

Low-Energy Spin Precession in the Molecular Field of a Magnetic Thin Film

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Electronic spin precession and filtering are measured in the molecular field of magnetic thin films. Lab-on-chip experiments allow injection of electrons with energies between 0.8 and 1.1 eV, an energy range not yet explored in spin precession experiments. While filtering angles agree with previous reported values measured at much higher electron energies, spin precession angles of 2.5° in CoFe and 0.7° in Co per nanometer film thickness could be measured which are 30 times smaller than those previously measured at 7 eV. On the basis of ab initio calculations, the results are explained and it is shown that the band structure and layer roughness are playing a key role at low energy.

1. Introduction


Since the discovery of Giant Magneto-Resistance (GMR) in 1988^[1,2] the spintronics research field has become the ground of intense investigations. Accompanying the growth of fundamental knowledge on spin transport in solid-state devices, numerous proposals for applications have emerged (see for instance refs. ^[3] and ^[4]). Among those, some have already hit the market as hard drive read heads, magnetic fields sensors, and spin transfer torque based magnetic random access memories. Despite the apparent maturity of the field, fundamental points

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remain to be clarified.^[5] In particular, very little is known about the electronic spin behavior in its out-of-equilibrium state (i.e., beyond the Fermi sea) even if it is acknowledged to be a source of spin-transfer torque.^[6]

When a beam of electrons with an initial spin polarization vector P_0 is injected into a region of space where a magnetic field H is present, the polarization vector P will exhibit a precessional motion around the magnetic field. Two angles can then be defined: the filtering angle, θ , that describes the reorientation of P

toward H and the precession angle, ϵ , that describes the precession of P around H (**Figure 1** in which the blue arrow is the field H). The precession frequency is given by the Larmor frequency $\omega_L = \gamma H$ with $\gamma \approx 1,7 \cdot 10^{11} \text{ rad s}^{-1} \text{ T}^{-1}$ the gyromagnetic ratio. If the incident polarized electrons are considered to move at a typical speed of $2 \cdot 10^6 \text{ ms}^{-1}$ (close to the Fermi velocity of many metallic elements), a precession angle per μm and per Tesla, $\tilde{\epsilon}$, of about $0,17 \text{ rad T}^{-1} \mu\text{m}^{-1}$ is expected. Large precession angles can thus be achieved either by a short travel distance in a strong magnetic field or by a long travel distance in a small magnetic field. This latter scheme was used in metals by Jedema et al.,^[7] as well as in semiconductors by Appelbaum et al.,^[8] and Awschalom et al.,^[9] while the first strategy was employed by Oberli et al.^[10] in their free-electron beam experiments. By injecting a spin polarized electron beam into a magnetic layer, the so-called molecular field of a ferromagnetic layer is estimated to be of the order of several 100 to 1000 T. Oberli et al. achieved experimentally precession angles of several tens of degrees per nanometer. Unfortunately, measurements with a free electron beam at electron energies (with respect to the Fermi level) below the vacuum level (4–5 eV for ferromagnetic metals, such as Co and Fe) are not possible. However, this is exactly the energy range of interest for all spintronics applications (typical bias voltages applied to tunnel junctions are 1 to 2 V). Therefore, we have conducted lab-on-chip experiments allowing us to measure at these low energies the spin precession induced by the molecular field of thin ferromagnetic layers.

2. Results and Discussion

Three elements are essential to perform such experiments: a spin polarizing layer, an active precession layer, and an analyzing layer. In order to study the dependence of the precession angle as a function of electron energy, the lab-on-chip has to host an

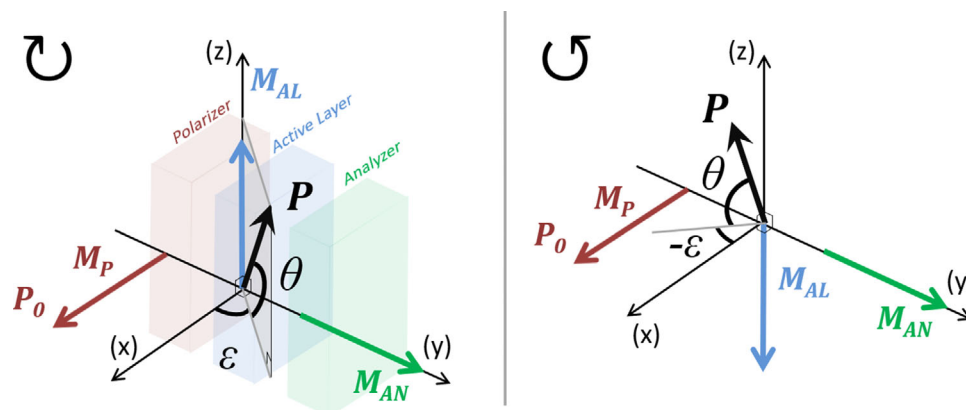


Figure 1. Angles of precession and filtering of the spin polarization vector of an electron beam injected into a magnetic layer with magnetization M_{AL} or a field H oriented along M_{AL} . See text for further details.

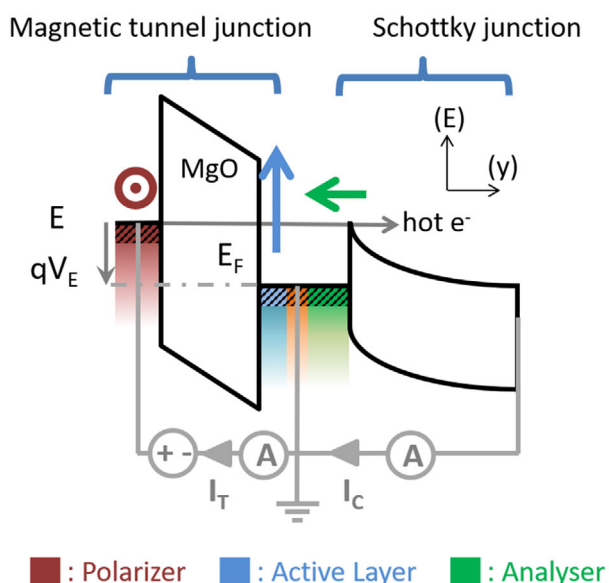


Figure 2. All solid state device based on a magnetic tunnel transistor that allows measuring the spin precession at low energies in the molecular field of a thin magnetic layer. The dotted red circle represents the arrow of the magnetization of the polarizer layer pointing perpendicular to the paper sheet. The blue arrow represents the magnetization of the active layer and the green arrow represents the magnetization of the analyzer layer. See text for further details or Figure 1 for the magnetic configuration.

electronic device that allows varying the injection energy of the electrons. In this work the electron injection is accomplished by a magnetic tunnel junction (Figure 2). After having aligned the spin polarization of the injected electrons within the polarizing layer along its magnetization direction, the electrons are transported via the tunnel effect through the MgO barrier. As the tunnel transport is spin conservative, the spin polarization of the electrons arriving in the active layer is perpendicular to the active's layer magnetization direction and will consequently precess around it during the electron's propagation. Changing the bias voltage across the tunnel barrier varies the injection energy in the active layer and thus allows a spectroscopic analysis

of the precession angle. This angle is analyzed through the GMR effect occurring in the active layer / Cu / analyzer spin valve (blue/orange/green rectangles in Figure 2). Note that the analyzer magnetization is orthogonal to the magnetization of both the active and the polarizing layers. Finally, one last key ingredient to the precession angle analysis is a Schottky diode (Figure 2). It allows a dual analysis. First of all, it ensures that the collected electrons in the semiconductor have always energy higher than 0.7 eV (the height of the Schottky barrier) after having passed the spin valve. Thus, only the spin precession of hot electrons is analyzed, while all thermalized electrons are reinjected by the tunnel barrier through the spinvalve's electron recovery circuit. Second, it defines an acceptance cone with an opening angle θ_c of only 4.5° at $E = 1$ eV at the Schottky interface due to the conservation of the momentum parallel to the Cu/Si interface (see additional material). This angular wave vector filtering effect is reinforced by another angular filtering taking place during the tunneling process. Thus, for an injection energy of around 0.7 eV, the collected electrons in silicon have been transported across the spin valve in an almost ballistic manner and practically perpendicular with respect to the multilayer interfaces.

A typical stack used in this study is as follows: Pt(5)/IrMn(7.5)/Co(2)/Ta(0.5)/CoFeB(2)/MgO(2.5)/X(y)/Cu(3.5)/[Ni(0.6)/Co(0.2)] \times 5/Ni(0.6)/Cu(5)/Ta(1)/Cu(5)//Si(100), where numbers in brackets indicate the layer thicknesses in nm. The multilayer is grown by sputtering on a hydrofluoric acid cleaned Si substrate (see method section for supplementary details). The CoFeB layer is the polarizer and X is the active layer. The [Ni(0.6)/Co(0.2)] \times 5/Ni(0.6) multilayer represents the analyzer. To determine a precession angle in our lab-on-chip experiment it is required to stabilize the aforementioned 3D magnetic configuration. The way to obtain a crossed configuration of spin valve magnetizations has been reported in a previous study.^[11] The crossed configuration in the Pt/IrMn/Co/Ta/CoFeB/MgO/X(y) tunnel junction is obtained by establishing an exchange bias field at the IrMn/Co interface to set the magnetization easy axis of the polarizer along the x-direction. The multilayer deposition is followed by an annealing process under an applied field to initiate the exchange bias field and by four steps of optical lithography to define the electrical contacts on the different layers of interest (see method section for supplementary details). The

annealing also promotes the crystallization of the CoFeB/MgO interface through diffusion of B that helps to increase the tunnel magnetoresistance of the tunnel junction. Precession was studied in two ferromagnetic layers, Co and CoFe (B has diffused out of the CoFeB layer after thermal anneal). These two materials have been chosen for the good quality of the tunnel barriers that can be achieved when the MgO is deposited on top. Co and CoFe are also useful for comparison with previous reports measured at higher energy.^[10] The thicknesses of the active layer were varied between 1 and 10 nm (1, 3, and 4 nm for CoFeB, 3 and 10 nm for Co).

The collected hot electron current I_C can be expressed as $I_C = I_C^\perp (1 + MC^\perp P \cdot M_{An})$ where P is the hot electron spin polarization vector after propagation through the active layer and M_{An} indicates a unit vector pointing along the magnetization direction of the analyzer.^[12] MC^\perp is the magneto-current ratio defined as $\frac{I_C^\parallel - I_C^\perp}{I_C^\perp}$ where I_C^\parallel and I_C^\perp are the collected currents when P and M_{An} are parallel or perpendicular to each other, respectively. Since we plan a spectroscopic analysis, the tunnel junction bias voltage (V_E) will be changed and so will the injected current. It is then convenient to normalize the collected current by the injected one. This defines the transfer ratio as:

$$TR = TR^\perp (1 + MC^\perp P \cdot M_{An})$$

which could also be expressed as $TR^\perp [1 + MC^\perp P_0 \cos(\theta) \sin(\epsilon)]$. The $\cos(\theta) \sin(\epsilon)$ product containing our angles of interest can thus be nicely obtained experimentally by measuring the transfer ratio in three different magnetic configurations: parallel \parallel (in this configuration, the active layer and analyzer are parallel and $P \cdot M_{An} = P_0$), clock wise \curvearrowright and counter clock wise \curvearrowleft as illustrated in Figure 1. The three transfer ratios TR^\parallel , TR^\curvearrowright , and TR^\curvearrowleft , can be expressed as $TR^\parallel = TR^\perp (1 + MC^\perp P_0)$ and $TR^\curvearrowright/\curvearrowleft = TR^\perp [1 \pm MC^\perp P_0 \cos(\theta) \sin(\epsilon)]$. As a result, the $\sin(\epsilon) \cos(\theta)$ product as a function of experimentally available quantities writes as:

$$\sin(\epsilon) \cos(\theta) = \frac{TR^\curvearrowleft - TR^\curvearrowright}{2TR^\parallel - (TR^\curvearrowright + TR^\curvearrowleft)}$$

Measurements of the three aforementioned TR versus V_E performed on the sample having an active layer composed of a 1 nm thick CoFeB film are reported in **Figure 3a**. They provide clear evidence of a precessional effect in the CoFeB layer since TR^\curvearrowright and TR^\curvearrowleft are not superimposed. Precession angles ϵ obtained for different filtering angles θ ranging from 0° to 85° are shown in **Figure 3b**. One must note that the obtained values are always smaller than the ones reported by Weber et al.^[13] and this is the case even if strong filtering effects are considered. Increasing θ toward 90° naturally increases ϵ toward Weber's values but such a high spin filtering effect is not expected for such a thin magnetic layer. In order to determine ϵ , the determination of θ is mandatory.

Quantitative values are obtained by varying the active precession layer thickness and taking into account that electrons overcoming the Schottky barrier have $\vec{k}_{\parallel} = \vec{0}$ (see additional material). In this case, the distance traveled by the electrons equals the thickness d of the active magnetic layer. Since the hot electron current is exponentially decreasing as a function

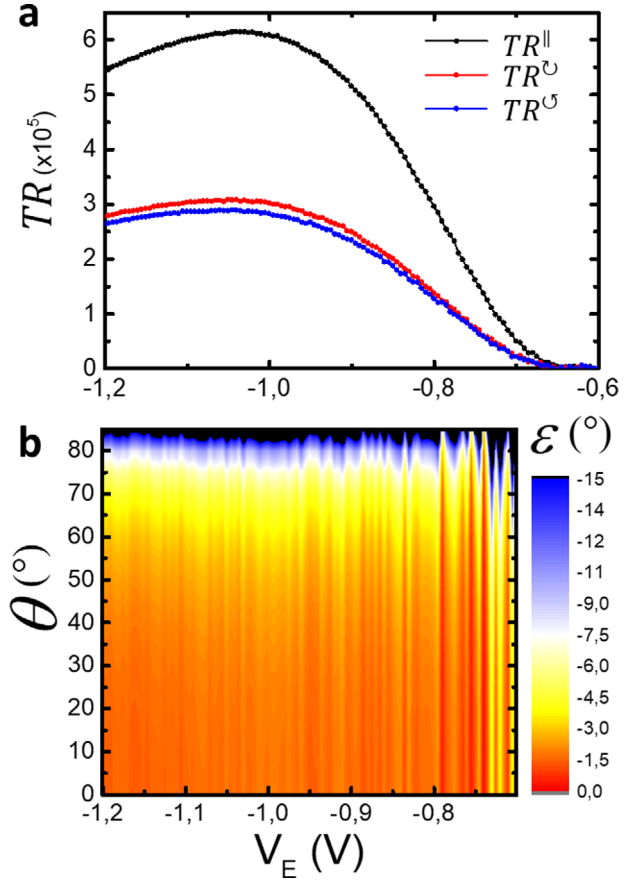


Figure 3. CoFeB (1 nm) precession layer. a) TR^\parallel (black), TR^\curvearrowright (red), and TR^\curvearrowleft (blue) versus applied voltage on the tunnel barrier measured at 60 K. b) Precession angle calculated from TRs as a function of applied voltage on the tunnel barrier considering different filtering angles θ (measured at 60K).

of d (see additional material), the $\sin(\epsilon) \cos(\theta)$ product can be rewritten as:

$$\sin(\epsilon) \cos(\theta) = \sin(\epsilon^* d) \cosh\left(\frac{d}{2\lambda^-}\right)^{-1}$$

where ϵ^* is the precession angle per nanometer and $\frac{1}{\lambda^-} = \frac{1}{\lambda^\uparrow} - \frac{1}{\lambda^\downarrow}$ (λ^\uparrow being the minority/majority inelastic electron mean free paths).

Fits of the $\sin(\epsilon) \cos(\theta)$ product versus d (reported in additional material) allows then the extraction of ϵ^* and λ^- as a function of hot electron energy. As both ϵ^* and λ^- are mostly constant over the energy window studied, their mean values are presented in **Table 1** for comparison to reported values.

First, let's look at λ^- for which values concerning Co can be found in the literature. All of them are in good agreement (our work, Weber et al., Van Dijken et al.),^[14] suggesting only a slight energy dependence of λ^- (see additional material). These values of λ^- indicate that Co thicknesses larger than 10 nm are necessary to almost completely turn the spin polarization vector of the hot electrons into the direction of the magnetization of the active layer. When Fe is inserted in the Co layer λ^- decreases to 0.81 nm

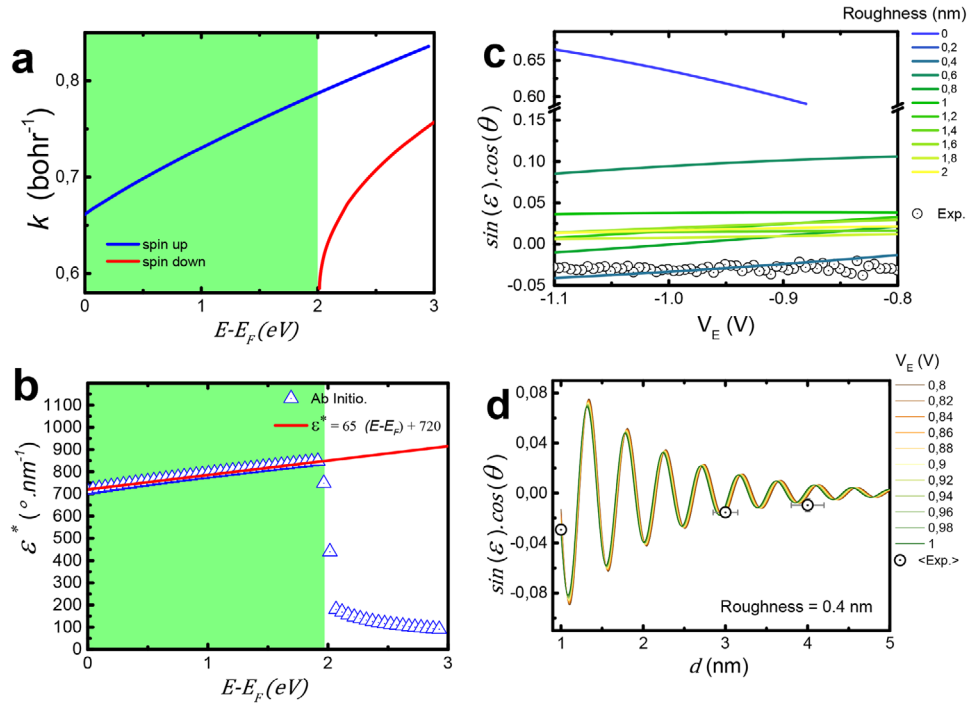


Figure 4. a) Ab initio computation of the band structure along Δ direction in the CoFe(100) case. b) ϵ^* versus $E-E_F$ from ab initio computation in the CoFe case. c) Mean $\sin(\epsilon)\cos(\theta)$ versus V_E . Line for an active layer composed of a 1 nm thick CoFeB active layer and for various variation of travel distance. Points are experimental data. d) Mean $\sin(\epsilon)\cos(\theta)$ versus d for different values of V_E . Points: experimental values of $\sin(\epsilon)\cos(\theta)$ for $d = 1, 3$, and 4 nm.

Table 1. Summary of the experimental values extracted from our work for Co and CoFeB and comparison to previous reports at higher energy.

Active Layer	Results comparison table			
	Present work		W. Weber et al.	
	CoFeB	Co	Co	Fe
Energy (above E_F)	1 eV	1 eV	7 eV	7 eV
ϵ^*	$2.4^\circ \text{ nm}^{-1}$	$0.7^\circ \text{ nm}^{-1}$	19° nm^{-1}	33° nm^{-1}
λ^-	0.56 nm	1.38 nm	1.54 nm	1.49 nm

for $\text{Co}_{84}\text{Fe}_{16}$ in Van Dijken et al. and to 0.56 nm in our work for $\text{Co}_{50}\text{Fe}_{50}$, which is linked to the decrease of λ^\perp . For CoFeB almost full spin filtering occurs for thicknesses of 5 nm. The fact that λ^- does only slightly vary in our energy window has been pointed out theoretically by Nechaev et al.:^[15] λ^- is mainly determined by the small value of λ^\perp that does not change with energy.

The surprise of our study relies on the values of the precession angle. Since no other data are available in this low energy range (E_F+1 eV), we can only compare with the work of Weber et al. which has been performed with the same material (Co) but at a much higher electron energy (E_F+7 eV): the precession angle per nanometer is 30 times smaller at E_F+1 eV than at E_F+7 eV. By changing the active layer material to CoFeB, that is, Co is partly replaced by Fe, we find in our lab-on-chip experiments an increase of the precession angle by a factor of 3.5. This tendency seems

to be followed in the free electron beam experiments at higher electron energy when going from Co to Fe.

When a spin-polarized electron beam is injected into a region of space where a perpendicular magnetization exists, the precession of the spin-polarization vector is theoretically given by $\epsilon^*(E) = \Delta k$ where $\Delta k = k^\uparrow - k^\downarrow$ is the difference in k -vector for both spin bands at energy E (see additional material). When only the spin-up band is accessible as for instance in **Figure 4a** at energies below 0.7 eV we have $\Delta k = k^\uparrow$ (see additional material). As a result, the value of ϵ^* should be strongly dependent on the spin-dependent band structure of the active layer. Ab initio calculations have been performed to get the band structure of CoFe(100) in the Δ direction (Figure 4a) that allows the determination of ϵ^* (Figure 4b). At energies above 1.95 eV, the two spin bands are accessible and increasing the energy leads to a decrease of ϵ^* as observed experimentally by Weber et al. Furthermore, the values are in rough agreement with the experimental report at higher energies. However, for energies below 1.95 eV, only one band can be accessed. In this case, $\epsilon^* = k^\uparrow$ such that huge values of ϵ^* should be measured experimentally. In real samples, however, fluctuations in the hot electron travel distance should be considered. Using the values and the linear variation of ϵ^* from the ab initio calculations and the experimental values of λ^- , we calculated a mean value of $\sin(\epsilon)\cos(\theta)$ considering a travel distance varying from d to $d + \Delta d$ for a sample with $d = 1$ nm and by varying Δd :

$$\sin(\epsilon)\cos(\theta) = \frac{1}{\Delta d} \int_d^{d+\Delta d} \frac{\sin(\epsilon^* \cdot t)}{\cosh\left(\frac{t}{2\lambda^-}\right)} dt.$$

The experimental values of $\sin(\epsilon)\cos(\theta)$ could be reproduced with $\Delta d = 0.4$ nm (Figure 4c). This difference in travel distance cannot be related to specular electron travelling: it would correspond to an angle of 44.4° , which is completely out of the acceptance cone. However, a layer roughness of 0.4 nm is reasonable and can be considered as being constant as a function of layer thickness. The mean values of $\sin(\epsilon)\cos(\theta)$ can then be calculated as a function of d and injection energy with a fixed value of $\Delta d = 0.4$ nm. Oscillations could be calculated that are in agreement with our experimental results as seen in Figure 4d. Furthermore, even if a strong variation of ϵ^* is expected theoretically with energy, the theoretical mean values $\sin(\epsilon)\cos(\theta)$ are almost constant as observed experimentally.

3. Conclusion

In conclusion, we show for the first time the manipulation of the spin direction in an unexplored energies range, thanks to an all solid-state device. As forecasted theoretically, ϵ^* is huge and requires a better control of the active layer roughness to access to high values. This result is the starting point for new studies in which materials, crystallographic orientations, band structure are parameters that can affect precession and pave the way for exploration of far richer spin transport properties than the one at high energies.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Author Contributions

M.H. and D.L. conceived the project. C.V., M.H., and D.L. were in charge of the thin-film growth and optimization of magnetic properties. C.V. patterned the samples. Y. L. designed the lithography masks and optimized

the Cu/Si Schottky barrier. C.V. and D.L. conducted the electronic transport under applied field experiment. C. T., M. C., C.V., W.W., D.L., and M.H. analyzed the data and D.L. and M.H. wrote the manuscript. All authors contributed to the discussion.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

hot electrons, magnetic tunnel transistors, molecular fields, spin precession, spintronics

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