

Spin Filtering of Free Electrons by Magnetic Multilayers: Towards an Efficient Self-Calibrated Spin Polarimeter

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An asymmetrical ferromagnetic cobalt bilayer (18 nm Au/0.8 nm Co/2.2 nm Au/1.3 nm Co/1.5 nm Au) operates as a self-calibrated spin polarimeter with a high spin selectivity for free electrons injected at a few eV above the Fermi level. We present the analysis of transmitted currents as a function of the incident energy, based on a model of spin polarization dilution into the first gold layer and ballistic transport close to the vacuum level throughout the sample.

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Since the Stern and Gerlach experiment, spin filtering has been demonstrated in systems associating ferromagnetic and nonmagnetic materials. One of its most spectacular manifestations is the giant magnetoresistance in ferromagnetic multilayers [1]. It refers to the conductivity at the Fermi level, which is strongly correlated to the orientation of magnetization. In contrast, spin polarized electron spectroscopy offers the opportunity to address spin-dependent transport of ballistic electrons traveling at energies of a few tens of eV above the Fermi level. In order to do so, one can examine the transmission of an unpolarized electron beam from a nonmagnetic substrate through an ultrathin 3D ferromagnet [2,3]. Unpolarized electrons are then found to become polarized after traversing the magnetic layer. This so-called overlayer technique shows the preferential emission of a spin direction from the ferromagnetic layer, defining an electron spin filter effect. The spin polarization can be explained by the electron inelastic scattering involving unoccupied d states above the Fermi level [4,5]. The scattering rate for minority spin electrons is then enhanced with respect to that of majority spin electrons due to the excess of minority spin holes.

The first attempts to characterize electron spin filtering well above the Fermi level were attained by measuring the *direct* transmission of a spin polarized free electron beam through a *freestanding* Au/Co/Au film [6,7]. It was shown that the transmitted current depends on the relative orientation of the incident spin polarization with respect to the cobalt layer magnetization. This approach offers the advantages of tuning the energy and the spin polarization of the incoming electrons, and of separating the inelastic and the elastic contributions to the transmitted current. By analogy to spin polarimetry, an ultrathin ferromagnetic layer can be described by its spin discriminating power, the so-called Sherman function s . It is the polarization acquired by an unpolarized beam when passed through the spin filter [8]. The Au/1 nm Co/Au structure, which has an s value of 0.34, behaves as an electron spin detector [9].

In this Letter, we show that the Au/Co/Au/Co/Au cobalt bilayer constitutes a self-calibrated spin polarimeter.

Two uncoupled ferromagnetic layers of different thickness d_1 and d_2 have different coercive fields. Therefore a bilayer structure can be magnetized in four saturated configurations: two of them, that we call “F” and “-F” correspond to parallel saturated magnetizations in both layers, and the other two labeled “AF” and “-AF” refer to antiparallel saturated magnetizations. The two reversed configurations F (AF) and -F (-AF) are equivalent for the transmission of an unpolarized beam, whereas they yield different transmissions for a polarized beam. It is therefore possible to perform three separated measurements: the transmission asymmetry of an unpolarized beam between F and AF configurations and the transmission asymmetries of a polarized beam between F and -F and between AF and -AF configurations. As a consequence, one obtains separately three quantities: the spin discriminating power s_1 and s_2 of each layer and the polarization P_0 of the incident beam. Furthermore, from the microscopic analysis of s_1 and s_2 in terms of spin-dependent inelastic scattering at the interfaces and in the bulk of the ferromagnetic layers, we determine a characteristic spin-discriminating length in bulk cobalt and the spin-filtering power of Au/Co interfaces. We also show evidence that spin transport above the Fermi level is mainly governed by the creation of a low energy secondary electrons distribution, followed by a ballistic transmission at an energy close to the vacuum level. The formation of the electron distribution leads to a dilution of the polarization by secondary electrons.

The transmission experiment consists of focusing a longitudinally polarized electron beam photoemitted from a GaAs source on a cobalt bilayer with perpendicular magnetization (18 nm Au/0.8 nm Co/2.2 nm Au/1.3 nm Co/1.5 nm Au). The spin polarization P_0 of the incident beam lies along the direction of the sample magnetization. Experimental details on sample preparation can be found in Ref. [9]. The sample is magnetized *in situ* by pulses of the magnetic field, which allows one to select the four distinct states of saturated magnetization. The electron beam enters the thickest gold side. The low energy injection threshold is given by the vacuum level of the entrance side, whereas

the lowest possible energy of the transmitted electron is the vacuum level E_V of the exit side of the cap Au layer. The vacuum level of both gold faces was lowered at best by 3 eV by cesium deposition. The electrons emerging from the multilayer build up an energy distribution curve, which includes, besides the elastic electrons, a distribution of inelastically scattered electrons. A retarding-field analyzer with an energy resolution of 0.2 eV FWHM allows one to separate these two parts. The inelastic contribution is located at the energy E_V and originates from electrons having lost energy and from secondary electrons excited from the Fermi sea. In the following, we will discuss only the inelastic part of transmitted beam, which is in our case essentially responsible for the spin-dependent transport above E_F [9].

The transmitted currents I_P^μ are measured at E_V , where μ stands for the bilayer magnetization configuration and P for the polarization values (0 or $\pm P_0$). We separate I_P^μ into its spin-independent contribution I_0^μ and its spin-dependent contribution ΔI_P^μ , $I_P^\mu = I_0^\mu + \Delta I_P^\mu$. I_0^μ is actually the transmitted current for an unpolarized beam. Let us remark that changing P into $-P$ is equivalent to changing μ into $-\mu$, so that $I_0^\mu = \frac{1}{2}(I_{P_0}^\mu + I_{-P_0}^\mu) = \frac{1}{2}(I_{P_0}^\mu + I_{P_0}^{-\mu})$ and $\Delta I_{P_0}^\mu = \frac{1}{2}(I_{P_0}^\mu - I_{-P_0}^\mu) = \frac{1}{2}(I_{P_0}^\mu - I_{P_0}^{-\mu}) = I_{P_0}^\mu - I_0^\mu$.

First, when injecting an unpolarized electron beam ($P = 0$) at very low incident energy $E_{\text{inc}} = 3.4$ eV (referred to the Fermi level E_F of the sample), we measure the transmitted current I_0^μ at $E_V = 1.7$ eV versus pulse amplitudes of the magnetic field [diamonds in Fig. 1(a)]. Such a measurement is equivalent to complete the hysteresis cycle of the sample. The largest transmissions are reached at high magnetic fields in the configurations

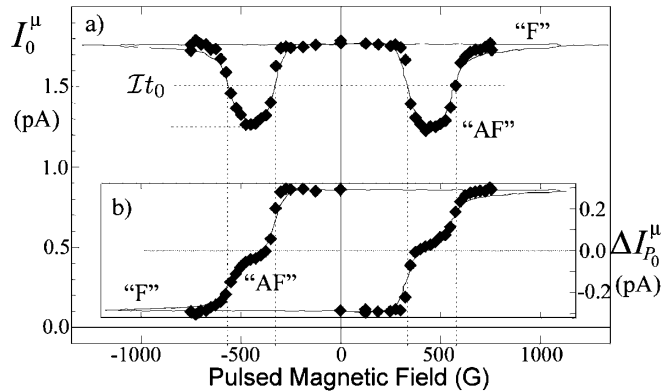


FIG. 1. (a) Transmitted current I_0^μ of an unpolarized beam injected at 3.4 eV above the sample's Fermi level versus magnetic field (\blacklozenge). The vacuum level of the exit face E_V is lowered to 1.7 eV. Corresponding fit calculated from Eq. (2) (solid line), I_{t_0} being the transmitted current when one Co layer is demagnetized (horizontal dotted line). (b) Spin-dependent transmitted current $\Delta I_{P_0}^\mu$ versus magnetic field (\blacklozenge). Scaled magneto-optical effect hysteresis loop showing the "F" and "AF" magnetization states (solid line). Vertical lines visualize the coercive fields of each Co layer. Both current measurements are normalized to 100 nA of incident current.

identified as F and $-F$ and sharply drop by about 30% when the bilayer is set to the AF or $-AF$ configurations. This transmitted asymmetry of an unpolarized electron beam between the parallel and antiparallel magnetization configurations is analogous to the spin-valve effect (or polarizing-analyzing effect) present in spin-valve transistors [10,11] and ballistic electron emission microscopy experiments [12]. The first cobalt layer polarizes the beam by passing a spin direction, whereas the second layer detects the projected spin component. Conditions of total extinction will be defined below.

Second, working with an incident beam of polarization $P = P_0$, the relevant quantity is $\Delta I_{P_0}^\mu$, which is reported on Fig. 1(b) versus the pulsed magnetic field. $\Delta I_{P_0}^\mu$ is directly measured by modulating the incident polarization between P_0 and $-P_0$ and using a lock-in amplifier. $\Delta I_{P_0}^\mu$ reproduces the magnetization cycle of the sample. One can easily identify the four magnetization states F, $-F$, AF, and $-AF$.

To describe quantitatively the transmission above E_F , we propose the following model. At very low E_{inc} , the electron distribution propagates ballistically within an energy range close to E_V by following an exponential attenuation versus distance. In this transport regime, no spin relaxation in the cobalt layers and in the 2.2 nm thick gold layer is assumed. At low energy, the electron inelastic mean-free path is large, i.e., of the same order as the cobalt layer thickness. Magnetization resulting from the distribution of opposite domains can be described by the respective domain fractions $\frac{1}{2}(1 \pm \eta_l)$, with l indexing the cobalt layers and η_l varying continuously from -1 to 1 . We define I as the current entering the *first* spin filter and \mathcal{P} its polarization. From the formalism of spin polarimetry [8,13,14] applied to the bilayer, the transmitted current is

$$I_P^\mu = I_{t_0}(1 + \eta_1 \eta_2 s_1 s_2 + \mathcal{P}(\eta_1 s_1 + \eta_2 s_2)), \quad (1)$$

where $\eta_1 s_1$ and $\eta_2 s_2$ are the Sherman functions of the Co layer (analyzer), and t_0 is the product of the spin-average transmissions of the four layers from the first magnetic layer to the exit surface. From Eq. (1), one obtains

$$I_0^\mu = I_{t_0}(1 + \eta_1 \eta_2 s_1 s_2), \quad (2)$$

and

$$\Delta I_{P_0}^\mu = I \mathcal{P} t_0 (\eta_1 s_1 + \eta_2 s_2). \quad (3)$$

Note that (2) implies that (i) a perfect spin-valve effect ($s_1 = s_2 = 1$) would yield a zero transmitted current I_0^μ in AF states ($\eta_1 \eta_2 = -1$) (extinction condition), and (ii) I_{t_0} is the transmitted current, for an unpolarized electron beam, when at least one of the Co layers is demagnetized (η_1 or $\eta_2 = 0$). In Fig. 1, the corresponding magnetization states are indicated by vertical dotted lines set at the coercive fields of the two Co layers. The

I_0^μ values associated with these states are the averaged transmitted currents between the F and the AF states $\frac{1}{2}(I_0^F + I_0^{AF}) = I_{t_0}$.

By using η_1 and η_2 values extracted from the hysteresis loop associated with each Co layer, Eq. (2) successfully fits the step variation of I_0^μ [solid lines in Fig. 1(a)]. For an unpolarized incident beam ($P = 0$), the asymmetry A_{I_0} in transmitted currents between F and AF states, deduced from Fig. 1(a), yields directly the value of $s_1 s_2 = A_{I_0} = (I_0^F - I_0^{AF}) / (I_0^F + I_0^{AF}) = 0.17 \pm 0.02$. On the other hand, for a spin polarized incident beam ($P = P_0$), the relevant asymmetry $A_{\Delta I_{P_0}}$ between the F and the AF states calculated from Fig. 1(b) gives the ratio $s_1 / s_2 = A_{\Delta I_{P_0}} = (\Delta I_{P_0}^F - \Delta I_{P_0}^{AF}) / (\Delta I_{P_0}^F + \Delta I_{P_0}^{AF}) = 0.70 \pm 0.03$. Obviously, this nonunity value expresses that in the AF configurations the saturated magnetizations of the two layers do not compensate, which is expected since both Co layers have different thicknesses. The spin selectivities of the two layers deduced from the above values of $s_1 s_2$ and s_1 / s_2 are $s_1 = 0.34 \pm 0.02$ and $s_2 = 0.49 \pm 0.03$. By analogy with usual spin polarimetry, we write

$$A_{I_{P_0}} = (I_{P_0}^F - I_{P_0}^{AF}) / (I_{P_0}^F + I_{P_0}^{AF}) = s^F \mathcal{P}, \quad (4)$$

which defines the Sherman function of the bilayer in the F state [8]. From the previous definition of I_0^μ and $\Delta I_{P_0}^\mu$, $A_{I_{P_0}}$ is equal to $\Delta I_{P_0}^F / I_0^F = 0.17 \pm 0.02$ and, using Eqs. (2) and (3), $s^F = (s_1 + s_2) / (1 + s_1 s_2) = 0.7 \pm 0.04$. From Eq. (4), we then obtain $\mathcal{P} = 0.24 \pm 0.02$, which is precisely the value of the polarization P_0 produced by the GaAs source that we use. This result shows that a calibration of the initial spin polarization is achieved and also validates the ballistic transport assumption at very low E_{inc} .

In order to verify the ballistic transport assumption at higher E_{inc} , we investigate the variation of I_0^μ relative to E_{inc} restricting ourselves to the F and AF states. In this experiment, the work function of the exit face has raised up to $E_{\text{inc}} = 2.5$ eV. The net increase in I_0^μ reveals the creation of secondary electrons, as seen on Fig. 2(a). As expected, the transmission is much higher in the F state. When $E_{\text{inc}} \gg E_V$, the central result is that the asymmetry $A_{I_0} = 0.12$ does not depend upon E_{inc} [Fig. 2(a)]. Since s is a decreasing function of the transport energy inside the spin filter [15], this invariance shows that transport in the ferromagnetic layers takes place at the same energy. The subjacent model is the creation of secondary electrons in the first atomic gold layers followed by ballistic propagation at energy close to E_V across the Co layers [16]. Consequently, according to (2) the current I entering the first cobalt layer increases almost linearly as a function of E_{inc} . Since secondary electrons are generated into the first Au layer, the initial polarization P_0 should be diluted by unpolarized secondary electrons excited from the Au conduction band. A first hint is given by the measurement of $\Delta I_{P_0}^\mu$, which is constant [see Fig. 2(b)]. This

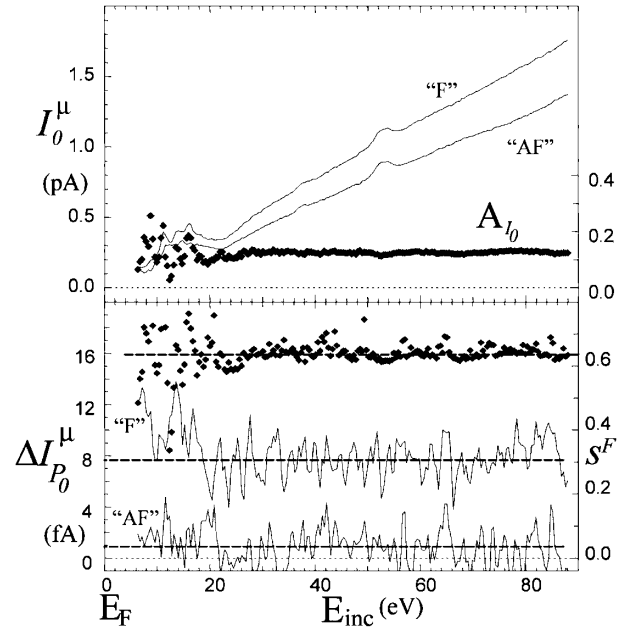


FIG. 2. (a) Inelastically transmitted currents I_0^F and I_0^{AF} in the F and AF configurations (left scale) and the asymmetry A_{I_0} measured versus the incident beam energy E_{inc} (right scale). (b) Difference in transmitted currents between P_0 and 0 spin polarized electron beam for the F and AF configurations (left scale) and the spin filter selectivity s^F . $E_{\text{inc}} = 0$ corresponds to the sample's Fermi level. Dotted lines give the averaged values. Data recorded for $E_V = 2.5$ eV.

means that $I\mathcal{P}$ is constant and therefore implies that \mathcal{P} is a decreasing function of E_{inc} (t_0 is constant since the ballistic transport occurs at the same energy E_V). To determine accurately this variation, we first calculate s^F from the measured asymmetries A_{I_0} and $A_{\Delta I_{P_0}}$. Figure 2(b) displays s^F as a constant $s^F = 0.62 \pm 0.05$, a lower averaged value than the previous one, since at $E_V = 2.5$ eV electrons travel farther above the Co d bands. Figure 3 shows the polarization $\mathcal{P} = A_{I_{P_0}} / s^F$ versus E_{inc} . The sharp fall is a direct evidence of the polarization dilution. The decrease is well reproduced by the following relation $\mathcal{P} = P_0 E_V / E_{\text{inc}}$, which asserts that the initial polarization P_0 is diluted by the multiplication factor E_{inc} / E_V produced by secondary electrons. This ratio can be derived from a model, in which δ -like d bands are considered at the Fermi level [14,17]. At each collision, an electron loses half of its energy by exciting a secondary electron. The mechanism of energy loss iterates until E_V is reached. This model is valid as long as E_V and the width of the d bands are negligible relative to E_{inc} . The data fit, excluding the measurements at low E_{inc} given by the polarization dilution model, is plotted in Fig. 3. The extrapolation of the fit to $E_{\text{inc}} = E_V$ gives again $\mathcal{P} = 0.24 = P_0$.

Coming back to the microscopic analysis of s_1 and s_2 , we introduce the spin-discriminating length δ and the average inelastic mean-free path in cobalt λ by $1/\delta = (1/\lambda_- - 1/\lambda_+)/2$ and $1/\lambda = (1/\lambda_- + 1/\lambda_+)/2$, where

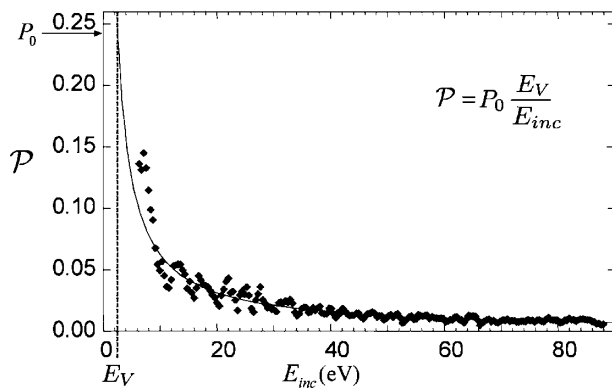


FIG. 3. Variation of the diluted polarization \mathcal{P} versus the incident energy E_{inc} . The polarization dilution law $\mathcal{P} = P_0 E_V / E_{inc}$ is drawn as a solid line. By extrapolating at $E_{inc} = E_V$, the initial polarization P_0 is retrieved.

λ_+ (λ_-) is the inelastic mean-free path in cobalt for majority (minority) spins. For each of the two layers l , we separate the bulk and interface discriminating powers, s_b and s_i defined in terms of transmission asymmetries as in [13]. Then, $s_{lb} = (t_+ - t_-)/(t_+ + t_-) = th(d_l/\delta) = th(a_b d_l/\lambda)$ and $s_i = th(2a_i)$, where $t_{\pm} = \exp(-d_l/\lambda_{\pm})$, $a_b = \lambda/\delta$ is the intrinsic spin-discriminating factor in cobalt, and a_i is the intrinsic spin-discriminating factor of one Co/Au interface. Then, the total Sherman function for the layer l is

$$s_l = (s_i + s_{lb})/(1 + s_i s_{lb}) = th(2a_i + a_b d_l/\lambda). \quad (5)$$

The Sherman function depends on the intrinsic quantities of the magnetic material a_i and a_b and on the reduced thickness d_l/λ . Using the values of $s_1 = 0.34$, $s_2 = 0.49$, $d_1 = 0.8$ nm, and $d_2 = 1.3$ nm, one obtains the following from Eq. (5): $\delta = 2.7$ nm and $2a_i = 0.06$. The value of δ , at 1.7 eV above the Fermi level, is in good agreement with the values previously reported [12]. The comparison of the value of $2a_i$ with $d_1/\delta = 0.30$ and $d_2/\delta = 0.48$ shows that the interface effect is much smaller than the bulk effect (it is equivalent to the bulk effect in a layer of thickness of 0.16 nm). The contribution of the two interfaces to the Sherman function is in fact of the order of the experimental uncertainty, since s_1 , s_2 , d_1 , and d_2 are not known with an accuracy better than 5%.

In conclusion, the analysis of the transmission through a ferromagnetic bilayer of free spin polarized electrons provides the physical quantities, which describe the spin-dependent transport: the spin-discriminating length in the ferromagnet, the Sherman function of the two layers, and

the bulk and interface contributions to the spin-filtering effect. Finally, the incident beam polarization P_0 is measured without the need for any external calibration and without relying upon any microscopic model of spin-dependent transport. Multilayered ferromagnetic structures open up the way to new self-calibrated electron spin polarimeters with high selectivity.

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