Spin Filtering of Hot Holes in a Metallic Ferromagnet

T. Banerjee, E. Haq, M. H. Siekman, J. C. Lodder, and R. Jansen

MESA$^+$ Institute for Nanotechnology, University of Twente, 7500 AE Enschede, The Netherlands

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Spin-dependent transport of nonequilibrium holes in ferromagnetic thin films and trilayers is investigated using ballistic hole magnetic microscopy. For Co, the hole attenuation length is short and increases from 6 to 10 Å in the energy range 0.8 to 2 eV. The hole transmission of a $\mathrm{Ni_{81}Fe_{19}/Au/Co}$ trilayer is clearly spin dependent, resulting in a surprisingly large current change by a factor of 2.3 in a magnetic field. The energy and spin dependence of the hole transmission cannot be explained by the phase space available for inelastic decay of the hot holes.

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Spin-dependent transmission of nonequilibrium, so-called hot electrons in ferromagnetic metals such as Co, Ni, and Fe has been extensively studied for several decades [1–3]. From the electronic structure of such ferromagnetic metals it is evident that a spin asymmetry of the inelastic lifetime should exist, since there is a large difference in the number of unoccupied final states above the Fermi level (E_F) into which the hot electrons can scatter. This was confirmed by time- and spin-resolved two photon photoemission experiments [4]. Significant progress towards calculation of spin-dependent quasiparticle lifetimes in ferromagnets was made recently [5]. Transport experiments using ballistic electron magnetic microscopy [6], spin-valve transistors [7], and magnetic tunnel transistors [8] have given valuable insight into the factors that control spin-dependent hot-electron transmission. For several ferromagnets, the attenuation length for the majority spin hot electrons was determined [6–8] to be between 25 and 80 Å. The minority spin attenuation length is typically two to six times shorter. The roles of interfaces and scattering by spin waves have also been studied [9]. Spin filtering of hot electrons has been successfully applied in magnetic sensor and memory devices [7,10], spin injection into semiconductors [11], current induced precessional magnetization reversal [12], and nanoscale magnetic imaging [6].

While spin-dependent transport and lifetimes of hot electrons above E_F has been well addressed, spin transport of nonequilibrium holes below E_F has not yet been explored. However, in many nonequilibrium phenomena, such as ultrafast laser-induced demagnetization, [13] nonequilibrium electrons and holes coexist and a complete picture requires understanding of the spin-dependent behavior of the hot holes, too. In this Letter, we report on the first experimental observation of spin-dependent transport of holes in ferromagnets for hole energy up to 2 eV below E_F, using a hybrid structure of ferromagnetic metals and a p-type semiconductor. We determine the hole attenuation length in ferromagnetic Co to be 6.5 ± 1 Å at 0.8 eV, increasing to 10 ± 1 Å at 2 eV. We observe a clear spin dependence of the hole transmission in a $\mathrm{Ni_{81}Fe_{19}/Au/Co}$ trilayer resulting in a current change by a factor of 2.3 in a magnetic field. Such a large field sensitivity for holes is surprising. Efficient spin filtering of hot electrons is readily understood using Fermi's golden rule and the fact that minority spin electrons have a much larger number of empty states into which to decay (see the electronic structure [14] of Co in Fig. 1). However, for holes of different spin in Co there is no significant difference in the number of states below E_F into which to decay. Therefore, a large spin asymmetry, comparable in magnitude to that of hot electrons, was not expected for hot holes and indicates that other factors such as the propagation velocity of the holes are important. A second important implication of the results is that it presents a first step in the development of complementary spintronics in which complementary structures based on n-type or p-type semiconductors are used together. This requires spin-dependent transport phenomena that work for electron as well as for hole transport, which has now been achieved in a single ferromagnet-silicon hybrid structure in the form of spin filtering of nonequilibrium carriers (hot electrons and now also holes).

To probe hole spin transport, we employ ballistic hole magnetic microscopy (BHMM, see Fig. 1). The technique is derived from ballistic electron emission microscopy (BEEM) [15] in which nonequilibrium carriers are emitted from the tip of a scanning tunneling microscope and locally injected into a (ferromagnetic) metal overlayer on a n-type semiconductor. In BHMM, a p-type semiconductor is used and the emitter bias is positive, such that a current of hot holes [16] is injected by tunneling. After spin filtering in the ferromagnetic layers, these are transmitted into the valence band of the semiconductor, provided the energy and momentum criteria for crossing the metal-semiconductor Schottky barrier are satisfied. All data reported here were obtained in ultrahigh vacuum (UHV) using PtIr metal tips. Measurements were done at 150 K to reduce noise in the metal-semiconductor collector diode. Details of the BEEM setup have been described elsewhere [17]. The samples are p-Si/Au(70 Å)/FM/Au(30 Å) where FM denotes either a single Co layer or a $Ni_{81}Fe_{19}/Au/Co$ trilayer. The metal layers are grown on a H-terminated *p*-Si(001) substrate by thermal evaporation in a molecular-beam epitaxy system at a base pressure of 10^{-10} mbar and transferred *ex situ* to the UHV-BEEM system [17]. At the *p*-Si/Au interface, a high quality Schottky barrier of 0.3 ± 0.03 eV is formed.

Figure 2 shows a typical spectrum of the collected hole current (I_{hole}) as a function of hole-injection voltage of a 100 Å thick Au layer on a *p*-Si substrate along with a series of spectra for different Co layer thickness (10–36 Å). The sign of the current corresponds to holes flowing from the metal stack to the Si and into the Ohmic back contact. With an increase in tip voltage we observe a rather abrupt onset of I_{hole} at around 0.30 ± 0.03 eV corresponding to the Schottky barrier height of the *p*-Si/Au interface. We confirmed that I_{hole} scales linearly with the injected tunnel current. The hole transmission of 16% at +2 V for Au is rather large, indicating a long inelastic lifetime of holes in Au, in agreement with recent calculations [18].

Introduction of a single Co(10 Å) layer sharply reduces I_{hole} by 2 orders of magnitude, and a further decrease is observed for increasing Co thickness up to 36 Å (Fig. 2). The large reduction of I_{hole} is the cumulative effect of attenuation of holes in the bulk of Co as well as at the Co/Au interfaces. The bottom panel of Fig. 2 shows I_{hole} normalized to its value at 2 eV. If the attenuation length for holes is not dependent on energy, then all the curves at different Co thickness would collapse onto a single curve. However, we observe a distinct change in the direction of curvature. This demonstrates that the attenuation length for holes in Co depends on energy and is shorter for hole energy closer to E_F.

From the data for the hole transmission in single Co layers we obtain the hole current per nA of injected holes as a function of Co thickness, for different injection voltage of 2, 1.4, and 0.8 eV (see top panel of Fig. 3). We observe that the hole transmission decays exponentially with Co thickness d. We extract the attenuation length λ for hole transmission from $I_c(d, E) = I_c(0, E)\exp[-d/\lambda(E)]$, where E is the hole-injection energy. We find that the attenuation length for hole transmission in Co at 0.8 eV is 6.5 ± 1 Å and it increases to 10 ± 1 Å at an energy of 2 eV. Hence, we find that the hole attenuation length is quite short, and shorter by at least a factor of 2 when compared to the majority spin attenuation length for hot electrons [6–8]. The energy dependence of λ for holes is shown in the bottom panel of Fig. 3. We see that λ for holes in Co increases with energy, despite the fact that for holes of higher energy there is a larger phase space of final states into which to decay. According to Fermi's golden rule, this would result in a shorter attenuation length at higher energy, as is observed in noble metals and also for majority spin hot electrons. This issue will be addressed later.

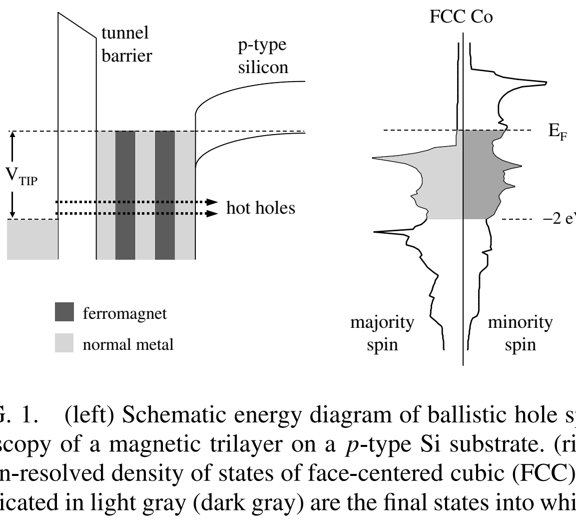

FIG. 1. (left) Schematic energy diagram of ballistic hole spectroscopy of a magnetic trilayer on a *p*-type Si substrate. (right) Spin-resolved density of states of face-centered cubic (FCC) Co. Indicated in light gray (dark gray) are the final states into which a hot hole in the majority (minority) spin bands can decay from an initial state of 2 eV below E_F.

From the top panel of Fig. 3 we can also determine the interface attenuation by extrapolating the exponential curves to zero Co thickness and comparing it with the hole current for a pure Au film (i.e., without Co). We

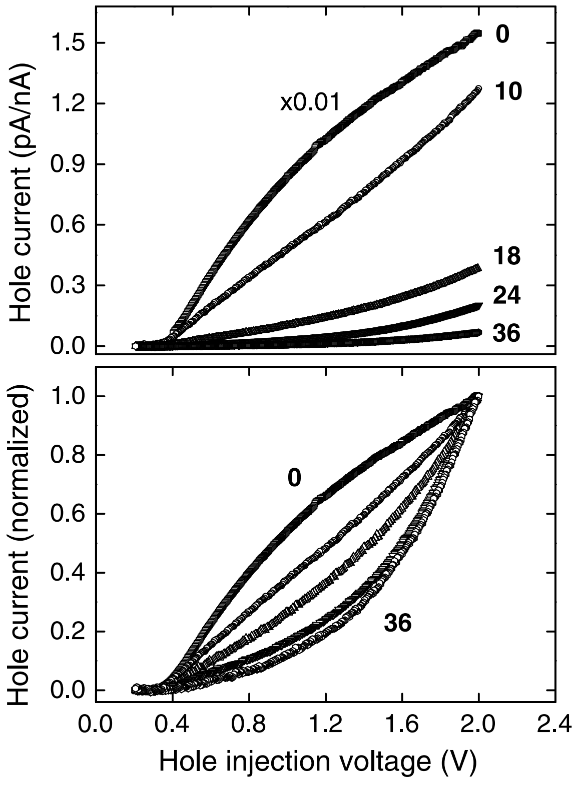

FIG. 2. (top panel) Hole current per nA of injected tunnel current versus tip voltage for *p*-Si/Au(70 Å)/Co/Au(30 Å), with Co thickness of 0, 10, 18, 24, and 36 Å as indicated by the labels. The curve for zero Co thickness has been divided by 100. Each curve represents an average over an area of at least 150 nm square. $T = 150$ K. (bottom panel) Data normalized to a tip bias of 2 eV.

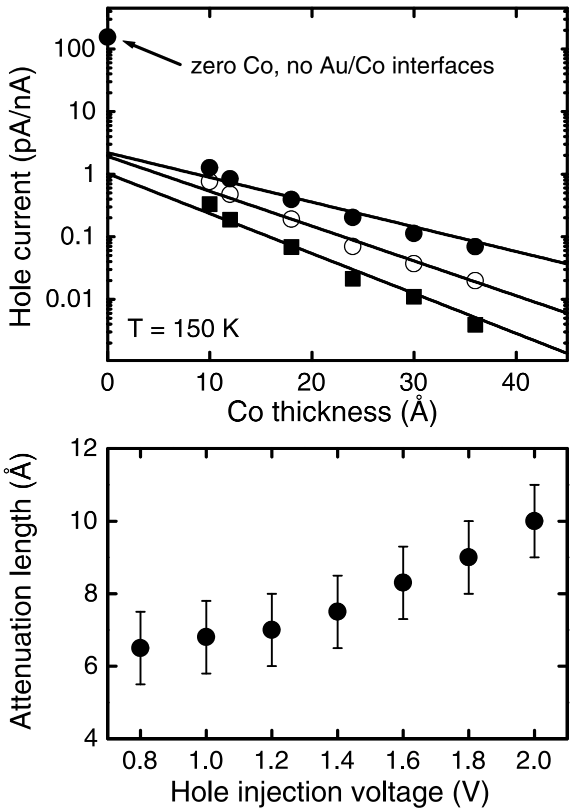

FIG. 3. (top panel) Hole current per nA of injected tunnel current in p-Si/Au(70 Å)/Co/Au(30 Å) as a function of Co thickness for emitter bias of 2 eV (solid circles), 1.4 eV (open circles), and 0.8 eV (solid squares). Solid lines represent exponential decay with attenuation length of 10 Å, 7.5 Å, and 6.5 Å. (bottom panel) The attenuation length as a function of hole-injection voltage.

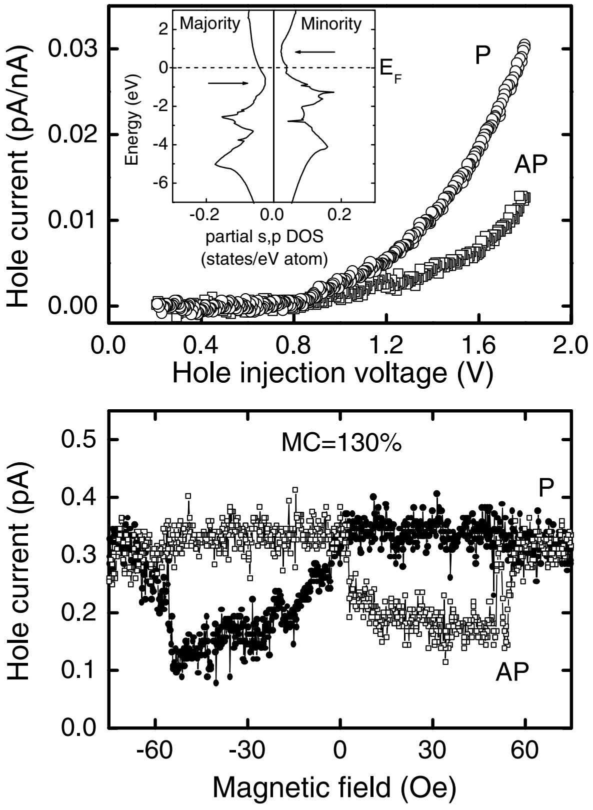

FIG. 4. (top panel) Hole current versus tip voltage for p-Si/Au(70 Å)/$Ni_{81}Fe_{19}$(18 Å)/Au(70 Å)/Co(18 Å)/Au(30 Å) at a magnetic field of +100 Oe and −30 Oe, respectively, corresponding to parallel (P) and antiparallel (AP) alignment of the $Ni_{81}Fe_{19}$ and Co magnetization. The inset shows the spin-resolved s, p partial density of states for FCC Co, with arrows indicating the pronounced dips near the top of the d bands. (bottom panel) Hole current versus magnetic field at +2 V and 10 nA hole-injection current.

find an attenuation by a factor of 35 caused by the introduction of two additional Au/Co interfaces. The interface attenuation for hot electrons in identical Co films [17] is less than a factor of 2. The interfacial attenuation is a combination of the mismatch of the electronic states at both sides of the interface and elastic scattering due to interface disorder, defects, etc.

While a single ferromagnetic layer may act as a spin filter, to demonstrate the spin dependence of hole transmission, one requires two ferromagnetic layers such that a spin-valve effect is obtained when the relative orientation of their magnetization is varied. Results for a $Ni_{81}Fe_{19}$(18 Å)/Au(70 Å)/Co(18 Å) trilayer are shown in Fig. 4. The curves in the top panel show that $I_{\rm hole}$ is smaller due to the extra $Ni_{81}Fe_{19}$ layer and below the detection limit for voltage under 0.8 V. The hole current depends strongly on magnetic field and is largest when the magnetization of the two ferromagnetic layers are aligned parallel (P), and about a factor of 2 smaller in the antiparallel (AP) configuration. Keeping the tunnel current constant at 10 nA and tip bias at 2 V, we have recorded $I_{\rm hole}$ while the magnetic field is swept from +100 Oe in steps of 0.1 Oe to −100 Oe and back. The magnetizations were first saturated to the P state in a magnetic field of +100 Oe. Sweeping the field to negative values first causes a transition into the AP state due to reversal of the soft $Ni_{81}Fe_{19}$ layer, followed by a transition around −50 Oe back to the P state when the Co magnetization is also reversed. On the retrace a similar behavior is observed, with the expected magnetic hysteresis. This clear spin-valve effect of the hole current unambiguously demonstrates that the transmission of holes in both the ferromagnetic metals is spin dependent.

From the change of $I_{\rm hole}$ of 0.34 pA in the P state to 0.15 pA in the AP state we obtain a hole magnetocurrent (MC), defined as $(I^{\rm P}_{\rm hole} - I^{\rm AP}_{\rm hole})/I^{\rm AP}_{\rm hole}$, of 130% ± 30%. The difference between $I^{\rm P}_{\rm hole}$ and $I^{\rm AP}_{\rm hole}$ is related to the asymmetry in spin-dependent transmission of majority T^M and minority T^m spin hot holes in the ferromagnetic layers via

$$I_{\mathrm{hole}}^{\mathrm{P}} \propto T_{\mathrm{NiFe}}^{M} T_{\mathrm{Co}}^{M} + T_{\mathrm{NiFe}}^{m} T_{\mathrm{Co}}^{m}, \tag{1}$$

$$I_{\mathrm{hole}}^{\mathrm{AP}} \propto T_{\mathrm{NiFe}}^{M} T_{\mathrm{Co}}^{m} + T_{\mathrm{NiFe}}^{m} T_{\mathrm{Co}}^{M}. \tag{2}$$

An initially unpolarized current after transmission of a ferromagnetic film acquires a spin polarization P given by $\mathrm{P} = (T^M - T^m)/(T^M + T^m)$. This directly relates to magnetocurrent via $\mathrm{MC} = (2\mathrm{P}_{\mathrm{NiFe}}\mathrm{P}_{\mathrm{Co}})/(1 - \mathrm{P}_{\mathrm{NiFe}}\mathrm{P}_{\mathrm{Co}})$. If both ferromagnets are the same, the measured MC of 130% corresponds to $\mathrm{P} = 63\%$ (or -63%) for each of the 18 Å films. This gives us a value of the spin asymmetry of transmission $T^M/T^m = 4.4$ (or 0.23), which is quite large indeed.

As noted in the introduction, such a large spin asymmetry cannot be explained by the phase space available for hole decay. We propose that the difference in the group velocity of the states in which holes of different spin propagate may be responsible. We note that the attenuation length is a product of the inelastic lifetime and the group velocity (v_g) of the corresponding spin bands. In general, v_g in pure or *s, p*-like states is larger than in the highly localized *d*-like states. In ferromagnets such as Co, the *s* and *p* character of hole states is reduced due to hybridization with *d* states. As shown in the inset of Fig. 4, near the top of the *d* band where the density of states peaks and consists of highly localized *d* states, there is a pronounced suppression (dip) in the *s, p* character of the states [19]. This dip occurs above E_F for the minority spin bands, since the top of the minority *d* band lies above E_F. In contrast, for majority spins the dip in the partial density of *s, p*-like states occurs for states below E_F that are relevant for hole transport. Thus, holes in the majority spin bands are expected to have smaller group velocity than holes in the minority spin bands, making the former more susceptible to inelastic scattering during the longer time spent in the ferromagnetic layer. This provides a possible explanation for the observed spin asymmetry of the hole transmission and suggests $T^m > T^M$, which is opposite to that of hot electrons. Also note that the partial density of *s, p* states increases again for energies further below E_F, consistent with the observed increase of attenuation length at higher energy.

Other explanations such as hole-phonon interactions are too weak to yield attenuation lengths below 10 Å at $T = 150$ K. However, spin-dependent interface transmission or the interaction of holes with spin waves cannot be ruled out at this point. For hot electrons, spontaneous excitation of (nonthermal) spin waves has been suggested [9] as a source of spin asymmetry. Because of spin conservation, spontaneous emission of a spin wave must be accompanied by a spin flip of an electron from minority to majority spin. Therefore, the process is forbidden for majority spin hot electrons. For holes, the spin asymmetry is opposite and spin-wave emission is forbidden for hot holes in the minority spin bands. However, spin-wave emission is possible for a majority spin hot hole if it is annihilated when an equilibrium electron is spin-flipped from minority to majority, leaving a hole in the minority spin band. The interaction with spin waves thus predicts a shorter lifetime for holes in the majority spin bands ($T^m > T^M$). Theoretical efforts are needed to examine whether this interaction is sufficiently large in ferromagnets like Co with strong exchange interaction.

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