

Hall effect in ferromagnetic nanomagnets: Magnetic field dependence as evidence of inverse spin Hall effect contribution

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We measure magnetic field dependence of the Hall angle in a metallic ferromagnetic nanomagnet where the adopted mechanisms of Hall effect predict linear plus a constant dependence on the external field originating from the ordinary and anomalous Hall effects, respectively. We suggest that the experimentally observed deviations from this dependence are caused by the inverse spin Hall effect (ISHE) and develop a phenomenological theory, which predicts a unique nonlinear dependence of the ISHE contribution on the external magnetic field. Perfect agreement between theory and experiment supports the considerable role of the ISHE in Hall transport in a ferromagnetic metal.

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The Hall effect (HE) describes a generation of an electric current perpendicularly to the bias current, which flows along an applied electric field. The magnitude of the HE is measured by the Hall angle $\alpha_{\text{HE}} = \sigma_{xy}/\sigma_{xx}$, which is defined as the ratio of nondiagonal σ_{xy} and diagonal σ_{xx} conductivities. Early experiments of a measurement of the HE in a nonmagnetic metal and semiconductor detected the simplest counterpart of the phenomenon, which is called the ordinary Hall effect (OHE) [1]. Further studies of the HE in magnetic materials revealed several other mechanisms, each of which is fundamentally different from the OHE.

Historically, the first considered mechanism of the HE is the OHE, in which the carrier motion perpendicular to the external magnetic field and bias current is generated by the Lorentz force. The OHE contribution is proportional to the external magnetic field $\sim \alpha_{\text{OHE}}H$, where α_{OHE} is the OHE coefficient. Another HE contribution, which exists in a ferromagnetic material, is the anomalous Hall effect (AHE). The AHE occurs due to the scattering of carriers on the aligned local magnetic moments in a ferromagnet and therefore the AHE is proportional to the total local magnetic moment [2]. The AHE contribution is independent of external magnetic field H if the local magnetic moments are field independent. The joint contribution of the OHE and AHE to the Hall angle is the sum of two contributions: The field-independent AHE $\sim \alpha_{\text{AHE}}$ and the linear OHE $\sim \alpha_{\text{OHE}}H$. The above constant plus linear dependence, which is the standard prototypical case encountered in plenty of the magnetic compounds, is a target of the recent theoretical efforts [3].

More complex nonlinear behavior of the HE, which does not fit into the constant plus linear dependence, points either to the high-field corrections of prototypical OHE and AHE contributions or to an additional mechanism of HE. In the following, the lowest order external field corrections $\sim H$ are excluded due to our special symmetrized experimental scheme. For example, the so-called planar Hall effect [4] is

even in magnetic field and cannot contribute to our results. Among other mechanisms of the nonlinear HE we exclude the quantum Hall effect, which requires a cryogenic temperature and a high magnetic field [5], which is not the case of our room temperature measurement in a field less than 1 Tesla. We can also exclude tiny nonlinear corrections to the OHE because even the contribution of the OHE itself is standardly small in metallic conductors due to electron-hole compensation. Accordingly, our subsequent measurements show that the OHE is more than an order of magnitude smaller than the observed $d\alpha_{\text{HE}}/dH \approx 30$ mdeg/kG. One can also neglect the impurity driven nonlinear contribution to HE [6] in a ferromagnet because, in contrast to a paramagnet, the spins of local defects are firmly aligned along the local magnetic moments and cannot be independently altered by an external magnetic field. Hence, these spins contribute only to field-independent AHE.

Therefore, we are aware of only one possible candidate for the observed nonlinearity. It is the inverse spin Hall effect (ISHE), which describes the fact that an electrical current is created perpendicularly to the flow of the spin-polarized current [7–9]. The ISHE originates from the spin dependence of electron scatterings due to the spin-orbit interaction. For example, in the case of a spin-polarized electron gas, the scattering probabilities of spin-up and spin-down electrons are different for the left and right scattering directions. Since there are more electrons that are scattered, e.g., to the left than to the right, an electron current flows from the left to the right. This mechanism of the ISHE is called the skew scattering mechanism [10]. The ISHE requires the existence of the spin-polarized electrons and, therefore, it occurs only in a spin-polarized electron gas. In a ferromagnetic metal the electron gas is spin polarized even in an equilibrium and any electrical current in a ferromagnetic metal is spin polarized. As a result, any electrical current in a ferromagnetic metal always experiences the ISHE and there is a flow

of the Hall current perpendicularly to the bias current. The ISHE-type Hall current is proportional to the number of spin-polarized electrons in the ferromagnetic metal. There are several methods to change the number of spin-polarized electrons in a ferromagnetic metal and therefore the magnitude of the ISHE. For example, the spin polarization of the conduction electrons can arise due to the influence of an external magnetic field, the mechanism considered by Landau and Lifshitz a long time ago [11]. This contribution is not merely proportional to the external magnetic field because of the influence of other processes with a field-independent rate, such as, e.g., the relaxation of spin polarization or an additional spin polarization induced by scattering of polarized magnetic moments.

Despite numerous studies of the HE, the possibility of the ISHE contribution in a ferromagnetic metal was never addressed experimentally or theoretically. In contrast, the ISHE contribution to the HE in a paramagnetic material has been verified experimentally. In equilibrium the electron gas is not spin polarized in a nonmagnetic material and there is no ISHE contribution. Only when the spin polarization is externally created can the ISHE contribution be detected and identified. For example, the spin polarization in a paramagnetic AlGaAs/GaAs heterojunction was created by circularly polarized light. The dependence of the measured Hall angle on the degree of circular polarization and therefore on the spin polarization is clearly detected [12], confirming the existence of the ISHE contribution to HE. Additional proof of the ISHE contribution is that the measured Hall angle exponentially decreases with an increased distance between the focused spot of a laser beam and the Hall probe as would be expected for a diffused spin current.

A measurement of the ISHE contribution in a ferromagnetic metal is more difficult, because of the existence of the ISHE contribution even in equilibrium. The presence of the ISHE can be evaluated from the nonlinear dependence of the Hall angle on an external magnetic field. Another difficulty in detecting the ISHE lies in the fact that the average local magnetic moment can be field dependent too, usually due to reordering of magnetic domains, causing nonlinearity in the AHE contribution. One cannot pin down the ISHE if alignment of localized moments occurs due to an external magnetic field as happens in a paramagnet [6,13]. An experimental identification of a pure ISHE contribution requires appreciable independence of the local magnetic moment on the external magnetic field. A nanomagnet made of a ferromagnetic metal with the perpendicular magnetic anisotropy (PMA) [14] is a unique object for such a measurement. In the nanomagnet, the magnetic moments are firmly aligned perpendicularly to the film surface due to the strong PMA effect. The nanosize of the nanomagnet ensures a one-domain state, in which all localized moments are aligned in one direction. As a result, the AHE contribution becomes essentially independent of the external magnetic field, and the joint contribution of the OHE and AHE is strictly a sum of the field-independent AHE α_{AHE} and linear OHE $\sim \alpha_{\text{OHE}}H$ terms. Such a simple dependence gives a unique possibility to detect the ISHE phenomenon unambiguously, because the deviation of the field dependence of α_{AHE} from the “constant plus linear” dependence can occur solely due to the ISHE.

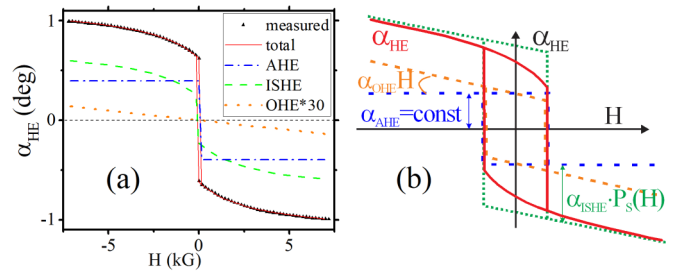


FIG. 1. Hall angle vs perpendicular magnetic field H . (a) Experimental data as a sum of three contributions: Triangles denote the total measured angle $\alpha_{\text{HE}}(H)$, the red line is the sum of contributions, and the blue/green/orange lines are individual AHE/ISHE/OHE contributions, respectively. (b) Schematic plot showing the shape of the hysteresis loop for different contributions. The solid red line is the measured (total) angle. The dashed blue horizontal line is the hypothetical loop in the absence of OHE and ISHE. The slanted dashed orange line is the loop in the absence of only ISHE. The dotted green line is the loop in the hypothetical case when all conduction electrons are spin polarized, $P_S = 1$.

In this Rapid Communication we develop a phenomenological theory of the ISHE dependence on the external magnetic field H and perform field-dependent measurements of the HE angle in numerous nanomagnets made of FeB and $\text{Fe}_{0.4}\text{Co}_{0.4}\text{B}_{0.2}$, which are the ferromagnetic metals with strong PMA effect. A comparison of the theoretical and experimental dependence of the Hall angle on the external magnetic field provides unambiguous proof of the importance of the ISHE in a metallic ferromagnet.

Theory. The Hall angle α_{HE} in our measurement can be presented as a sum of three terms:

$$\alpha_{\text{HE}}(H) = \alpha_{\text{OHE}}H + \alpha_{\text{AHE}} + \alpha_{\text{ISHE}}P_S(H). \quad (1)$$

All coefficients $\alpha_{\text{AHE},\text{OHE},\text{ISHE}}$ can be considered as field independent and the function $P_S(H)$ is the spin polarization of the conduction electrons in the external field H . Figure 1 shows how $\alpha_{\text{HE}}(H)$ can be divided into three contributions.

The ISHE originates from the spin-dependent scatterings of the conduction electrons [15–22]. For example, in the case of the skew-scattering mechanism [10], the amounts of spin-polarized conduction electrons scattered into the left/right directions are different leading to the ISHE which is, naturally, linearly proportional to the number of spin-polarized electrons. Since the Hall angle is a ratio of the nondiagonal σ_{xy} and diagonal σ_{xx} conductivities, the ISHE contribution to the Hall angle is proportional to the relative spin polarization

$$P_S(H) = n_{\text{SP}}/(n_{\text{SP}} + n_{\text{SU}}) \quad (2)$$

in the external magnetic field H . Here, we divided all relevant conduction electrons $n_{\text{SP}} + n_{\text{SU}}$, which participate in charge transport, into the group of the spin-unpolarized electrons n_{SU} , in which the numbers of spin-up and -down states are equal, and the group of spin-polarized electrons n_{SP} , whose spin direction is the same. Both the spin-polarized and spin-unpolarized states $n_{\text{SP}} + n_{\text{SU}}$ contribute to the diagonal component σ_{xx} of the conductivity, whereas only the spin-polarized states n_{SP} contribute to σ_{xy} .

The conversion rate between spin-polarized n_{SP} and spin-unpolarized n_{SU} electrons can be calculated as [23]

$$\frac{\partial n_{\text{SP}}}{\partial t} = \left[\frac{n_{\text{SU}}}{\tau_M} - \frac{n_{\text{SP}}}{\tau_{\text{rel}}} \right] + \frac{n_{\text{SU}}}{\tau_H}, \quad (3)$$

where $1/\tau_M$ and $1/\tau_H$ are the rates of spin-pumping processes $n_{\text{SU}} \rightarrow n_{\text{SP}}$, whereas the rate $1/\tau_{\text{rel}}$ describes the spin relaxation $n_{\text{SP}} \rightarrow n_{\text{SU}}$ into spin-unpolarized states. The spin-pumping rate $1/\tau_M$ is set by spin-dependent scattering on the aligned magnetic moments M , and $1/\tau_H$ describes the rate of the spin-polarization processes caused by the external magnetic field. The magnetic field alignment rate in the lowest order is proportional to H , $1/\tau_H = \varepsilon H$, where ε is the rate per magnetic field unit. The process of the spin alignment is described by the damping term of the Landau-Lifshitz equation [11], which is linearly proportional to the magnetic field. Recent rigorous calculation of the conversion rate [23] led to the same result.

The expression for spin polarization $P_S(H)$ follows from Eqs. (2) and (3) and balance condition $\partial n_{\text{SP}}/\partial t = 0$,

$$P_S(H) = \frac{P_s^{(0)} + (H/H_S)}{1 + (H/H_S)}, \quad (4)$$

where the spin polarization in the absence of the external magnetic field is $P_s^{(0)} = \tau_{\text{rel}}/(\tau_{\text{rel}} + \tau_M)$ and H_S is the scaling relaxation magnetic field $H_S = 1/(\varepsilon\tau_{\text{rel}})$, which is determined by the relation between the spin-depolarization relaxation rate $1/\tau_{\text{rel}}$ and the magnetic field alignment rate per field unit ε [24]. Spin polarization $P_S(H)$ is a monotonic function growing from $P_S(H=0) = P_s^{(0)} < 1$ to $P_S(H \rightarrow \infty) = 1$. The values $P_s^{(0)} = 0$ and $P_s^{(0)} = 1$ in a ferromagnet are unphysical because its required for the spin relaxation to be either infinitely fast, $\tau_{\text{rel}} \rightarrow 0$, or infinitely slow, $\tau_{\text{rel}} \rightarrow \infty$.

Experiment. The Hall angle α_{HE} was evaluated from the measured Hall voltage using the standard relation [25]. The Hall voltage is measured by the ‘‘Hall bar’’ measurement setup shown in Fig. 2.

We measured around 100 of the FeB and FeCoB samples. In the following we show the data of a specimen representing plenty of samples showing qualitatively, and even quantitatively, the same phenomenon (see Supplemental Material [26] for the ranges of parameters of the large number of studied samples).

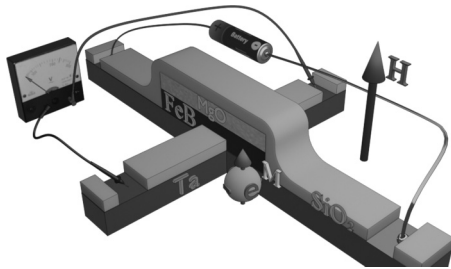


FIG. 2. Measurement setup. An FeB (1 nm) nanomagnet was fabricated on top of a Ta (3 nm) nonmagnetic nanowire. Nanomagnet size is 200 nm \times 200 nm. The Hall voltage was measured by a pair of Hall probes aligned to the nanomagnet position. An external magnetic field H is applied along the magnetization direction and perpendicularly to the nanomagnet surface.

The measured longitudinal conductivity σ_{xx} is between 0.03 and 0.06 S/m². The magnetoresistance $\leq 0.03\%$ is negligible, hence the whole change of α_{HE} can be attributed to σ_{xy} . FeB and FeCoB are standard well-studied materials [27–32] for magnetic random access memory. The nanomagnet thickness is in the range 0.8–1.3 nm because only in this range is the magnetization perpendicular to the surface, which is crucial for stability of the magnetic moment. The nanomagnet size varied from 50 \times 50 nm to 3 \times 3 μm . The condition of field independence of localized magnetic moments requires the use of only monodomain nanomagnets, whose monodomain nature we confirmed by a detailed study of hysteresis, magnetization dependence of the in-plane component on the in-plane magnetic field, and measurements on the nucleation domain sizes [33] (see Supplemental Material [26]). The AC results gave the same values as the DC ones but had smaller noise. All terms in (1) reverse their sign, when M and H are reversed. In order to avoid a systematic error due to a possible misalignment of the Hall probe, the Hall angle was measured as $\alpha = [\alpha(H, M) - \alpha(-H, -M)]/2$. Additionally, symmetry requirements allow one to exclude all linear $\sim H$ corrections to the prototypical OHE and AHE contributions.

Comparison with experiment. The measured variation of $\alpha(H)$ vs H can be explained in terms of the relation (1) depending on five fit parameters, namely, α_{OHE} , α_{AHE} , α_{ISHE} , H_S , and $P_s^{(0)}$. Figure 3 shows that the fit of the experimental data with the above relation is excellent proving the importance of the ISHE contribution to the HE in a ferromagnetic metal. Although, as shown below, the parameters used to obtain fit in Fig. 3 are ambiguous, the nonlinearity of experimental data requires that coefficient $\alpha_{\text{ISHE}} \neq 0$ indicating the inevitable presence of the ISHE contribution.

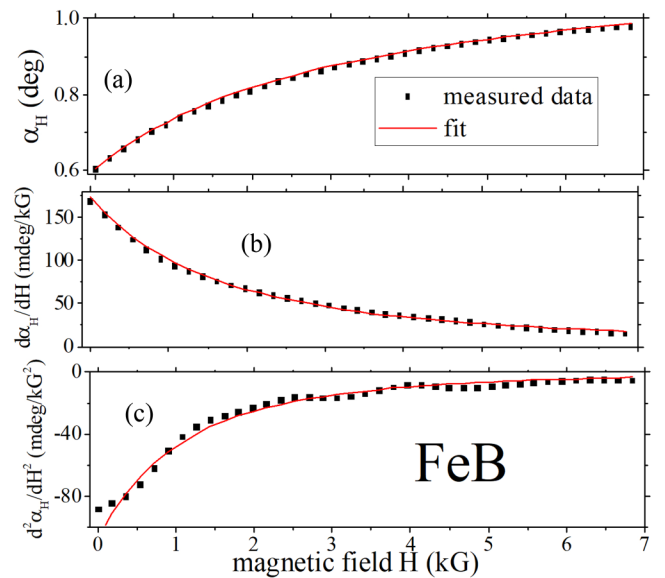


FIG. 3. Comparison of experimental data (circles) and theoretical fit (lines) for FeB: (a) Hall angle α_{Hall} as a function of the applied magnetic field H ; (b) first derivative, and (c) second derivative of α_{Hall} . The fit parameters are $H_S = 3.13$ kG, $P_s^{(0)} = 0.2$, $\alpha_{\text{ISHE}} = 664$ mdeg, $\alpha_{\text{AHE}} = 490$ mdeg, and $\alpha_{\text{OHE}} = 0.2$ mdeg/kG.

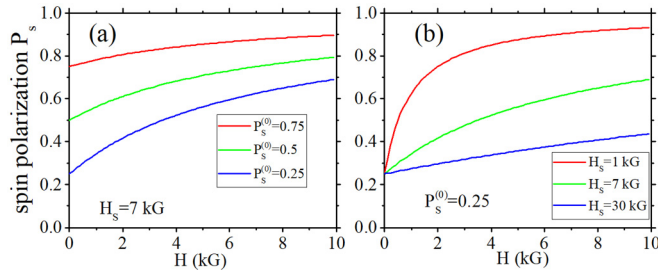


FIG. 4. Spin polarization P_s vs external magnetic field H : (a) for different zero-field spin polarization $P_s^{(0)}$ and fixed $H_s = 7$ kG and (b) for different scaling relaxation magnetic fields H_s and fixed zero-field spin polarization $P_s^{(0)} = 0.25$. We note that every set of parameters here gives absolutely the same theoretical curve for description of the experimental quantities shown in Fig. 3.

Some of the fitting parameters are ambiguous because the functional dependence (1) does not change (see Supplemental Material [26]) when the set of three initial fitting parameters $\{\alpha_{\text{AHE}}, \alpha_{\text{ISHE}}, P_s^{(0)}\}$ changes to a new set $\{\alpha'_{\text{AHE}}, \alpha'_{\text{ISHE}}, P_s^{(0')}\}$, which is related to the initial set as

$$\alpha'_{\text{ISHE}} = \alpha_{\text{ISHE}}(1 - P_s^{(0)})P_s^{(0')}/(1 - P_s^{(0')})/P_s^{(0)}, \quad (5)$$

and

$$\alpha'_{\text{AHE}} = \alpha_{\text{AHE}} + \alpha_{\text{ISHE}}(P_s^{(0)} - P_s^{(0')})/P_s^{(0)}(1 - P_s^{(0')}). \quad (6)$$

For FeB, with unambiguously determined parameters $H_s = 3.13$ kG and $\alpha_{\text{OHE}} = 0.2$ mdeg/kG one obtains an identical fit for $P_s^{(0)} = 0.4/0.3/0.2/0.1$ at $\alpha_{\text{AHE}} = 268/395/490/563$ mdeg and $\alpha_{\text{ISHE}} = 886/759/664/590$ mdeg. Note, the decrease of α_{AHE} is correlated with the increase of α_{ISHE} . A similar set of possible parameters can be obtained in $\text{Fe}_{0.4}\text{Co}_{0.4}\text{B}_{0.2}$: $H_s = 6.18$ kG, $\alpha_{\text{OHE}} = 0.2$ mdeg/kG, $P_s^{(0)} = 0.5/0.4/0.3/0.2$, $\alpha_{\text{AHE}} = 657/788/851/951$ mdeg, and $\alpha_{\text{ISHE}} = 783/653/560/490$ mdeg.

The important fact is that the measured Hall angle α_{HE} and consequently the evaluated spin polarization $P_s(H)$ is nonlinear for any value of $P_s^{(0)}$ [Fig. 4(a)] provided $P_s^{(0)} \neq 0$ and $P_s^{(0)} \neq 1$. Therefore, nonlinear contribution from ISHE is a robust feature which is not influenced by the ambiguity of the fitting parameters. The parameter, which defines the nonlinear dependence, is the scaling relaxation magnetic field H_s . When the H_s becomes larger, the ISHE contribution becomes close to linear i.e. for $H_s \geq 30$ kG [Fig. 4(b)]. Since the nonlinearity is the key distinguished feature of the ISHE, it is difficult to distinguish experimentally the nonlinear ISHE contribution from the linear OHE contribution in the case of a large H_s . The H_s is large when the spin relaxation is fast. Since in the studied sample $H_s = 3.13$ kG $\ll 30$ kG, the ISHE contribution can be easily distinguished (see Fig. 3). Note, regardless of the ambiguity of the fitting constants, the experimental first derivative of $\alpha_{\text{HE}}(H)$ in Fig. 3 is not constant and the second derivative

is nonzero. This implies that the nonlinear theoretical function $P_s(H)$, which describes the ISHE, has a nonzero nonlinear contribution to the full Hall angle (1).

We verified the stability of the obtained results and proved that the magnetic moment of the studied nanomagnets is in the single-domain state and is not realigned by comparison with the measurement of HE in multiple devices and at multiple current densities (see Supplemental Material [26]). We note that the first and second derivatives remain nearly the same, indicating that the OHE and ISHE contributions are nearly the same in different samples and their temperature dependence is weak. This nearly identical dependence of derivatives in different nanomagnets excludes the possibility of the existence of any magnetic domains, because the movement of the domain wall should be individual and different for each nanomagnet due to different distributions of fabrication defects, edge irregularities, and a slight difference in shape of the nanomagnets.

It should be noted that the firm alignment of local magnetic moments along the magnetic field is the critical requirement for the described measurements. This requirement makes it difficult to perform a similar measurement in an antiferromagnetic, compensated ferromagnetic or paramagnetic material. Additionally, it requires a measurement of the second derivative of α_{HE} with a reasonably high signal-to-noise ratio. It is only possible in a material having a low α_{OHE} and a long spin-relaxation time. The amorphous FeB and FeCoB are well fit to all these requirements.

We measured the Hall effect in a ferromagnetic nanomagnet with a strong perpendicular magnetic anisotropy, in which the local magnetic moments are very stable and well aligned along the easy axis of the nanomagnet. The magnetization direction does not change under an external magnetic field applied along the magnetization. The adopted expectation is that the Hall angle in a such nanomagnet must be a sum of a linear term proportional to the external magnetic field, which is the contribution from the ordinary Hall effect, and a field-independent constant, which is the contribution of the anomalous Hall effect in a sample with field-independent local moments. Our measurement revealed an extra nonlinear contribution indicating an additional contribution to the Hall effect. We interpreted this contribution as the inverse spin Hall effect, which is originated from the spin-imbalanced scatterings of the spin-polarized conduction electrons, and developed a phenomenological theory describing the spin polarization of conduction electrons of a ferromagnet in an external magnetic field. The theoretical expression of our phenomenological approach is in perfect agreement with the experimentally obtained nonlinear contribution giving, thus, a proof of the importance of the inverse spin Hall effect in a ferromagnetic metal.

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- [26] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.102.100404> for details of (i) theoretical fit ambiguity, (ii) experimental proof of single-domain nature of nanomagnets, and (iii) fabrication details and characterization of specimens.
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