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Perturbative measurement of magnetoresistance

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In this paper, we report the development of a novel technique in which the magnetoresistance of nanostructures is perturbatively measured by transversally modulating the DC magnetic field. It measures the electrical transport counterpart of the transverse magnetic AC-susceptibility. The technique was developed in a conventional four-probe configuration in which a small DC current flows through the sample and a small transverse AC-field perturbs the equilibrium position of the sample magnetization. Lock-in detection, in-phase with the AC-perturbation, is used to measure the voltage signal between the two inner electrodes of the four-probe station. We successfully demonstrated that this signal is proportional to the product of the first derivative of sample resistance with respect to the equilibrium position of the magnetization times the second derivative of the energy with respect to the equilibrium position of the magnetization. These dependences add new features to the technique investigated here that were not captured by the investigations previously reported on the same subject. To show the effective use of the technique, we discuss its application in measuring magnetic properties of thin magnetic films in the non-saturated regime. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5047661>

I. INTRODUCTION

Measurements of the magnetic properties of nanostructures are usually performed by means of a variety of techniques which depending on the type of the external excitation and the time scale of response can be considered as static, quasi-static, or dynamic techniques.^{1,2} The basic principles on which the experimental techniques are created rely on different magnetic phenomena, such as magnetic induction,^{3–6} magnetic force,^{7,8} magneto-optical properties,^{9–11} magnetic resonances,^{12–15} light scattering by magnetic excitations,¹⁶ and magnetoresistance.^{17,18} To be suitable for investigation of magnetic properties of nanostructures, the development of new techniques has to consider two key aspects: (i) being able to measure weak signals and (ii) providing information on magnetic phenomena that are not easily obtained by well-established techniques. For example, the magnetic field values in which the magnetization reversal processes start in magnetic nanostructures¹⁹ are not easily measured by existent techniques. Therefore, the development of magnetic instrumentation dealing with weak signals, which operates in the non-saturation of the magnetization regime of field values, is of current interest.

Here we present the development of a perturbative measurement technique based on magnetoresistance effects, termed Perturbative Magnetoresistance (PMR), which can be used to characterize the magnetic properties of nanostructures in magnetic field ranges of non-saturation of the magnetization regime. The physical principle of the PMR can be understood as an extension of the AC-AMR measurements introduced by Venus *et al.*²⁰ In the AC-AMR technique, the authors used a double modulation detection where the magnetization was

perturbed by means of a small AC-field (100 Hz) superimposed with an AC-current (40 kHz) that flows through the sample. In Ref. 20, the double modulation detection was carried out with no external DC-field. In the PMR technique, the DC-field is always applied to the sample; therefore, the magnetization direction can be slowly reversed in the presence of a perpendicular weak oscillating field. The electrical excitation is obtained by applying a DC current through the sample, and the PMR signal is measured by means of a lock-in amplifier phase-locked to the reference signal. The principle of operation of the perturbative magnetoresistance (PMR) technique and the required instrumentation are described in Sec. II. Section III describes the interpretation of the data with an analytical model. This section also includes the validation of the technique and comparison of the data obtained by means of PMR with data obtained by the conventional techniques of the vibrating sample magnetometer (VSM) and conventional collinear magnetoresistance (MR). Conclusions are presented in Sec. IV.

II. PERTURBATIVE MAGNETORESISTANCE: PRINCIPLE OF OPERATION AND INSTRUMENTATION

In the PMR setup, perpendicular AC and DC magnetic fields are simultaneously applied to the sample. As shown below, this configuration makes the measured response be proportional to dR/dh_{ac} , where R is the sample resistance and h_{ac} is the perturbative magnetic field. On the other hand, if the AC field is applied parallel to the DC field, the response is proportional to dR/dH_{dc} .²¹ While the parallel configuration has been named magnetoresistance AC-susceptibility,

FIG. 1. Illustration of the PMR setup. While the fixed DC-current is applied between the outer probes, the voltage is measured between the inner probes. A spurious signal is induced between the inner probes due to the magnetic flux variation. The lock-in reference signal is provided by the same signal generator used to feed the AC field.

we named the perpendicular configuration as perturbative magnetoresistance.

To avoid any contact-resistance contribution, we employed the collinear four-point probe technique.²² Figure 1 illustrates the experimental setup used to perform PMR measurements. Four gold-coated spring contact probes were tightly pressed onto the sample surface, the DC electrical current is applied between the two outer probes, and the voltage signal is measured between the two inner contacts. The current varies from hundreds of microamps to few milliamps. In some measurements, we used an additional low-noise pass-band preamplifier (Stanford SR560) before the lock-in detection. The DC field is generated by using an electromagnet, and the perpendicular AC field is generated by using a pair of Helmholtz coils which is sinusoidally driven by a 90 W audio amplifier. In a typical measurement, the amplitude and frequency of the sinusoidal AC field at the sample are 1.0 Oe and 470 Hz, respectively. Since we have time dependent magnetic fields and closed circuits through the probes, two spurious signals need to be eliminated before measurement takes place: (i) An induced AC voltage, due to the perturbative magnetic flux, is generated in between the inner probes by h_{ac} (hatched area of Fig. 1). Before turning on the DC power supply, this spurious AC signal is detected by measuring the 1st harmonic component of the lock-in amplifier. This constant background signal (typically 80 μ V) does not depend on the DC field and is later subtracted from the magnetic field dependent signal. (ii) As the DC field is stepped upward or downward, there also appears a spurious contribution to the signal. To eliminate this spurious signal, the data acquisition program has a dead time between the acquisition of two consecutive data points that can be adjusted from 1.0 s to 15 s.

The physical principle of the PMR technique relies on the change in the electrical resistance of the sample when the

FIG. 2. Illustration of the equilibrium position of the magnetization $\vec{M}$ in the presence of the external magnetic fields H_{dc} and h_{ac} . The dashed arrows represent the time-varying positions of magnetization, oscillating around the equilibrium position given by ϕ_M . Here, the uniaxial anisotropy field is parallel to the x -axis.

magnetization weakly oscillates around its equilibrium position under the action of a weak time varying and spatially uniform external magnetic field. Figure 2 shows the vector configuration of the magnetization ($\vec{M}$) and applied magnetic fields H_{dc} and h_{ac} . The in-plane equilibrium position of $\vec{M}$ is given by the angle ϕ_M , and the time varying positions are illustrated by dashed arrows. Due to h_{ac} , the magnetization vector will oscillate around its equilibrium position that is obtained by minimization of the Zeeman and anisotropy energies.²³ As the electrical resistance weakly oscillates, all the harmonic components can be measured by means of the lock-in amplifier. By varying the magnetic DC-field, the equilibrium position of the magnetization $\phi_M(H_{dc}, h_{ac})$ and the electrical resistance $R = V(H_{dc}, h_{ac})/I_{dc}$ vary as well. Here, V is the measured voltage and I_{dc} is the DC-electrical current, which is kept constant. Thus, for a fixed value of H_{dc} and considering h_{ac} as a perturbation (smaller than the anisotropy fields involved), we can expand $V(H_{dc}, h_{ac})$ using Taylor's series expansion as

$$V(H_{dc}, h_{ac}) = V(H_{dc}, 0) + \left. \frac{\partial V}{\partial h_{ac}} \right|_{h_{ac}=0} h_{ac} + \frac{1}{2} \left. \frac{\partial^2 V}{\partial h_{ac}^2} \right|_{h_{ac}=0} h_{ac}^2 + \dots, \quad (1)$$

where $h_{ac}(t) = h_0 \cos \omega t$, $h_0 \cong 1.0$ Oe, and $f = \omega/2\pi = 470$ Hz. We used a lock-in amplifier (model EG&G 7260) to measure the first harmonic component of $V(H_{dc}, h_{ac})$, in-phase with the external AC-excitation. As the period of h_{ac} is around 2 ms, the time constant of the lock-in low-pass filter can be as low as 50 ms with no change in the result. Depending on the level noise, the time constant can be as high as 500 ms.

Defining V_{PMR} as the amplitude of the 1st harmonic term of Eq. (1), it can be written in terms of different variables,

$$V_{PMR} = h_0 \left. \frac{\partial V}{\partial h_{ac}} \right|_{h_{ac}=0} = h_0 I_{dc} \left. \frac{\partial R}{\partial h_{ac}} \right|_{h_{ac}=0} = h_0 I_{dc} \left. \frac{\partial R}{\partial \phi_M} \frac{\partial \phi_M}{\partial h_{ac}} \right|_{h_{ac}=0}. \quad (2)$$

To correctly interpret the V_{PMR} signal, we need to know the analytical expressions for both $\partial R/\partial \phi_M$ and $\partial \phi_M/\partial h_{ac}$ terms that appear in the above equation. The term $\partial R/\partial \phi_M$

accounts for the modulation of the resistance and depends on the underlying mechanism responsible for the magneto-transport change. The term $\partial\phi_M/\partial h_{ac}$ depends on the equilibrium position and is obtained by minimizing the free energy of the sample that contains all the relevant contributions to the magnetic free energy such as Zeeman, anisotropy, demagnetization, and exchange coupling. Therefore, the equilibrium position of magnetization (ϕ_{M_0}) is obtained from the minimization conditions of the magnetic free energy E given by

$$\left. \frac{\partial E}{\partial \phi_M} \right|_{\phi_{M_0}} = 0 \text{ and } \left. \frac{\partial^2 E}{\partial \phi_M^2} \right|_{\phi_{M_0}} > 0. \quad (3)$$

From the total derivative of $\partial E/\partial \phi_M$ with respect to h_{ac} , we obtain the expression for $\partial\phi_M/\partial h_{ac}$,

$$\begin{aligned} \frac{d}{dh_{ac}} \left(\frac{\partial E}{\partial \phi_M} \right) &= \left[\frac{\partial}{\partial \phi_M} \left(\frac{\partial E}{\partial \phi_M} \right) \right] \frac{\partial \phi_M}{\partial h_{ac}} + \frac{\partial}{\partial h_{ac}} \left(\frac{\partial E}{\partial \phi_M} \right) \\ &= \frac{\partial^2 E}{\partial \phi_M^2} \frac{\partial \phi_M}{\partial h_{ac}} + \frac{\partial^2 E}{\partial h_{ac} \partial \phi_M} = 0. \end{aligned} \quad (4)$$

Therefore the $\partial\phi_M/\partial h_{ac}$ term is given by

$$\frac{\partial \phi_M}{\partial h_{ac}} = - \frac{\frac{\partial^2 E}{\partial h_{ac} \partial \phi_M}}{\frac{\partial^2 E}{\partial \phi_M^2}}, \quad h_{ac} \rightarrow 0, \quad (5)$$

where the derivatives are evaluated at the equilibrium position ϕ_{M_0} .

To calculate the $\partial R/\partial \phi_M$ term, we consider the anisotropic magnetoresistance (AMR) effect¹⁷ as the mechanism responsible for the magnetoresistance. Thus,

$$\frac{\partial R}{\partial \phi_M} = -\Delta R \sin(2\phi), \quad (6)$$

where $\Delta R = R_{\parallel} - R_{\perp}$ is the AMR amplitude, $R_{\perp}$ is the resistance when DC current is applied perpendicular to $\mathbf{M}$, $R_{\parallel}$ is the resistance when DC current is applied parallel to $\mathbf{M}$, and ϕ is the angle between the magnetization and the DC-electrical current direction. As mentioned above, the terms given by Eqs. (5) and (6) are fundamental to interpret the measurement of the PMR signal given by Eq. (2).

III. THEORETICAL MODEL AND INTERPRETATION OF EXPERIMENTAL DATA

In this section, we apply the theoretical model developed in Sec. II to investigate the PMR of ferromagnetic thin films. By writing the magnetic free energy of the system and then using the minimization procedure, we obtain the PMR signal as given by Eq. (2). Later, this theoretical model will be applied to explain measurements carried out in polycrystalline thin films of permalloy ($\text{Ni}_{80}\text{Fe}_{20}$). The magnetic free energy density includes two contributions, Zeeman and uniaxial anisotropy energies, given by

$$\varepsilon = -\mathbf{M} \cdot (\mathbf{H}_{dc} + \mathbf{h}_{ac}) - K_u \left(\frac{\mathbf{M} \cdot \hat{\mathbf{u}}}{M} \right)^2, \quad (7)$$

where $\mathbf{M}$ is the saturation magnetization, $\mathbf{H}_{dc}$ and $\mathbf{h}_{ac}$ are the static and time-varying magnetic fields, $\hat{\mathbf{u}}$ defines the

uniaxial anisotropy direction, K_u is the uniaxial anisotropy constant such that the uniaxial anisotropy field is given by $\mathbf{H}_u = (2K_u/M)\hat{\mathbf{u}}$. In terms of the angles shown in Fig. 2, where the x -axis is parallel to $\hat{\mathbf{u}}$, the energy density is given by

$$\begin{aligned} \varepsilon &= -MH_{dc} \cos(\phi_M - \phi_{H_{dc}}) - Mh_{ac} \cos(\phi_M - \phi_{h_{ac}}) \\ &\quad - \frac{1}{2}MH_u \cos^2(\phi_M). \end{aligned} \quad (8)$$

Equation (2) requires the calculation of the second derivative of the energy with respect to h_{ac} and ϕ_M . Thus,

$$\frac{\partial^2 \varepsilon}{\partial h_{ac} \partial \phi_M} = -M \cos(\phi_M - \phi_{H_{dc}}) \quad (9)$$

and

$$\begin{aligned} \frac{\partial^2 \varepsilon}{\partial \phi_M^2} &= MH_{DC} \cos(\phi_M - \phi_{H_{dc}}) + Mh_{ac} \sin(\phi_M - \phi_{H_{dc}}) \\ &\quad + MH_u \cos(2\phi_M), \end{aligned} \quad (10)$$

where $\phi_{h_{ac}} = \phi_{H_{dc}} + \frac{\pi}{2}$. Using Eqs. (9) and (10) and taking limit of $h_{ac} \rightarrow 0$, the PMR signal given by Eq. (2) becomes

$$V_{PMR} = h_0 I_{dc} \Delta R \frac{\sin(2\phi_M) \cos(\phi_M - \phi_{H_{dc}})}{H_{dc} \cos(\phi_M - \phi_{H_{dc}}) + H_u \cos(2\phi_M)}, \quad (11)$$

where the DC-electrical current is applied parallel to the hard axis of magnetization, y -axis of Fig. 2. It is important to mention that to interpret the PMR data with Eq. (11). Finally, we need to calculate the equilibrium position of the magnetization as a function of H_{dc} , using Eq. (8). Also, in Eq. (6), we used $\phi = \phi_M - \phi_I = \phi_M - \frac{\pi}{2}$.

A. PMR signal validation

In this subsection, we discuss experimental data that are used to confirm the theoretical interpretation given previously. For instance, Eq. (2) shows that the PMR signal depends linearly on both the intensity of the DC-current (I_{dc}) and the AC-field amplitude (h_0). This linear dependence is only valid in the limit of $h_{ac} \rightarrow 0$, in which Taylor's expansion is applicable. For h_0 values higher than a threshold value, the linear dependence given by Eq. (2) is no more valid. It is important to mention that this threshold value depends on intrinsic magnetic parameters of the sample under investigation. To discuss the above assumptions, we investigate a sample with large PMR signal, a bilayer of Py(10 nm)/CoO(3 nm)/MgO(100). Here, Py is a permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) film deposited on a very thin layer of CoO, which is not in the antiferromagnetic phase. The films were sputter deposited on a substrate of MgO(100) by the DC magnetron sputtering technique.

Figures 3(a) and 3(b) show the DC field dependence of the PMR signal for different I_{dc} and h_0 values. Figure 3(a) shows the PMR signal as a function of H_{dc} for different h_0 values, where I_{dc} was kept constant at 4.0 mA and h_0 was varied from 0 to 9 Oe. The inset shows the PMR dependence for larger values of H_{dc} . The field value, where the PMR signal nulls, is named transition field (H_t). In the perturbative approximation, the H_t value should not depend on the external excitation h_0 . As shown in Fig. 3(c), H_t remains almost constant for

FIG. 3. (a) and (b) show the PMR signal as a function of H_{dc} , measured at room temperature for the sample Py(10 nm)/CoO(3 nm)/MgO(100). In (a), the DC current is fixed at 4.0 mA and the AC field amplitude is varied. In (b), the AC field amplitude is fixed at 2.9 Oe and the DC current is varied. (c) Transition field dependency as a function of h_0 . (d) Linear behavior of the amplitude of the PMR signal as a function of h_0 , for $I_{dc} = 4.0$ mA (hollow squares), and as a function of I_{dc} for $h_0 = 2.9$ Oe (solid circles).

$0 < h_0 \leq 2.5$ Oe, but it decreases for values higher than 2.5 Oe. Therefore, $h_0 = 2.5$ Oe characterizes the upper limit of the AC field and is the maximum field that should be used as the perturbative excitation for this sample. As shown by the pink symbols in Fig. 3(a), the PMR dependence with H_{dc} changes strongly for $h_0 \geq 9.0$ Oe. In Fig. 3(b), the AC-field amplitude (h_0) is kept constant at 2.9 Oe, while I_{dc} is varied. In this case, the dependence of the PMR signals with the magnetic field is a well behaved and symmetrical hysteresis curve. The H_t values do not depend on I_{dc} , and the PMR amplitude exhibits linear behavior as a function of both the AC-field amplitude and DC-current, as expected from the model introduced in Sec. III [Eq. (2)]. The value of the signal amplitude, shown in the vertical axis of Fig. 3(d), was obtained by taking the difference between maximum and minimum signals, divided by 2, i.e., $sign.amp. = \frac{PMR_{max} - PMR_{min}}{2}$.

B. Experimental results and physical interpretation

The experimental data discussed in this subsection were obtained by investigating the PMR signal of a single film of permalloy, sputter deposited on Si(100) with a thickness of 36 nm. To induce a large in-plane uniaxial anisotropy, the film was obliquely deposited with a tilt angle of 75° . As known, obliquely deposited metal films develop columnar microstructures that induce very large uniaxial anisotropy fields as well as large coercivities.^{24,25} Before the PMR measurements, the Py film was investigated by means of magnetoresistance (MR) and vibrating sample magnetometer (VSM) techniques. To understand the physical meaning of the PMR signals, we compared them with the magnetization hysteresis measurements obtained with VSM. Figure 4(a) shows the magnetization curve (black symbols) and PMR signal (blue symbols) as a

function of the DC field applied along the easy axis of magnetization. As the Py film was obliquely grown, it exhibits a coercive field of $H_C \approx 32$ Oe which is larger than typical values of H_C for Py polycrystalline films. A PMR signal, of few μV , was obtained by applying a DC current of 3.0 mA and $h_0 \approx 1.0$ Oe. Observe that in the saturated regime ($H > H_{sat}$ and $\phi_M = \phi_{H_{dc}} = 0$ or π), V_{PMR} is zero as expected from Eq. (11). Figure 4(b) shows a zoomed-in view of Fig. 4(a) in which the transition field ($H_t \approx 38$ Oe) measured by the PMR signal occurs for a field value larger than the coercive field value ($H_C \approx 32$ Oe). This is an important property of the transition field measured by the PMR technique. As the DC field value decreases, for instance, along the upper branch of the hysteresis curve, and the magnetization starts to deviate from the saturated state, the PMR signal smoothly increases (blue symbols) reaching a maximum at around the value of the coercive field, and then as the field increases, it suddenly jumps down. The region in which the PMR signal jumps from the maximum to the minimum value occurs in the region between the two vertical dashed lines, which is defined as the switching field distribution (SFD). SFD is the region between the fields at which the magnetization drops to +90% to -90% of the saturation value. To compare with a quantitative model, we calculated the PMR signal, supposing the simplified model of Stoner-Wohlfarth (SW) described by Eqs. (8)–(11), in which the magnetization direction was obtained by numerical energy minimization, using $H_u = 55$ Oe. The PMR signals, given by the solid red and blue lines, exhibit a qualitative agreement with the experimental data showing a sharp peak at the transition field, H_t . Actually, a model to explain the experimental PMR data needs to take into account the detailed multidomain magnetic configuration as well as the Py grain distribution. Magnetoresistance data exhibit the same behavior as the PMR

FIG. 4. Measurements performed with DC field applied parallel to the easy axis of magnetization for the obliquely deposited Py(36 nm)/Si(100). (a) Magnetization curve (black symbols) and PMR signal (blue symbols). (b) Zoomed-in view of (a) showing the down-field sweep (blue symbols) and the up-field sweep (red symbols) of the PMR signal. The solid lines represent the PMR signal calculated using the SW model of coherent rotation of the magnetization. In spite of being an approximation, the sharp peaks are associated with the transition field measured experimentally. (c) MR data in which the down-field sweep is given by the blue solid circles and the up-field sweep by the solid red circles, superimposed to the PMR signal.

signal as shown in Fig. 4(c), where the MR data (solid circles) are superimposed to the PMR signal (open circles).

Figure 5(a) shows the magnetization curve (black symbols) and PMR signal (blue symbols) as a function of the DC field applied along the hard axis of magnetization. The SW scenario, when the magnetic field applied is along the hard axis, predicts that the magnetization equilibrium angle rotates

FIG. 5. Measurements performed with the DC magnetic field applied parallel to the hard axis of magnetization for the obliquely deposited Py (36 nm)/Si(100). (a) Magnetization curve (black symbols) and PMR signal (blue symbols). (b) Zoomed-in view of (a) showing the down-field sweep (red symbols) and the up-field sweep (blue symbols) of the PMR signal. The solid lines were numerically obtained from the SW model that considers coherent rotation of the magnetization. The sharp peaks of the PMR signal are associated with the switching field that shows a small variation at the VSM curve. (c) MR data in which the down-field sweep is given by the red solid circles and the up-field sweep by the blue circles, superimposed to the PMR signal.

continuously toward either side of the hysteresis curve with no switching field and no hysteresis.^{26,27} In the sample investigated here, the magnetization curve shown in Fig. 5(a) exhibits a quasi-typical behavior of a hard axis curve with a saturation field value of around 125 Oe, a small hysteresis, and a very weak magnetization switching at ± 50 Oe. The weak magnetization switching, which is hard to be measured by VSM, can

easily be detected by the PMR technique, as given by the abrupt transitions also at ± 50 Oe, as shown in Fig. 5(a). Figure 5(b) shows a zoomed-in view of Fig. 5(a), in which the PMR signal is given by the red and blue open circles. The red and blue solid lines were obtained by means of the SW model, similar as in Fig. 4(b), but now considering the DC field applied parallel to the hard axis of magnetization. This is a clear demonstration that PMR can be used to detect feeble variation of the magnetization in fields out of the saturation range. Figure 5(c) shows the MR data (solid circles) superimposed to the PMR signal (open circles).

IV. CONCLUSIONS

In this paper, we present a novel characteristic of the perturbative magnetoresistance measurement technique, which was not explored by previous papers. We measured the 1st harmonic of the resistance signal, modulated by a small AC field perpendicular to a DC field. We named the signal obtained by the PMR signal. Equations (2), (5), and (6) show that the PMR signal depends on the product of the second derivative of the magnetic energy with respect to the equilibrium position of the magnetization times the first derivative of the resistance with respect to the equilibrium position of the magnetization. A parameter sensitive to the 2nd derivative is, for instance, the switching field of the magnetization that is defined as the point of instability close to the saturation state. It is difficult to measure the switching or nucleation field in the low field regime by usual techniques (see Refs. 19 and 27 to understand the difference between switching and nucleation fields). We verified that the amplitude of the AC field cannot be larger than a threshold above which the PMR line shape changes significantly. The PMR technique is very effective to measure the magnetic phenomena that vary significantly in the low field regime, such as the magnetization switching.^{28–31} Certainly, the principle of functioning of PMR and the results presented in this paper add a novelty to the set of techniques that use the AMR phenomenon to probe the magnetization state in magnetic thin films and multilayers.³² Also, it will be of interest to scientists and engineers working with the development of magnetic instrumentation that operates in the low magnetic field regime.

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