NANO LETTERS

2007
Vol. 7, No. 4
927−931

Surface Spin-Valve Effect

I. K. Yanson,[†] Yu. G. Naidyuk,[†,‡] V. V. Fisun,[†,‡] A. Konovalenko,[‡] O. P. Balkashin,[†] L. Yu. Triputen,[†] and V. Korenivski*[,‡]

B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, 47 Lenin Avenue, 61103 Kharkiv, Ukraine, and Nanostructure Physics, Royal Institute of Technology, SE-10691 Stockholm, Sweden

Received December 2, 2006; Revised Manuscript Received February 10, 2007

ABSTRACT

We report an observation of spin-valve-like hysteresis within a few atomic layers at a ferromagnetic interface. We use phonon spectroscopy of nanometer-sized point contacts as an in situ probe to study the mechanism of the effect. Distinctive energy phonon peaks for contacts with dissimilar nonmagnetic outer electrodes allow localizing the observed spin switching to the top or bottom interfaces for nanometer thin ferromagnetic layers. The mechanism consistent with our data is energetically distinct atomically thin surface spin layers that can form current- or field-driven *surface spin-valves* within a *single* ferromagnetic film.

Spin-valves in the current perpendicular to the plane geometry are usually nanopillars having two ferromagnets (F1 and F2) of different anisotropy, such that one is magnetically hard and the other is magnetically soft separated by a nonmagnetic spacer layer (N). The conductivity of such a spin-valve is governed by the giant magnetoresistance effect[1,2] and depends on the mutual orientation of the magnetization in F1 and F2.[3] Recent studies have shown[4−7] that nonmagnetic metal contacts to single ferromagnetic films (N−F) exhibit spin torque effects similar to those observed in F1−N−F2 spin-valves. For single N−F interfaces, non-hysteretic singularities of magnetic origin observed in the conductance are explained as arising from spin wave excitations in the ferromagnetic film.[8,9] Similar to the nanopillar case,[3] these peaks are observed only for one polarity of the bias current, namely for the electron current flowing from N into F, and their position on the current/voltage axis is proportional to the magnitude of the external magnetic field.[5−7] Another pronounced and rather unexpected feature of N−F nanocontacts, being the trade mark of the F1−N−F2 spin-valves, is hysteresis in resistance versus voltage, resulting in a bistable resistance state near zero bias. Its origin is under debate, with proposed interpretations ranging from surface exchange anisotropy[10] and magnetoelastic anisotropy[11] to spin vortex states.[12] In this work, we investigate the mechanism of the hysteretic conductance in magnetic point contacts (PC's) by combining PC phonon spectroscopy[13] and magnetoconductance on the scale down to ∼1 nm in the contact radius, which is inaccessible by today's lithographic means. We focus in particular on measuring thin films, where the magnetic film thickness (t) can be smaller than the PC diameter (d) and comparable or smaller than the ferromagnetic exchange length in the material. Such experimental configuration prevents formation of volume domains on the scale of the contact. Furthermore, we investigate the regime of dimensional cross over in t vs d, the latter scale defining the contact core where the current density is maximum. In the limit $t < d$, both magnetic interfaces can be inside the high current density region and therefore actively contribute to spin transport, whereas for $t > d$, only one interface is expected to contribute to magnetoconductance. Such nanometer-scale probing into the ferromagnetic surface, with an in situ spectroscopic detection of the location of the nanocontact core, leads us to conclude that the observed magnetic effects are due to atomically thin surface spin layers acting as current- or field-driven spin-valves with respect to the magnetization in the interior of the ferromagnetic layer.

Our samples are Co films deposited onto oxidized Si substrates buffered with a nonmagnetic bottom electrode (N2 = Cu, Au) of 30−100 nm in thickness. The thickness of the Co films (F) ranged between 2 and 100 nm. For several samples, the Co layer was capped with a 3 nm Au layer in order to prevent oxidation of the Co surface. The other nonmagnetic electrode (N1 = Cu, Ag, Au, W, Mo) is prepared in the form of a sharp tip, mechanically manipulated at low temperature to gently touch the F surface. All measurements are done at 4.2 K, with the samples always cooled from room temperature in zero field. The magnetic field is applied parallel to the film plane. For each contact, a set of complementing transport characteristics were recorded: the differential resistance $R(V) \equiv dV/dI(V)$, magnetoresistance $R(H, V \approx 0)$, and the so-called PC spectrum,

* Corresponding author.
[†] B. Verkin Institute for Low Temperature Physics and Engineering.
[‡] Nanostructure Physics, Royal Institute of Technology.

10.1021/nl0628192 CCC: $37.00
Published on Web 03/03/2007

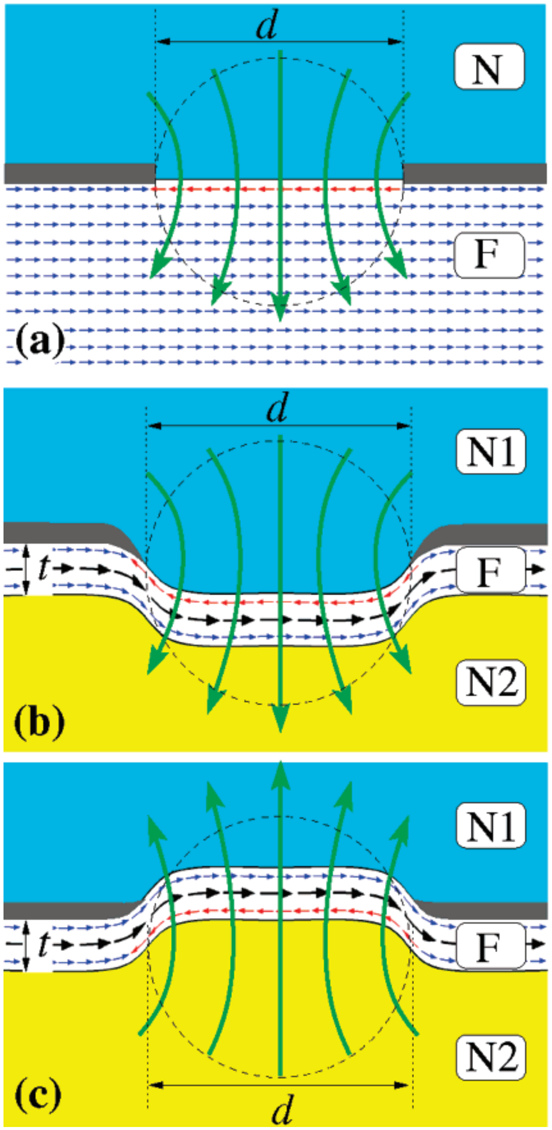

Figure 1. Schematic of three generic point contacts: (a) a nonmagnetic tip in contact with a bulk or thick film ferromagnet ($t \gg d$); a thin F layer ($t < d$) between two nonmagnetic metals N1 and N2, placed in the lower (b) or upper (c) part the contact core. The electrodes are separated by a nonconductive surface layer, shown in dark gray. Green arrows indicate the electron current flow through the PC core, which is schematically shown as a thin dashed circle. Small red arrows illustrate spin switching of the interface spins with respect to the interior spins (black arrows) by the high density current in the PC core. Notice that the polarity of the electron current in (b) and (c) are opposite, illustrating the cases of maximum current density and hence strongest spin torques at the top and bottom F surface, respectively.

$d^2V/dI^2(V)$, provided the contact was mechanically stable for a sufficient period of time. A great many shorter-lived contacts have been measured, resulting in a large library of still very informative subsets of magnetotransport data. The static resistance measured at low bias showed essentially the same hysteretic behavior as the differential resistance. In the discussion to follow, we therefore make no distinction between the two and show the data for dV/dI.

Three generic geometries of a point contact are illustrated in Figure 1. Figure 1a shows a schematic view of a nonmagnetic metal (N) in contact with a bulk or a thick film ferromagnet. The two metals are separated electrically except for a small circular orifice of diameter *d*, the point contact size. This model corresponds to the experimental configuration studied previously.[5,7,10] In this case, only the upper interface between N and F is located in the region of high current density, hence only this interface plays a role in magnetotransport.

Figure 1b illustrates the case where a thin F film is located in the lower half of the contact core (dashed circle). The upper F−N interface is located in the region of maximal current density and therefore contributes most to the spin torque effects. Notice that the contact core in this case is filled predominantly with N1 material, which therefore should dominate the PC spectrum. For a Co layer thinner than *d*, its contribution to the PC spectrum is expected to be small.[13] The Co phonon peaks are therefore not resolved in the PC spectra discussed below.

A third characteristic, albeit less probable geometry, is shown in Figure 1c. Here, because of a supposed protrusion on the surface of N2, the Co nanolayer is elevated to the upper half of the contact core filled predominantly by N2, which therefore is expected to dominate the phonon PC spectrum. Consequently, the maximal current density is found at the bottom interface of the Co film. Thus, using PC phonon spectroscopy, we can determine in situ the relative weight of the two N/F interfaces in the magnetotransport for a given contact.

Figure 2a illustrates the *normal* hysteresis for a heterocontact Cu100/Co3-Au, a 100 nm Cu buffer electrode covered with a 3 nm thick Co film in contact with an Au tip. The higher resistance state is obtained for negative bias where the electrons flow from N1 to F. Cycling through a large positive bias (red curve), in this case approximately +40 mV, switches the contact into the low resistance state at about +10 mV. This state is preserved down to about −40 mV, followed by a smeared upward transition. Assuming the hysteresis is caused by a domain wall of type F1−DW−F2 within the Co layer, the large negative bias produces an antiparallel (AP, or at least a large relative angle) configuration of the F1 and F2 magnetic sublayers, while for the large positive bias, the spins in the two sublayers are parallel (P). The subsequently recorded PC spectrum shown in Figure 2b, which is the measure of the electron−phonon interaction in the contact,[13] shows that the contact core is occupied predominantly by Au (N1). This means that the Co layer (whose phonon spectral lines are not detected owing to its small thickness) is located in the lower half of the PC core and, therefore, it is the upper N/F interface that plays the main role in the spin torque driven hysteresis.

The typical resistance of our contacts is ~10 Ω, which correspond to the Sharvin diameter $d \simeq 10$ nm. This is estimated as follows (see Figure 3.9 and the accompanying text in ref 13 for additional details):

$$R_{\text{Sharvin}} = 16\rho l/3\pi d^2$$

which using the free electron approximation and the Fermi

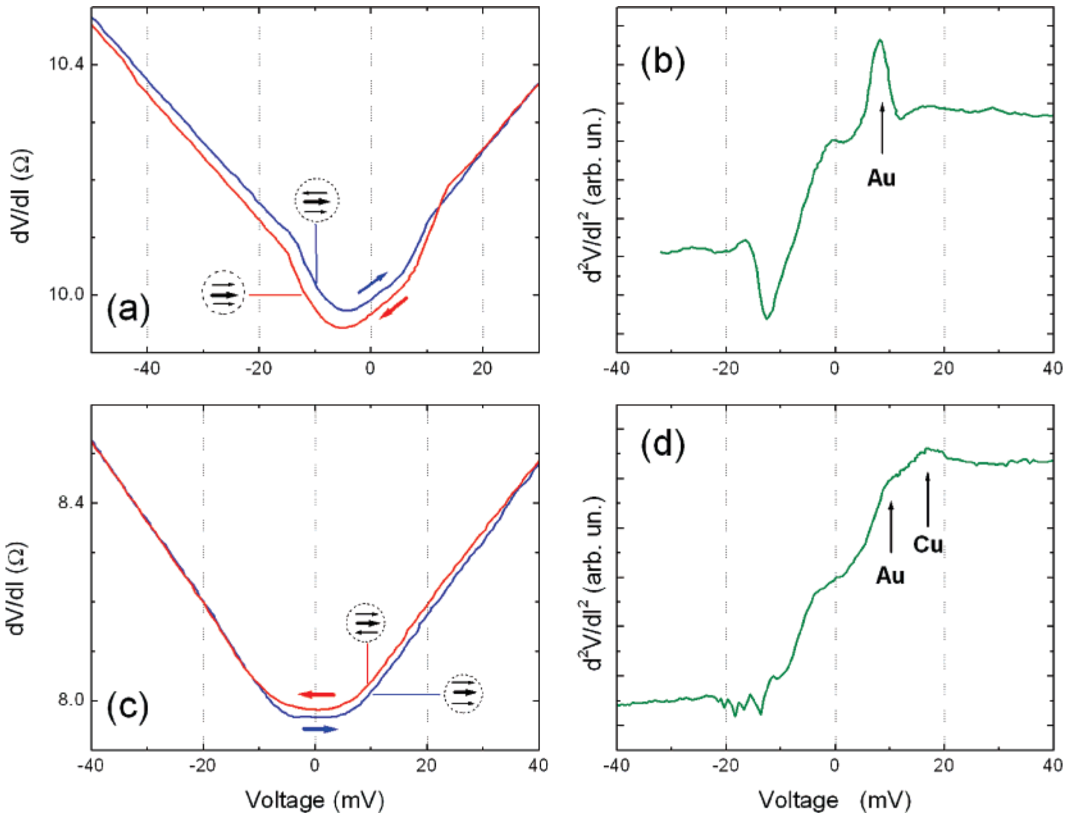

Figure 2. (a) *Normal* hysteresis in dV/dI(V) for a Cu100/Co3-Au contact; (b) PC spectrum for the same contact showing a pronounced Au transverse phonon peak; (c) *anomalous* hysteresis in dV/dI(V) for a nominally similar contact (Cu100/Co3-Au), along with its PC spectrum showing a dominant Cu phonon maximum (d). The black arrows in dashed circles in (a,c) schematically indicate the orientation of the surface and interior spins. The red and blue arrows in (a,c) as well as in Figures 3 and 4 indicate the bias sweep direction.

momentum k_F(Cu, Au) $\simeq 1.35 \times 10^8$ cm^{-1} yields

$$d\,[\mathrm{nm}] \simeq 30/\sqrt{R[\Omega]}$$

This value somewhat underestimates the true contact diameter because the Sharvin formula applies in the ballistic current regime. For the contacts reported herein, the regime is closer to diffusive (the electron mean free path $l < d$), hence the typical contact diameter should be $\gtrsim$10 nm for $R \approx 10\ \Omega$. This means that, for our nanometer thin ferromagnetic layers, the condition $t < d$ is fulfilled for typical contacts, hence both ferromagnetic interfaces can be expected to contribute to magnetotransport, depending on the microscopic layout of the contact (see Figure 1b,c).

Figure 2c illustrates the case where the bottom interface is dominant, which leads to a reversal of the hysteresis in conductance. Such inverse or anomalous hysteresis is observed much less frequently owing to the fact that protrusive point contacts (see Figure 1c) are less probable micromechanically. The outer electrodes were chosen with an aim to separate their main phonon peaks, which in the case of Cu and Au are found at approximately 17 and 10 mV, respectively.[13] The separation of 7 mV is large enough to reliably distinguish the peaks even for nonballistic contacts with smeared phonon maxima.[7] The dominant Cu maximum in Figure 2d, in contrast to the spectrum of Figure 2b, indicates that it is the bottom Co/Cu interface region that plays the main role in the electron and spin transport for this nanocontact. The corresponding hysteresis is *anomalous*, i.e., the large resistance state corresponds to the electrons flowing from the film into the tip (Figure 2c). The spectroscopic data provides a natural explanation, namely the contact core of highest current density is at the bottom F/N interface where the electrons flow from the Cu bottom electrode into Co. The transition to the high resistance state occurs +35 mV and the reverse transition at −8 mV.

Figure 3 illustrates the normal (a) and inverse (c) hysteresis in $R(V)$ accompanied by a spin-valve-like hysteresis in $R(H)$ (b,d) for Cu30/Co5-Ag contacts. Here the switching AP-to-P and P-to-AP occur as sharp steps in resistance, occurring under the influence of a nominally unpolarized current (insets a and c) or an externally applied magnetic field. Importantly, the resistance difference between the stable at zero bias AP and P states is the same for the current- and field-induced transitions, even though the mechanisms leading to hysteresis are different in the two cases. For $R(V)$, the bistability is caused by spin transfer torques, while for $R(H)$ with the external magnetic field parallel to the film plane, the behavior is identical to the nanopillar spin-valve magnetoresistance. This observation is true even for $R(V)$ curves with smooth transitions such as Figure 2a,c; the difference in resistance versus current and that versus field are the same. Notice also the maxima in dV/dI at approximately −20 mV, which are due to nonhysteretic magnetization excitations.[5,7,8]

Another important observation, based on our measurements of about 100 contacts with hysteresis, is that statisti-

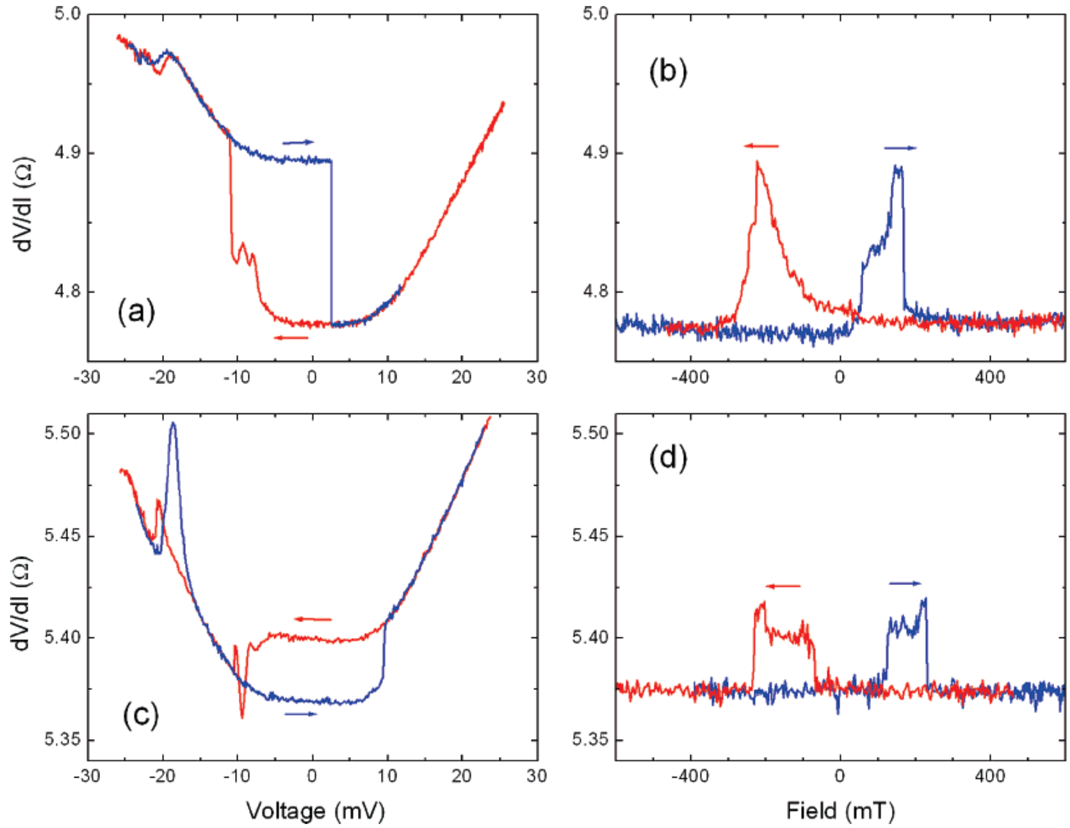

Figure 3. Current (a) and field (b) driven *normal* hysteresis for a Cu30/Co5-Ag contact; (c) and (d) same for a similar Cu30/Co5-Ag contact exhibiting *anomalous* hysteresis.

cally $\Delta R/R$ shows no trend as a function of the thickness of the Co layer in the range $t = 2-100$ nm, as illustrated in Figure 4a, strongly suggesting that the observed hysteresis is a surface effect. As shown in Figures 2, 3, and 4b, this surface effect can occur at either of the ferromagnetic interface and is indistinguishable in its current- and field-driven behavior from the standard three-layer spin-valve effect.

The above clear correlation between the dominant phonon maxima and the sign of hysteresis in the conductance of the heterocontacts is straightforward conceptually and reflects the limiting cases of the ferromagnetic layer being at the bottom or top of the contact core. Naturally, a richer behavior should be expected for very thin magnetic layers located symmetrically in the PC core. Namely, the spin torque effects from the two interfaces should superpose. An example of such behavior is shown in Figure 4b. Here, the polarity of the hysteresis near zero bias is *normal*, correlating well with the pronounced Au phonon maximum at $\approx$10 mV. At positive bias, expected to drive the contact into a low resistance state if one considers the top interface only, such a *normal* hysteresis transition indeed occurs at $\approx$15 mV. However, the presence of the bottom interface within the contact core, as evidenced by the Cu-phonon maximum in the PC spectrum, results in a superposed *anomalous* hysteresis at large positive bias. The spin structure of the ferromagnetic layer of only 2 nm in thickness consistent with these double-hysteresis data is a rather surprising double spin-valve of type $\downarrow\uparrow\uparrow$ or $\uparrow\uparrow\downarrow$. The data in Figures 2 and 3 with hysteresis of one type only would therefore represent the limiting cases where the PC conductance is dominated by either one of the two N/F interfaces.

The contact core, dominating the charge and spin current in PC's, approaches 1 nm scale for highly resistive contacts with $R \sim 100\ \Omega$. $R(V)$ for such a contact, exhibiting a pronounced current-driven hysteresis, is shown in Figure 4c for a 5 nm thick Co layer. We estimate the radius of this contact core to be $\approx$1.5 nm. Therefore the whole F1−DW−F2 magnetic sublayer structure producing the hysteretic switching must be limited in thickness to some 3−4 atomic unit cells at the interface, which provides a decisive evidence that the surface spin layer acting as the "free" layer is atomically thin. This consideration and the lateral extent of the contact of only $\approx$3 nm (to an exchange-length thin ferromagnetic film) disqualifies any interpretation of the phenomenon based on volumelike domains or vortex states. In particular, vortex states that are possible to produce in $\sim$100 nm ferromagnetic particles in nanopillars should be unstable for contacts to continuous films studied here, especially at zero bias, where circular Oersted fields are absent. We therefore rule out the vortex interpretation for our contacts, which are 1−2 orders of magnitude smaller than typical magnetic nanopillars.

The previously proposed interpretation[10] of the *normal* hysteresis as due to a surface exchange bias in naturally oxidized Co films is faced with difficulties as well. First of all, making a true metallic contact removes any oxide from the contact region. Furthermore, in contrast to ref 10, all of our measurements are done on samples cooled in zero field,

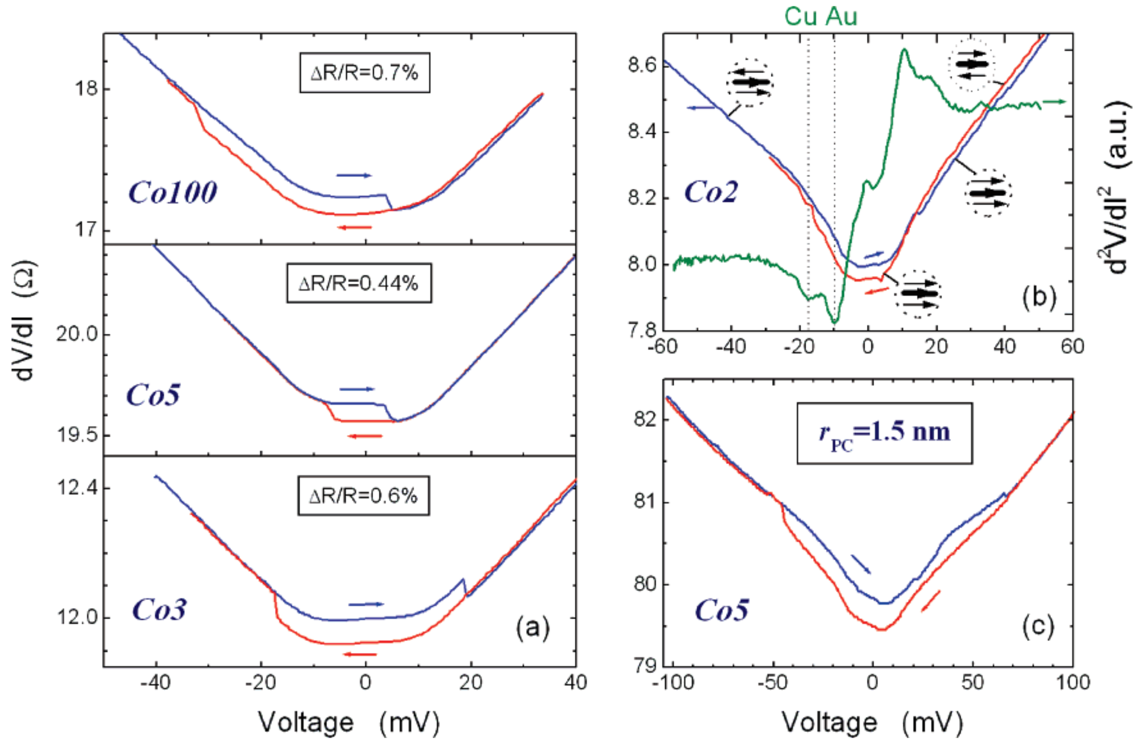

Figure 4. (a) Hysteresis in dV/dI(V) for contacts to Co films of varying thickness; (b) dV/dI(V) for a Cu100/Co2/Au3-Au contact showing a superposed hysteresis from the Au and Cu interfaces and the PC spectrum (green) exhibiting well-defined phonon maxima for both Au and Cu at ≈10 and ≈17 mV, respectively. The arrows in dashed circles schematically indicate the orientation of the surface and interior spins; (c) dV/dI(V) for a high R contact to a 5 nm Co film, $r_{PC} \approx 1.5$ nm.

which we find does not diminish the hysteresis effects. As shown in Figure 4b, hysteresis is also observed in contacts to ferromagnetic films capped with protective Au antioxidation layers (we have measured a number of such contacts). As shown in Figures 2, 3, and 4b, we also detect hysteresis due to the bottom ferromagnetic interface, which is produced in high vacuum and is certainly free from any oxide. Finally, surface exchange anisotropy anyway necessitates postulating a volumelike domain,[10] which can be excluded for our nanocontacts to nanometer thin films. We therefore can rule out this mechanism for the effects we observe. It is important to mention that we observe pronounced magnetic hysteresis effects on Permalloy ($Ni_{80}Fe_{20}$) films, which is additional evidence against interpretations based on surface exchange or stress-induced anisotropy.

Spins at ferromagnetic interfaces can have substantially different magnetic character and be weakly coupled to the interior spins, which finds support in the recent studies of surface and interface magnetism in Co and Fe.[14,15] The fundamental physics involved is that the interface spins have a lower coordination number and therefore fewer exchange bonds compared to the bulk spins. This can lead to a reconstruction of the interface spin order, resulting in quite different magnetic moment and anisotropy. The spin transport effect we observe is of quite general nature and should also be present in magnetic structures on a larger scale, provided that surface imperfections such as roughness do not lead to averaging out its contribution. With a proper control of the magnetic interface in nanodevices, this effect can offer a new way of manipulating the electron spin.

Acknowledgment. Financial support from the Swedish Foundation for Strategic Research (SSF), the Royal Swedish Academy of Sciences (KVA), and the National Academy of Sciences of Ukraine (NASU) under project NANO is gratefully acknowledged.

References

(1) Baibich, M. N.; Broto, J. M.; Fert, A.; Van Dau, F. N.; Petroff, F.; Eitenne, P.; Creuzet, G.; Friederich, A.; Chazelas, J. *Phys. Rev. Lett.* **1988**, *61*, 2472.
(2) Dieny, B.; Speriosu, V. S.; Parkin, S. S. P.; Gurney, B. A.; Wilhoit, D. R.; Mauri, D. *Phys. Rev. B* **1991**, *43*, 1297.
(3) Katine, J. A.; Albert, F. J.; Buhrman, R. A.; Myers, E. B.; Ralph, D. C. *Phys. Rev. Lett.* **2000**, *84*, 3149.
(4) Myers, E. B.; Ralph, D. C.; Katine, J. A.; Louie, R. N.; Buhrman, R. A. *Science* **1999**, *285*, 867.
(5) Ji, Y.; Chien, C. L.; Stiles, M. D. *Phys. Rev. Lett.* **2003**, *90*, 106601.
(6) Özyilmaz, B.; Kent, A. D.; Sun, J. Z.; Rooks M. J.; Koch, R. H. *Phys. Rev. Lett.* **2004**, *93*, 176604.
(7) Yanson, I. K.; Naidyuk, Yu. G.; Bashlakov, D. L.; Fisun, V. V.; Balkashin, O. P.; Korenivski, V.; Konovalenko, A.; Shekhter, R. I. *Phys. Rev. Lett.* **2005**, *95*, 186602.
(8) Polianski, M. L.; Brouwer, M. R. *Phys. Rev. Lett.* **2004**, *92*, 026602.
(9) Stiles, M. D.; Xiao, J.; Zangwill, A. *Phys. Rev. B* **2004**, *69*, 054408.
(10) Chen, T. Y.; Ji, Y.; Chien, C. L.; Stiles, M. D. *Phys. Rev. Lett.* **2004**, *93*, 026601.
(11) Konovalenko, A.; Korenivski, V.; Yanson, I. K.; Naidyuk, Yu. G. *J. Appl. Phys.* **2006**, *99*, 08G503.
(12) Özyilmaz, B.; Kent, A. D. *Appl. Phys. Lett.* **2006**, *88*, 162506.
(13) Naidyuk, Yu. G.; Yanson, I. K. *Point Contact Spectroscopy*; Springer Series in Solid-State Sciences, Vol. 145; Springer Science+Business Media, Inc.: New York 2005.
(14) Gruyters, M.; Bernhard, T.; Winter, H. *Phys. Rev. Lett.* **2005**, *94*, 227205.
(15) Zhao, H. B.; Talbayev, D.; Lupke, G.; Hanbicki, A. T.; Li, C. H.; van't Erve, M. J.; Kioseoglou, G.; Jonker, B. T. *Phys. Rev. Lett.* **2005**, *95*, 137202.

NL0628192