields and five different devices [20]. The relatively small uncertainty speaks of the reproducibility of our measurements.

In result, we measure a 100% degree of spin-polarization confirming that the diffusive electric current has also a half-metallic character in patterned Co_2MnSi at room temperature. Note that this is not a trivial result as previously we only had information about the film's surfaces density of states near the Fermi level [2–4].

Indeed, $\text{Co}_2\text{MnSi}/\text{MgO}/\text{Co}_2\text{MnSi}$ magnetic tunnel junctions measurements have shown that minority states contribute to the tunneling current [35,36]. Up to this point it was not clear if such interface states could contribute to the bulk diffusive transport of Co_2MnSi [37]. Notably, our result shows that the electric current has a half-metallic character which means that any possible surface or bulk minority electronic state at the Fermi level does not contribute significantly to the diffusive electron transport.

We stress that our result does not imply that the density of states is fully spin polarized. *Ab initio* work has shown how thermal spin-fluctuations can lead to shifts in the density of states and to a slight reduction of its spin-polarization in Co_2MnSi [38]. This is not in disagreement with our result, since the properties of the density of states do not translate directly to the electric current [39]. The current and its spin polarization (the quantities that are relevant for most spintronic devices) are also determined by how the electron bands shift, diffuse/broaden with temperature, and how they couple to other degrees of freedom like phonons and magnons [13,40].

To conclude this part, we remark that we measure systematically a fully spin polarized electron current in several patterned devices [20]. This is a true testament of the robustness of the half-metallic character of these films against nanofabrication.

B. Current-induced spin-wave amplitude change

Now that we have seen how the electric current shifts the resonance frequency of the spin waves, we turn to investigate how it modifies their attenuation.

From Eq. (1) we know that the nonadiabatic part of the STT translates into a current-induced modification of spin-wave losses. To estimate such change, we follow precisely the current-induced change of the amplitude of the propagating spin-wave signals ΔL_{ij} . In the linear regime we can describe this change with

$$\Delta L_{ij}(f, I) = (1 \pm \varepsilon_{ij}(I))g_{ij}(f + \delta\omega_{Dop}), \quad (3)$$

where $g(f)$ is a wave-form function (in practice we use a complex Gaussian function) and the $\varepsilon_{ij}(I)$ parametrize the current-induced amplitude change of the signal [20]. For our measurements, the factor $1 + \varepsilon_{ij} = e^{-T_2/t_p}$ (T_2 is the characteristic relaxation time given by the Gilbert damping [20,41]) accounts for this change during the apparent time delay $t_p = 1/f_p$ that it takes spin waves to travel between the two antennas (f_p being the period of the oscillation in Fig. 2, see also,

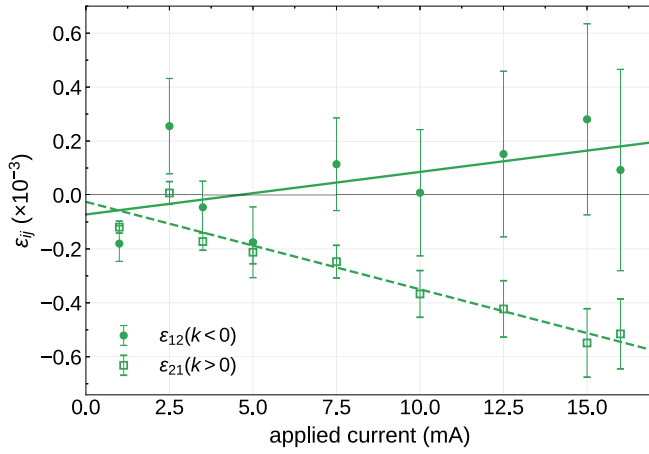


FIG. 4. Current-induced modification to the mutual inductance amplitude as function of the applied current for a wave vector $k_1 = 3.90 \text{ rad}/\mu\text{m}$ and applied magnetic field $\mu_0 H = 117 \text{ mT}$ for device WA22 in Table I in Ref. [20]. The lines are linear fits.

Ref. [20]). We can extract the ε_{ij} from our measured spectra [20] and plot them as functions of the applied current for the two propagation directions. In Fig. 4 we present an example of this data showing a linear behavior for both propagation directions. Despite the larger data dispersion for $k < 0$ due to the smaller signal-to-noise ratio, the overall trend of the ε_{ij} changes sign depending on the relative direction of spin-wave propagation and applied current. This indicates that the current-induced attenuation can either increase or decrease the net dissipation, in agreement with the expected effect of nonadiabatic STT [see Eqs. (1) and (2)]. Similar to the first part of the analysis, we use the two propagation directions and two current polarities to eliminate spurious contributions that do not follow the nonadiabatic STT symmetry. By fitting the data of several devices to the corresponding linear expressions [20], and extracting the slopes, we obtain an average current-induced modification to the damping parameter $\alpha - \beta = -0.04 \pm 0.01$. This is obtained as the weighted average between the values of $\alpha - \beta$ estimated for different devices and fields [20].

To further understand this observation, we measured the temperature dependence of the magnetic damping in a piece of material that underwent nanofabrication. Our ferromagnetic resonance measurements show that α follows a conductivity-like behavior as a function of the temperature [20]. According to the Kambersky's torque model [42], this means that α is dominated by intraband electron transitions, i.e. magnetization precession leads to fluctuations of the electron states occupation around the Fermi level (breathing Fermi surface model) [31]. In the present case of our 100% spin polarized Co_2MnSi , only majority electrons should contribute.

Now, if we consider our estimated $\alpha = 1.2 \pm 0.1 \times 10^{-3}$ [4,20], we obtain $\beta = 0.04 \pm 0.01$ at room temperature [43]. This means that although the magnetic damping in our material is the smallest of all metallic ferromagnets [2,4], the nonadiabatic STT remains relatively large and comparable to others (e.g., permalloy) [17,44].

This result becomes more surprising if one considers the historical development of STT around domain-wall motion

[30] and measurements in $\text{Ni}_{80}\text{Fe}_{20}$ where the estimated ratio β/α is close to unity [17,27]. Actually, under the Kambersky torque model, both α and β originate from the same electron transitions [31]. Yet, they differ in that each electron state contributes to β with a weight v_n/u proportional to its respective band velocity v_n [n is the electron band index and u is the magnetization velocity, see Eq. (2)] [31]. In a hypothetical fully Galilean invariant system (all electrons have the same velocity and $v_n/u = 1$) one would expect $\beta/\alpha = 1$ strictly. However, in real materials, while for α all electronic transitions contribute with the same weight, for β , electron states with large velocities would have the main contributions (i.e., $\beta/\alpha \neq 1$). Therefore, we argue that the ratio β/α could be interpreted as a probe of the type of electron states involved in both dissipative processes.

Indeed, the proven half-metallicity of the transport in our films (reduced type and number of electron transitions allowed) and the small value of α allows us to interpret the estimated ratio $\beta/\alpha \approx 40$ closely to Kambersky's model: in Co_2MnSi , the large β is an experimental signature that fast majority electron states around the Fermi level (highly conductive electrons) do in fact contribute to the magnetic dissipation.

IV. CONCLUSIONS

We have had a closer look at the nature of the electronic states involved in the spin transfer torque processes between an electron current and propagating spin waves in the Heusler compound Co_2MnSi .

Our study of the current-induced spin-wave frequency shifts reveals that the diffusive electric current is fully spin polarized at room temperature, i.e., it contains only majority electron states, in agreement with the predicted half-metallic character of the material. Remarkably, this occurs in patterned devices of thin films at room temperature, which entails a promising realistic functionality for spintronic applications.

Additionally, we measured the current-induced change of the mutual inductance amplitude. From this we estimated a relatively large nonadiabatic spin transfer torque parameter which is attributed to the contribution of highly conductive majority states to intra-electron-band magnetic dissipation processes. We think these experimental observables can assist in the study of electronic transitions and their contributions to magnetic losses particularly in half metals and highly spin-polarized-low-damping metallic ferromagnets.

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DATA AVAILABILITY

All data related to the article is available upon reasonable request.

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