Magnetic-field enhancement of nonlocal spin signal in $Ni_{80}Fe_{20}$/Ag lateral spin valves

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We observe a magnetic-field-induced enhancement of the nonlocal spin signal in $Ni_{80}Fe_{20}$/Ag lateral spin valves. The enhancement depends on the bias current polarity but not on the field direction. We present a theoretical model that explains our experimental results, taking into account the electron-spin relaxation of magnetic impurities. We find that the relaxation is about an order of magnitude weaker than Elliott-Yafet relaxation.

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Electronic transport in metals in the presence of magnetic impurities (MIs) has fascinated physicists for decades due to its rich physics.[1–3] MIs that affect the resistivity of metals via the Kondo effect[4] are responsible for saturation of the electron phase-coherence time in mesoscopic metal wires at low temperature[5] and for enhanced critical currents in superconducting nanowires exposed to external magnetic fields.[6] However, the effect of MIs on spin transport is an area of research[7] where unexpected phenomena continue to provide surprises. For example, for the recently observed giant magnetothermopower in Cu embedded with Co nanoclusters,[8] the origin of the electron-hole asymmetry may rest in the electron spin-flip scattering from polarized Co magnetic moments.[9] In addition, a giant spin Hall effect reported for Au[10] has been attributed to an orbital-dependent Kondo effect of Fe impurities,[11] although both the experimental results and its theoretical explanation are controversial.[12–14]

Lateral spin valves (LSVs) provide a preferred geometry for studying pure spin transport; however, there have been no reports to date on the effect of MIs on their nonlocal magnetotransport. Herein we show that MIs, likely residing at the interface of ferromagnetic and normal metal junctions of LSVs, give rise to an enhancement of the nonlocal (NL) spin signals when the LSVs are exposed to an external magnetic field. The observed field dependence is physically different from that arising from magnetization switching of ferromagnetic injector and/or detector electrodes[15–17] or from spin precession and dephasing (Hanle effect).[18,19] In addition to the experimental results, we also present a theoretical model that relates the NL spin signals to spin-relaxation rates in the presence of MIs.

Three LSV devices used in this study were fabricated on a SiN (100 nm)/Si substrate by *e*-beam lithography and shadow mask *e*-beam evaporation. They consist of two 25-nm-thick $Ni_{80}Fe_{20}$ (permalloy, Py) electrodes bridged by an 80-nm-thick Ag wire, while their widths range from ∼110–140 nm for the Py injector to ∼70–95 nm for the Py detector and ∼110–260 nm for the Ag wire. The measurements are carried out in the NL geometry, as shown in Fig. 1(a), where charge current and the measured voltage are taken apart from each other to minimize spurious signals arising from charge transport.[20] Unless otherwise noted, the measurements were carried out using a dc bias reversal technique with the bias current magnitude $I = 0.5$ mA. The NL resistance $R_{NL} = V/I$ depends on spin accumulation and diffusion in Ag and changes in response to external magnetic field B. For a parallel field $B_\parallel$, R_{NL} changes sign when the Py electrodes switch their magnetization orientation $\mathbf{M}$, giving rise to a NL spin-valve signal $\Delta R_s = R_{NL}^{\uparrow\uparrow} - R_{NL}^{\downarrow\uparrow}$ [see Fig. 1(b)]. When the field is oriented perpendicular, $B_\perp$, spin precession gives rise to characteristic oscillations of R_{NL} damped by the spin-flip-induced dephasing of polarization and its reduction due to tilting of $\mathbf{M}$ out of plane in the direction of $B_\perp$ [see Fig. 1(c)]. In both cases, when Py electrodes are oriented parallel to each other and to B, no further dependence on B is expected to occur due to the absence of spin dynamics and constant $\mathbf{M}$. However, we observed that R_{NL} monotonically increases, independent of the direction of B.

Figure 1(c) shows R_{NL} vs $B_\perp$ for initial parallel $\mathbf{M}$. Due to spin precession the R_{NL} value of 2.9 mΩ at 0 T initially decreases with increasing $B_\perp$. This behavior is reversed at ∼0.3 T when R_{NL} reduction due to spin precession and dephasing is overcome by an increasing contribution of spins parallel to $\mathbf{M}$ due to tilting of $\mathbf{M}$ in the direction of $B_\perp$.[19] While the latter is expected to restore R_{NL} to its initial zero-field value, we found that at about 0.6 T R_{NL} becomes larger than at 0 T, continues to increase with increasing $|B_\perp|$, and reaches ∼3.3 mΩ at 2 T. Thus the additional NL spin signal δR_{NL} at $|B_\perp| = 2$ T amounts to more than 12% of the initial R_{NL} value.

While nonzero δR_{NL} was initially observed for $B_\perp$, we found that it also appears in $B_\parallel$. Since spin precession is absent for this case, when magnetization of both injector and detector electrodes is parallel to the applied field, we can define $\delta R_{NL}(B) = R_{NL}(B_\parallel) - R_{NL}(0)$. Figure 2 shows representative plots obtained at 40 K on the same sample, the only difference being the orientation of the external field in the measurements, i.e., parallel and perpendicular to the initial $\mathbf{M}$. Note that in this measurement the field range is much larger than in Fig. 1(b): consequently, the common $\mathbf{M}$ of the injector and detector is opposite for positive and negative fields. One can see that the initial and the final values of R_{NL}

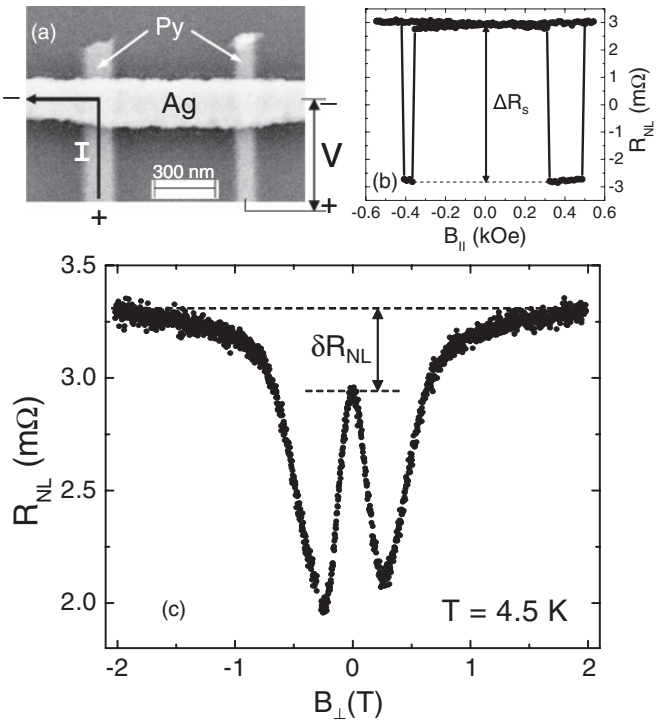

FIG. 1. (a) A scanning electron microscopy image of a Py/Ag LSV device adapted to show the nonlocal measurement configuration for a positive bias current polarity. (b) R_{NL} vs $B_{\|}$ at 4.4 K and the corresponding ΔR_s signal. (c) R_{NL} vs $B_{\perp}$ measured on the same sample at 4.5 K. δR_{NL} at $B \sim 2$ T is marked by an arrow.

for the two measurements are identical. Furthermore, the data practically overlap above 1.2 T when Py electrodes are fully saturated in the direction of B. This behavior was observed in all measurements. Thus the magnitude of $\delta R_{NL}(B)$ does not depend on the direction of the external field once the Py electrodes are saturated in this direction. This finding suggests that δR_{NL} does not arise from an orbital magnetoresistance effect since the latter would depend on the field orientation. Note also that the orbital magnetoresistance in a normal metal such as Ag is expected to decrease, rather than increase, R_{NL} since it reduces the mean free path and, consequently, the

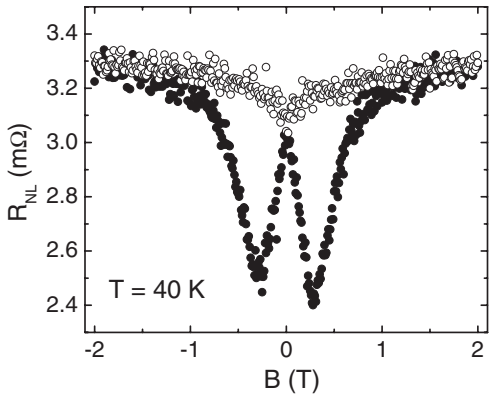

FIG. 2. (a) Magnetic-field dependence of R_{NL} measured at 40 K for $B_{\perp}$ (solid circles) and $B_{\|}$ (open circles) to the initial magnetization orientation of Py electrodes.

spin diffusion length in Ag. Therefore, $\delta R_{NL}(B)$ must have a different physical origin.

In order to elucidate the physics we studied $\delta R_{NL}(B)$ as a function of the temperature and the polarity of the LSV bias current I. Figures 3(a)–3(d) show $\delta R_{NL}(B)$ measured on a Py/Ag LSV for $B_{\|}$ between $+6$ and -6 T. Here the "dc" measurements were carried out using zero-bias current subtraction and $I = 0.3$ mA. The latter technique is less efficient in eliminating the magnetic noise voltage fluctuations, apparently resulting in noisier $\delta R_{NL}(B)$ values compared to Figs. 1 and 2. One can see that δR_{NL} vanishes for negative I (spins injected from Py into Ag), while finite δR_{NL} is observed only for positive I (spins extracted from Ag into Py). This strongly suggests that the physical mechanism responsible for the appearance of δR_{NL} depends on the orientation of the spin polarization in Ag since this parameter is changed when the polarity of I is switched.[19]

Since precession-related phenomena such as those combined with spin-orbit effects tend to suppress spin polarization rather than to enhance it,[21] we microscopically base our interpretation on spin-flip events due to unintentional MIs. For positive current, these should be randomly magnetized in zero field, thus providing an additional spin-relaxation mechanism whose strength is continuously reduced with increasing B. On the other hand, the experimentally observed absence of the field dependence for negative current suggests that, in this case, polarization of the impurities is already frozen at $B = 0$ in the direction of B to be applied (since increasing the value of B has no effect on electron-spin relaxation). The two situations can be realized in a system in which the magnetization of the MIs at $B = 0$ is determined by the competition between the coupling to the magnetization orientation of the FM electrodes and the polarization of the injected spins. For antiparallel orientation of the magnetization of the FM electrodes and the spin polarization in Ag (positive bias, electrons flowing from Ag to Py), just as is the case when a nonzero $\delta R_{NL}(B)$ is observed in Fig. 3, the two may oppose each other, resulting in a randomized impurity magnetization. This gives rise to higher spin scattering and the reduced nonlocal spin signal at zero field that is gradually recovered via application of an external field. In the opposite case, the spin-polarized current enhances the saturation of the impurities, effectively freezing them out, and increasing the value of B then has no further effect. Our previous work[22] also suggests that impurities in the form of ferromagnetic oxide nanoclusters are present at the Py/Ag interface in the junction region, thus making this assumption empirically plausible.

To model the experimental $\delta R_{NL}(B)$ data based on this assumption, we first write down an expression for $1/\tau_{sf}$, the spin-flip rate at the Py/Ag interface, which includes a microscopically constructed spin-flip rate due to the MIs $1/\tau_{mi}$, and then employ a resistor network model in order to connect the spin-flip resistance $R_{sf} \propto \tau_{sf}(B)$ to R_{NL}. Regarding the first step, the Fermi golden rule predicts the following spin-flip scattering rate between $s_z = \pm 1/2$ electrons on randomly magnetized impurities of density n[9]

$$\frac{1}{\tau_{mi}} = \frac{2\pi n J^2 S^2 D}{\hbar}\left(1 - \left\langle \frac{S_z^2}{S^2} \right\rangle\right). \quad (1)$$

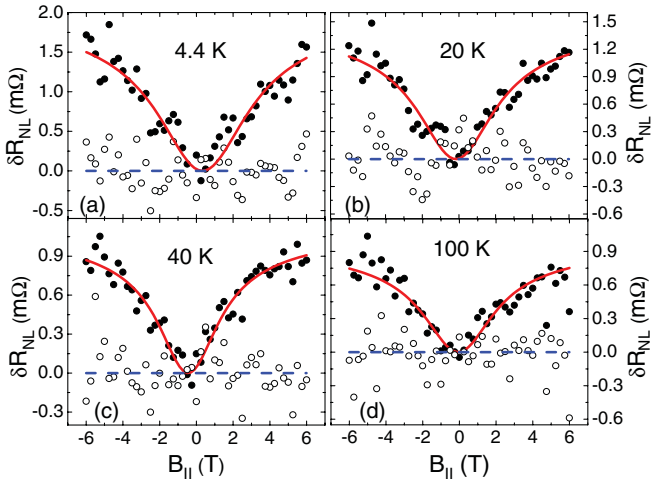

FIG. 3. (Color online) (a)–(d) δR_{NL} vs $B_{\parallel}$ for positive (solid circles) and negative (open circles) polarity of the LSV bias current. The fits to the data based on Eq. (4) are shown as solid red (positive bias) and dashed blue (negative bias) lines. The measurement temperatures are indicated in the graphs.

In this expression,[24] $\langle S_z^2/S^2\rangle$ is the ensemble thermal average of impurity magnetization along the z axis normalized to the impurity spin $S \gg 1$. Physically, Eq. (1) stems from the Schrieffer-Wolff-transformed Hamiltonian[23] that couples with strength J a single Anderson impurity to conduction electrons whose density of states is D. We also consider a parallel mechanism, such as Elliott-Yafet (EY),[25] to contribute to the spin-flip rate as $1/\tau_{ey}$. The total rate is then $(\tau_{sf})^{-1} = (\tau_{mi})^{-1} + (\tau_{ey})^{-1}$ and

$$\frac{\tau_{sf}(B)}{\tau_{sf}(B=0)} = \frac{a+\frac{2}{3}}{a+1-\left\langle\frac{S_z^2}{S^2}\right\rangle}. \tag{2}$$

The nonzero ratio $a = (\tau_{ey})^{-1}/(2\pi n J^2 S^2 D/\hbar)$ between the EY and MI scattering rates at zero field is a necessary assumption for our model to reproduce the experimentally observed $\delta R_{NL}(B)$.

In order to derive an expression that relates R_{sf} to R_{NL}, we employed a resistor network representation of a LSV[26,27] and adapted the network used by Jedema *et al.*[29] to our device structure. Interface scattering is allowed to cause spin flips.[28] In this representation $R_{sf} = \tau_{sf}(2D/e^2)$ (Ref. 29), while $R_{NL} = \Delta\mu/eI$ can be obtained from the potential difference $\Delta\mu$ between the device output terminals and the known driving current I. If MI scattering contributes only a small part to the total τ_{sf}, an assumption verified below, we can write

$$\frac{\delta R_{NL}(B)}{R_{NL}(0)} \approx \alpha_R\left[\frac{R_{sf}(B)}{R_{sf}(0)} - 1\right]. \tag{3}$$

Our resistor network is mathematically equivalent to a system of seven linear equations whose parameters include current polarization in Py, spin-diffusion lengths, conductivities, and ohmic resistances at zero field for each Ag/Py interface in both spin channels.[30–32] Effectively, the complete resistor network projects into the single dimensionless constant α_R whose value we find to be in the range between 4 and 6. Following the proportionality relation between R_{sf} and τ_{sf} and Eq. (2) and assuming a Langevin-like field-dependent polarization $\langle S_z/S\rangle$ of the MIs, we finally obtain

$$\delta R_{NL}(B) = \alpha_R R_{NL}(0)\left[\frac{a+\frac{2}{3}}{a+2\frac{L(\beta B)}{\beta B}} - 1\right], \tag{4}$$

where $L(x) = \coth(x) - 1/x$. This expression can be used to fit the measured $\delta R_{NL}(B)$ with two fitting parameters, a and β, if the value of α_R is known. Also, recall that $a \to \infty$ corresponds to purely EY spin relaxation, while $a = 0$ stands for the MIs acting alone.

The fits to $\delta R_{NL}(B)$ using Eq. (4) are shown in Figs. 3(a)–3(d). Here we allowed for a small offset in B, which could be due to a random trapped flux field in the superconducting magnet and/or to a current-induced magnetization of the impurities. All fits for negative bias (dashed blue lines) result in a constant $\delta R_{NL}(B) = 0$, i.e., an absence of impurity-induced field dependence of spin relaxation, as expected. For positive bias, the fitting curves (solid red lines) appear to be in good qualitative agreement with the data. If the impurities behave paramagnetically, $\beta = m/k_B T$ would depend on temperature (m is the magnetic moment of the impurity, and k_B is the Boltzmann constant). However, instead of following this T dependence, the inferred β values in Fig. 4(b) seem randomly distributed around the average value $(1.65 \pm 0.36)\ \mathrm{T}^{-1}$. Such behavior and value of β can correspond to shape anisotropy of nanometer-sized MIs with random form and orientation. We identify this mechanism to be responsible for the appearance of $\delta R_{NL}(B)$. Note that the parameter β mainly determines the shape of the $\delta R_{NL}(B)$ curve and is almost independent of the value of α_R used in fitting.

The latter, however, is not the case for parameter a, whose values extracted from fitting depend on the values of α_R. In the case of our samples, R_i and its T dependence are not well known;[22] thus precise α_R values cannot be determined. However, assuming T-independent $\alpha_R = 4$, which is a good order of magnitude estimate, we obtain a values in the range of ~8 to ~27 [see Fig. 4(a)]. This suggests that impurity spin-flip scattering is, indeed, weaker than the B-independent EY mechanism.

In conclusion, we observe enhancement of the nonlocal spin-valve signals in NiFe/Ag lateral spin valves exposed to external magnetic fields. The enhancement occurs only for the positive polarity of the bias current and is absent in the

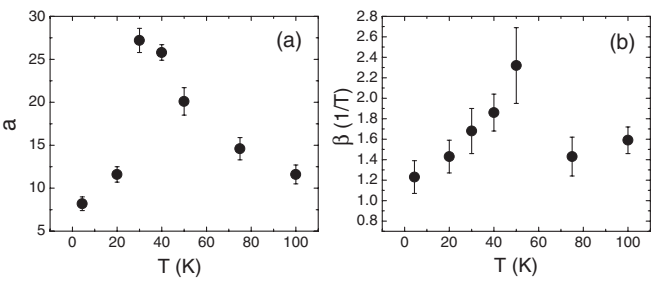

FIG. 4. T dependencies of (a) parameter a and (b) parameter β extracted from fitting $\delta R_{NL}(B)$ using Eq. (4). The value of $\alpha_R = 4$ was assumed in fitting.

opposite case. When the magnetization direction of the Py electrodes is aligned with the field, the enhancement does not depend on the field orientation. We presented a theoretical model that considers spin flip scattering on magnetic impurities in addition to the conventional field-independent Elliott-Yafet spin-relaxation mechanism and found agreement between the derived equations and the experimental data. By studying how the nonlocal signals change with magnetic field and temperature we conclude that scattering on magnetic impurities in our samples is not a markedly temperature-dependent mechanism (such as due to isotropic paramagnetic impurities), while the Elliott-Yafet mechanism still dominates the total spin relaxation.

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