

Asymmetric field dependence of magnetoresistance in magnetic films

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We study an asymmetric in field magnetoresistance that is frequently observed in magnetic films and, in particular, the odd longitudinal voltage peaks that appear during magnetization reversal in ferromagnetic films, with out-of-plane magnetic anisotropy. We argue that the anomalous signals result from small variation in magnetization and Hall resistivity along the sample. Experimental data can be well described by a simple circuit model, the latter being supported by analytic and numerical calculations of current and electric field distribution in films with a gradual variation in the magnetization and Hall resistance.

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Onsager's reciprocity relations¹ are the cornerstone in understanding the field symmetry of magnetotransport measurements. Magnetoresistance or longitudinal resistivity (measured along the current flow direction) is predicted to be an even function of magnetic induction B while transverse (Hall) resistivity is specified to be odd with respect to B when a magnetic field is applied perpendicular to the sample plane. General acceptance of these rules is so common that, in numerous experimental cases, when current and voltage contacts cannot be arranged in a well defined five-probe geometry, the magnetoresistance and Hall-effect data are, respectively, extracted as the even and odd in field components of the measured four-probe signal. However, asymmetric in field magnetoresistance is quite frequently observed in magnetic materials (also in samples with fully symmetric magnetic properties and properly arranged current and voltage contacts),^{2,3} although this is rarely mentioned and discussed.^{4,5} A seeming violation of Onsager's law only recently attracted attention when sharp distinctive peaks of magnetoresistance, odd with respect to applied field, were found at magnetization reversal of ferromagnets with an out-of plane magnetic anisotropy.^{6,7} As argued by Cheng *et al.*⁶ the effect can appear when a domain wall (DW), located between the voltage probes, runs perpendicular to both magnetization and current. Electric fields generated by the extraordinary Hall effect (EHE) have opposite polarities on both sides of the DW, which can produce a circulating current loop and a respective extra voltage contribution. The model was used to explain the odd in field longitudinal voltage peaks in a specially designed Co-Pt multilayer film with a single DW gradually propagating along the sample. However, the effect was also observed in other samples with multiple domains,^{6,8} and the applicability of the "single wall" model in this general case is dubious.

In this paper we present two typical cases of asymmetric magnetoresistance observed in magnetic films and analyze their origins. We shall argue that the anomalous behavior can consistently be explained by a gradual variation in magnetization and Hall resistivity along the sample.

Figure 1(a) presents the longitudinal voltage V_l measured in a 6-nm-thick Ni film at 4.2 K as a function of a magnetic field applied perpendicular to the film plane in both field polarities. Thin Ni films possess the surface induced out-of-plane anisotropy at low temperatures⁹ responsible for the

hysteresis in the magnetoresistance curve. Anisotropic magnetoresistance is the origin of the negative magnetoresistance when a field is applied perpendicular to the electric current direction. V_l reaches maximum at magnetic fields corresponding to the coercive field value when the macroscopic out-of-plane magnetization crosses zero. Notably, the magnitude of the maximal voltage is not equal at two field polarities although the location of the peaks is the same. Similar asymmetric maxima can be found in several publications.^{10–12} The measured voltage is Ohmic (linear in electric current) and, if interpreted as magnetoresistance, its asymmetry would mean a violation of the Onsager rule.

Figure 1(b) presents the transverse (Hall) voltage for the same sample. Hall voltage in magnetic films depends on magnetization as:¹³

$$V_t = \frac{I}{t}(R_0 B + \mu_0 R_{\text{EHE}} M), \quad (1)$$

where I is electrical current, t is thickness, R_0 and R_{EHE} are the ordinary and extraordinary Hall coefficients, and B and M are the out-of-plane components of magnetic field induction and magnetization, respectively. A clear hysteresis loop is seen in Fig. 1(b) which is proportional to magnetization (the EHE term contribution is much larger than the ordinary one; therefore we neglect the ordinary Hall component in the following discussion).

Another striking example of asymmetric in field magnetovoltage is presented in Fig. 2(a). The longitudinal voltage measured in a Co/Pd multilayer sample (10 bilayers of 0.2-nm-thick Co and 1.1-nm-thick Pd, total thickness of 13 nm) is shown as a function of a magnetic field normal to the film. The sample was prepared by sequential e -beam deposition of Co and Pd layers on a GaAs substrate. It has the six-contact Hall bar geometry that is 5 mm wide and 15 mm long. The distance between longitudinal and transverse voltage contacts is 5 mm. The sample has a strong out-of-plane anisotropy, typical for Co/Pd multilayers. Sharp antisymmetric peaks are clearly observed at about 0.44 T when magnetization reverses its polarity. The antisymmetric peaks are superimposed with a slightly asymmetric magnetoresistance curve. It is important to note that the polarity of the odd peaks (positive in the negative field and negative in the posi-

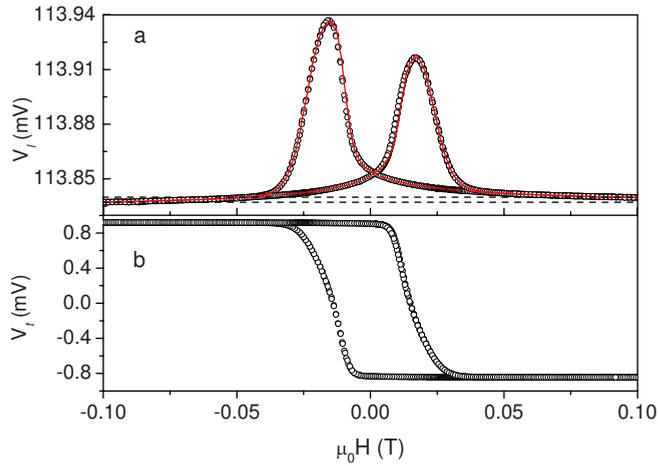


FIG. 1. (Color online) (a) Longitudinal voltage (\circ) measured in a 6-nm-thick Ni film at 4.2 K as a function of field applied perpendicular to the film plane. Solid line ($—$) is a fit calculated according to Eq. (8). Dashed lines are a guide for the eye that emphasizes the high-field asymmetry. (b) Hall voltage (\circ) measured simultaneously.

tive field) is reversed if measurement of the longitudinal voltage is done along the opposite edge of the film. A similar effect was found by us in FeTb films,¹⁴ and was previously reported in Co/Pt multilayers⁶ and (Ga,Mn)As epilayers⁷ with perpendicular magnetic anisotropy. Following Cheng *et al.*⁶ the odd in field longitudinal voltage signal can appear

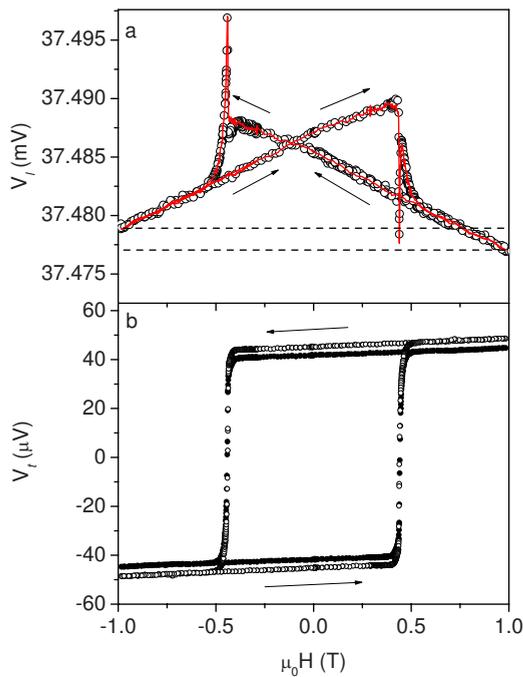


FIG. 2. (Color online) (a) Longitudinal voltage V_l (\circ) measured in the Co/Pd multilayer sample at 4.2 K as a function of applied field normal to the film plane. Solid line ($—$) is a fit according to Eq. (8). Dashed lines are a guide for the eye that emphasizes the high-field asymmetry. Arrows indicate the direction of the field sweep. (b) Hall voltages V_{H1} (\circ) and V_{H2} (\bullet) measured simultaneously at two locations along the sample.

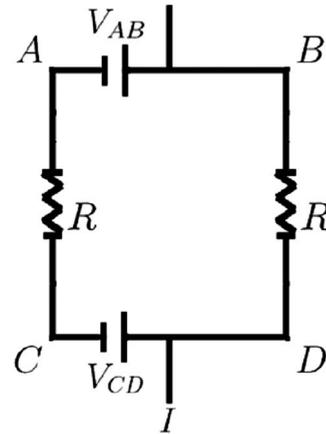


FIG. 3. Effective circuit representation of the sample.

when a domain wall separating two domains with up and down magnetizations is located between the voltage probes. Electric fields generated by the EHE depend on the local magnetization and have opposite polarities on both sides of the domain wall. These electric fields normal to the current can produce a circulating current loop around the domain wall and the respective additional voltage contribution along the sample. The model of a single domain wall assumes that magnetization is opposite at locations of the two longitudinal voltage contacts when the anomalous voltage peaks appear. This assumption can be tested experimentally by measuring the Hall voltage at two cross sections along the sample. Figure 2(b) shows V_H measured between two pairs of contacts transversal to the current direction, at two locations along the sample (\circ and \bullet), while the longitudinal voltage V_l , shown in Fig. 2(a), is measured simultaneously between a pair of longitudinal contacts. The magnetization reverses almost simultaneously at both locations {the difference in coercive fields is about 20 Oe, whereas the reversal width defined as the field span over which V_l varies between 10% and 90% is approximately 700 Oe [Fig. 2(b)]}. The antisymmetric longitudinal voltage peaks [Fig. 2(a)] appear with the reversal of magnetization. The width of the peaks is equal to the width of the magnetization (Hall voltage) reversal. This observation does not agree with the “single domain-wall” picture that predicts opposite Hall voltage polarities at two cross sections when the anomalous peaks appear. Two more experimental results are important for future discussion: (i) V_l signals at two cross sections are similar in shape but differ in magnitude in the magnetically saturated state at high fields by approximately 8%, and (ii) macroscopic magnetization is not uniform: there is a small but finite difference in coercive fields along the sample.

Although the single wall model is not in agreement with the experimental data, one can assume that the transverse voltage is not uniform along the sample. We, therefore, model the sample as a simple circuit shown in Fig. 3. $V_{AB}(H)$ and $V_{CD}(H)$ represent the transverse voltage generated by Hall effects at two cross sections AB and CD, while two equal resistors R are positioned between A and C, and B and D. Field dependence of the resistors $R(H)$ is the usual symmetric in magnetic induction magnetoresistance. Following Kirchhoff’s circuit laws, the longitudinal voltages at two edges of the sample are

$$V_{AC} = \frac{IR + V_{AB} - V_{CD}}{2},$$

$$V_{BD} = \frac{IR - V_{AB} + V_{CD}}{2}. \quad (2)$$

Voltage measured along the sample would differ from the ordinary Ohmic $\frac{1}{2}IR$ if $(V_{AB} - V_{CD}) \neq 0$, i.e., $V_{AB} \neq V_{CD}$. The field symmetry of $(V_{AB} - V_{CD})$ is even in the case of, e.g., nonuniform planar Hall-effect contribution,¹⁵ or odd when the ordinary and/or extraordinary Hall effects are present. In this case $(V_{AB} - V_{CD})$ is given by

$$V_{AB} - V_{CD} = \mu_0 I \left(\frac{R_{EHE,AB} M_{AB}}{t_{AB}} - \frac{R_{EHE,CD} M_{CD}}{t_{CD}} \right), \quad (3)$$

with t_{AB} and t_{CD} as the local thickness, M_{AB} and M_{CD} as the local magnetizations, and $R_{EHE,AB}$ and $R_{EHE,CD}$ are the EHE coefficients at cross sections AB and CD, respectively.

Several mechanisms can cause a nonuniform transverse voltage along the sample. The simplest is a gradual variation in thickness t [Eq. (1)] due to either unintended imperfection of fabrication or when wedge samples are studied. This argument is applicable to any material including nonmagnetic metals¹⁶ and semiconductors.¹⁷ In magnetic materials there are additional mechanisms that can affect the EHE coefficient R_{EHE} . In thin ferromagnetic films R_{EHE} depends on the thickness and diverges in the thin-film limit due to an enhanced surface scattering.^{3,18} In granular ferromagnetic or superparamagnetic films R_{EHE} depends on size, density, and shape of magnetic clusters that might not be uniform along the sample due to deposition and annealing procedures.⁴ If magnetization is uniform along the sample ($M_{AB} = M_{CD}$), Eq. (3) gives

$$V_{AB} - V_{CD} = \left(1 - \frac{R_{EHE,CD} t_{AB}}{R_{EHE,AB} t_{CD}} \right) V_{AB}. \quad (4)$$

The longitudinal voltage V_{BD} can then be presented as

$$V_{BD} = \frac{IR}{2} - \alpha V_{AB}, \quad (5)$$

where α is a coefficient that depends on thickness and R_{EHE} variation along the sample. The first term on the right-hand side of Eq. (5) is even with respect to the field while the second is odd and proportional to the transverse voltage. V_{AB} is a monotonic function of field [see Fig. 2(b)]; therefore Eq. (5) can explain the high-field asymmetry of the longitudinal voltage in Fig. 2(a) but not the antisymmetric peaks at the magnetization reversal. We then assume that magnetization is not uniform and reverses gradually along the sample with raise of the applied field. The local magnetization values $M_{AB}(H)$ and $M_{CD}(H)$ are connected by

$$M_{CD}(H) = M_{AB}(H) - \Delta H \frac{\partial M_{AB}(H)}{\partial H}, \quad (6)$$

where ΔH is the increase in applied field needed to propagate the magnetization reversal from the cross section AB to CD. Then, to the first order of ΔH :

$$V_{AC}(H) = \frac{1}{2} \left(IR(H) + \frac{\Delta H I \mu_0 R_{EHE,AB}}{t} \frac{\partial M_{AB}(H)}{\partial H} \right),$$

$$V_{BD}(H) = \frac{1}{2} \left(IR(H) - \frac{\Delta H I \mu_0 R_{EHE,AB}}{t} \frac{\partial M_{AB}(H)}{\partial H} \right), \quad (7)$$

where we assume that $t_{AB} = t_{CD} \equiv t$ and $R_{EHE,AB} = R_{EHE,CD}$. The second term in Eq. (7) is odd with respect to the field (ΔH is odd), and can be significant in materials with a large EHE coefficient and sharp reversal of magnetization, as in thin ferromagnetic films with the out-of-plane anisotropy. The shape of $\Delta H \frac{\partial M_{AB}(H)}{\partial H}$ has a strong peak at magnetization reversal; therefore, this term can account for the antisymmetric peaks, as in Fig. 2(a), or for a significant difference in the maximal resistance in Fig. 1(a). If both the gradual reversal of magnetization and the variation in the saturated high-field Hall voltage along the sample are considered, the combination of Eqs. (1), (5), and (7) gives

$$V_{BD}(H) = \frac{1}{2} \left(IR(H) - \Delta H \frac{\partial V_{AB}(H)}{\partial H} \right) - \alpha V_{AB}(H). \quad (8)$$

We applied Eq. (8) to fit the experimental data both for the Ni film [Fig. 1(a)] and Co/Pd multilayer [Fig. 2(a)] by using the measured transverse voltage $V_{AB}(H)$, and two fitting parameters α and ΔH . The asymmetric magnetoresistance of Ni [solid line in Fig. 1(a)] was calculated with $\Delta H = 2.2$ Oe and $\alpha = 1.4 \times 10^{-3}$. The fit for the Co/Pd multilayer, shown in Fig. 2(a) by a solid line, was calculated with $\alpha = 2 \times 10^{-2}$ and $\Delta H = 24$ Oe. This value of ΔH is in good agreement with the measured 22 Oe difference in coercive fields between cross sections AB and CD. It should be noted that only minor inhomogeneity (α) and nonuniformity of magnetization reversal (ΔH) along the sample are sufficient to generate large anomalous signals. A possible cause for variation in the coercive field along the sample is the thickness variation. Magnetization reversal was reported^{6,19} to propagate along wedge shaped samples with thickness variation of a few percent only. Other possible causes are variation in surface roughness and adhesion to the substrate which are suspected²⁰ of inducing a transition from nucleation dominated reversal to domain-wall-motion reversal.

Although the model presented above is in a good agreement with the experimental data, one can wonder if a simple circuit (Fig. 3), which has only two current channels, provides a reliable description of a macroscopic sample. In the following we present a more rigorous derivation of the electric potential along an infinitely long sample with variable thickness and Hall resistivity, and show that in the proper limit the result is identical to Eq. (8). In order to reduce the problem to two dimensions, we follow Ref. 16 and define the following two-dimensional (2D) fields:

$$\langle \vec{j}(x,y) \rangle \equiv \frac{1}{t} \int_0^{t(x,y)} \vec{j}(x,y,z') dz', \quad (9)$$

$$\langle \vec{E}(x,y) \rangle \equiv \frac{1}{t(x,y)} \int_0^{t(x,y)} \vec{E}(x,y,z') dz', \quad (10)$$

where E is electric field, j is the current density, t is the average sample thickness, and $t(x,y)$ is the actual sample thickness at each point. The two-dimensional current distribution is determined by

$$\langle \vec{E}(x,y) \rangle = \frac{t\vec{\rho}(x,y)}{t(x,y)} \cdot \langle \vec{j}(x,y) \rangle, \quad (11)$$

$$\vec{\nabla} \cdot \langle \vec{j}(x,y) \rangle = 0, \quad (12)$$

and to a good approximation by

$$\vec{\nabla} \times \langle \vec{E}(x,y) \rangle = 0, \quad (13)$$

where $\vec{\rho}(x,y)$ is the spatially dependent resistivity tensor. Boundary conditions are set to prevent current flow normal to the sample edges. Exponential variation in thickness along the sample with constant Hall resistivity was analyzed by Bruls *et al.*¹⁶ The equations were found to be identical to those describing a sample with an exponential variation in charge-carrier density.¹⁷ Adaptation of the latter case gives the field dependent potential along the sample as

$$\varphi(x,y,H) = \frac{-\rho(H)I\beta(H)\exp\left(\frac{-x+\beta(H)y}{a}\right)}{t\left[1-\exp\left(\frac{\beta(H)w}{a}\right)\right]}, \quad (14)$$

where $\rho(H)$ is resistivity, a is the length scale over which the sample thickness changes by a factor of e , w is the sample width, and $\beta(H)$ is the ratio of Hall and longitudinal resistivities. In the limit of $a \gg x$ and $a \gg \beta(H)w$, Eq. (14) can be reduced to

$$\varphi(x,y,H) = -\frac{\rho(H)I}{tw}[x+\beta(H)(w/2-y)] + \frac{\rho(H)Ix^2}{2twa} + \frac{\rho(H)I\beta(H)x(w/2-y)}{twa}, \quad (15)$$

where $a=Lt/\Delta t$, with L being the distance between the longitudinal voltage probes and Δt the change in sample thickness between locations of the longitudinal probes (at $x = \pm L/2$). The first term in Eq. (15) consists of the standard longitudinal and transverse voltages of a homogeneous sample. The second and third terms are corrections to the potential due to the thickness variation. The second term does not contribute to longitudinal voltage since it is symmetric in x . The third term is proportional to the Hall voltage and changes sign depending on the location of the probes (at $y=0$ or $y=w$).

Linear variation in Hall resistivity due to change in charge-carrier density along the sample was analyzed by Ilan *et al.*²¹ in 2D electron gas. In the case of magnetic materials we ascribe the gradient of Hall resistivity to linear variation in both magnetization and $R_{\text{EHE}}(t)$ along the sample so that

$$\frac{\partial \rho_{xy}(x,H)}{\partial x} = \mu_0 M(0,H) \frac{\partial R_{\text{EHE}}(t)}{\partial t} \frac{\Delta t}{L} + \mu_0 R_{\text{EHE}}(0) \frac{\partial M(x,H)}{\partial x}, \quad (16)$$

where $M(0,H)$ and $R_{\text{EHE}}(0)$ are the values of magnetization and R_{EHE} at $x=0$. For ρ_{xy} varying along the x coordinate only, and $l_x \gg w$, the potential along the sample is given by

$$\varphi(x,y,H) = -\frac{\rho(H)I}{tw}[x+\beta(H)(w/2-y)] - \frac{\rho(H)Ix(w/2-y)}{twl_x(H)}, \quad (17)$$

where $l_x(H) = \rho(H)[\partial \rho_{xy}(x,H)/\partial x]^{-1}$.²¹ The first term in Eq. (17) corresponds to the potential distribution in a homogeneous sample, whereas the second term is the correction due to a spatial variation in the Hall resistivity. Since the correction terms in Eqs. (15) and (17) are small and of different origins, they are additive (higher order corrections are neglected). The longitudinal voltage can be calculated from Eqs. (15) and (17) as

$$\begin{aligned} V_l(H) &= \varphi(x=-L/2,H) - \varphi(x=L/2,H) \\ &= \frac{\rho(H)IL}{tw} \pm \frac{I\mu_0 M(0,H)R_{\text{EHE}}(0)}{2t} \\ &\quad \times \left(\frac{\Delta t}{R_{\text{EHE}}(0)} \frac{\partial R_{\text{EHE}}(t)}{\partial t} - \frac{\Delta t}{t} \right) \\ &\quad \pm \frac{LI\mu_0 R_{\text{EHE}}(0)}{2t} \frac{\partial M(x,H)}{\partial x}, \end{aligned} \quad (18)$$

where the \pm sign stands for $y=0(+)$ and $y=w(-)$. The first term in Eq. (18) is simply $IR(H)$, the second term is the correction due to the thickness variation and is proportional to the transverse voltage, and the last term is the correction due to a nonuniform magnetization along the sample. Finally, by assuming a constant ratio between the change in an applied field ΔH and the propagation of the magnetization reversal over a distance L , we calculate

$$\begin{aligned} V_l(H) &= IR(H) \pm \frac{1}{2} \left(\frac{\Delta t}{R_{\text{EHE}}(0)} \frac{\partial R_{\text{EHE}}(t)}{\partial t} - \frac{\Delta t}{t} \right) \\ &\quad \times V_t(0,H) \pm \frac{\Delta H}{2} \frac{\partial V_t(0,H)}{\partial H}, \end{aligned} \quad (19)$$

with $V_t(0,H) = \frac{I\mu_0 R_{\text{EHE}}(0)M(0,H)}{t}$. Equation (19) is identical to Eq. (8) obtained from the circuit model.

The analytic calculation was done for an infinitely long sample. In order to treat a finite sample, numerical calculations were carried out. Following Hajjar and Mansuripur,²² the current distribution was calculated by taking the finite difference version of Eqs. (12) and (13) on a two-dimensional rectangular lattice. Boundary conditions were added along the length of the sample edges together with the current source and drain. The sample dimensions were chosen equal to the actual geometry of the Co/Pd multilayer film, in which $L=w$ and the total sample length is $3w$. The effective resistivity tensor that was used included both thickness and magnetization gradients along the sample:

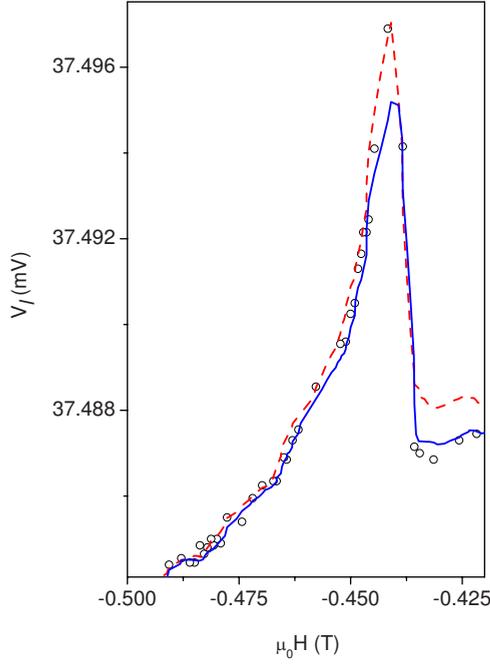


FIG. 4. (Color online) Longitudinal voltage V_l (\circ) measured in the Co/Pd multilayer sample at 4.2 K as a function of field normal to the film plane in the peak region. Solid line (—) is a fit calculated by using Eqs. (20) and (21) while dashed line (- -) is a fit according to Eq. (8).

$$\frac{t\rho(x,H)}{t(x)} = \frac{\rho(H)}{1+x\frac{\Delta t}{tL}} \begin{pmatrix} 1 & \beta(H) + \frac{x}{l_x(H)} \\ -\beta(H) - \frac{x}{l_x(H)} & 1 \end{pmatrix}. \quad (20)$$

In order to obtain $l_x(H)$ at each field value, the normalized change in magnetization between locations AB and CD was estimated as

$$\frac{\Delta M(H)}{M_S} = \frac{V_{CD}(H)}{V_{CD,S}} - \frac{V_{AB}(H)}{V_{AB,S}}, \quad (21)$$

where M_S is the saturation magnetization, and $V_{AB,S}$ and $V_{CD,S}$ are the saturated values of V_{AB} and V_{CD} , respectively. The normalized magnetization slope along the sample is then $\frac{\Delta M(H)}{LM_S}$. The solid line in Fig. 4 presents the simulation of the field dependent longitudinal voltage in the peak region for the Co/Pd multilayer sample with a single fitting parameter $\Delta t/t=0.05$. The dashed line was calculated by Eq. (8) and is shown here for comparison. Numerical results agree nicely with the experimental data (\circ). A snapshot of the simulated sample potential during a gradual magnetization reversal (magnetization is zero at $x=0$ at applied field of 0.44 T) is shown in Fig. 5. Large Ohmic component $\frac{\rho(H)lx}{tw}$ was subtracted for clarity. It is clearly seen that the potential gradient along the sample has opposite polarities at two edges of the

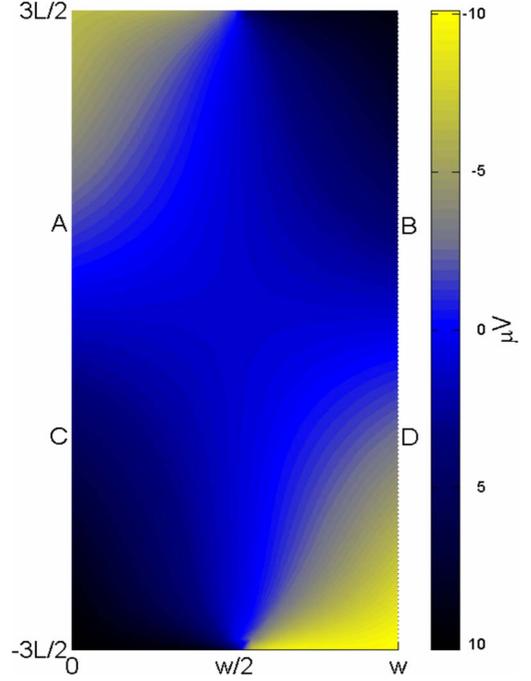


FIG. 5. (Color online) Numerical calculation of electric potential generated by a nonuniform magnetization reversal. The standard IR contribution is subtracted for clarity. A, B, C and D correspond to locations of the voltage probes.

sample. It is important to note that, due to the sharpness of magnetization reversal in films with an out-of-plane anisotropy, a minor delay in coercive field (20 Oe as compared with 700 Oe of the reversal width) results in a relative difference of magnetization of up to about 20% between cross sections AB and CD, which, respectively, leads to distinctive voltage peaks in magnetoresistance.

To summarize, we studied the asymmetric field dependence of magnetoresistance in magnetic films. We argue that minor variation in thickness, Hall coefficient, and nonuniform magnetization reversal along the sample can explain the anomalous phenomena. We show that a nonuniform variation in the Hall voltage along the sample generates an additional odd in field longitudinal voltage signal proportional to the field derivative of the transverse voltage. This additional signal can be significant when the Hall voltage varies sharply with the applied field, such as in the case of magnetization reversal in films with perpendicular magnetic anisotropy, studied here, at superconducting transitions or in materials demonstrating the quantum Hall effect. The fingerprint of the mechanism is the reversal of the asymmetry when the longitudinal voltage is measured along the opposite edge of the sample.

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