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Opposite Spin Asymmetry of Elastic and Inelastic Scattering of Nonequilibrium Holes Injected into a Ferromagnet

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The spin asymmetry of elastic and inelastic scattering of nonequilibrium holes injected into Co thin films is examined using a *p*-type magnetic tunnel transistor. Spin-dependent transmission yields a positive or negative magnetocurrent depending on Co thickness and hole energy. Up to a critical thickness of about 3 nm, (quasi)elastic scattering dominates with a short attenuation length (< 1 nm) and preferential attenuation of holes in the majority spin bands, consistent with spin-wave emission. At a larger Co thickness, inelastic scattering dominates with a larger attenuation length (~ 4 nm) and opposite spin asymmetry.

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Spin-dependent scattering of electrons in a ferromagnet (FM) is fundamental to the understanding and application of various magnetic systems [1,2]. The spin-dependent transport can involve electrons at the Fermi energy (E_F) but in many cases also nonequilibrium, so-called hot carriers. One example is the magnetic tunnel junction (MTJ) which exhibits a large tunnel magnetoresistance (TMR) at low bias voltage where electrons near E_F are of relevance, while the TMR drops significantly at higher bias where states away from E_F also participate in the tunneling process [3]. Studies of the transmission of hot electrons injected into the empty states above E_F of FM thin films has shown [4–8] that the attenuation length of majority spin hot electrons is much longer than that of minority spins, believed to be due to the spin dependence of the number of states available for inelastic scattering by electron-hole (e - h) pair excitation. This spin filtering of hot electrons has been applied in magnetic devices [9–11], spin injection into a semiconductor [12], and magnetic imaging [13]. One can also achieve spin transfer torque switching of the magnetization by injecting spin-polarized hot electrons into a FM layer in a MTJ [14]. When the spin-polarized hot electrons are injected into the states above E_F of one FM electrode, holes are simultaneously injected into the states below E_F of the counterelectrode. Also, in other nonequilibrium magnetic phenomena, hot electrons and holes are both present, such that it is relevant to understand the spin-dependent scattering of both hot electrons and holes.

The transport of nonequilibrium carriers below E_F (hot holes) is not well understood. Recently, the first demonstration of spin-dependent hole transmission in a FM has been reported using ballistic hole magnetic microscopy [15], where a clear spin-valve effect of 130% and attenuation lengths as short as 0.6 nm were observed. In this Letter, we report on the spin-dependent transmission of holes injected into a ferromagnetic Co layer using a *p*-type magnetic tunnel transistor (MTT) and investigate the spin-dependent attenuation of holes due to elastic and inelastic

scattering, respectively. Surprisingly, we observe a sign reversal of magnetocurrent (MC) as a function of the Co thickness and the hole energy. It shows that nonequilibrium holes, when injected into a FM, scatter (quasi)elastically in the first few nanometers of material due to spin-wave emission, while at larger depth into the film the attenuation is dominated by inelastic scattering by e - h pair excitations, but with opposite spin asymmetry.

The *p*-type MTT [16] combines a magnetic tunnel junction and a *p*-type Si substrate and consists of a FM tunnel emitter, a base with a single FM layer, and a Au/*p*-Si Schottky diode collector (see Fig. 1). When an emitter bias (V_E) is applied between the emitter and the base, spin-polarized hot holes are injected into the states below E_F of the FM base by a tunneling process. After spin-dependent transmission through the FM base, holes are collected in the valence band of the *p*-type semiconductor if they have proper energy and momentum to overcome the Schottky barrier of height ~ 0.3 eV formed between Au and *p*-type Si. The MTT

FIG. 1. Left: Schematic energy diagram of a *p*-type MTT of $\text{Ni}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3/\text{Co}/\text{Au}/p\text{-type Si}$. The curve in the Al_2O_3 tunnel barrier represents the energy distribution of tunneling electrons (holes). Right: Collector hole current (I_C) versus emitter bias (V_E) in a *p*-type MTT with a 3.6 nm Co base.

structures of $\text{Ni}_{80}\text{Fe}_{20}$ (5 nm)/ Al_2O_3 (2 nm)/Co(1.5–12 nm)/Au(7 nm)/*p*-type Si(100) were fabricated as previously described [16]. The magnetization of the $\text{Ni}_{80}\text{Fe}_{20}$ emitter is pinned by an antiferromagnetic CoO layer whose blocking temperature is around 240 K. Special care was taken to assure that the Co films in the base are smooth, continuous, and free of pinholes down to the lowest thickness of 1.5 nm [17]. Transport measurements were conducted at 82 K using a four-point geometry for the emitter-to-base tunnel junction and a separate Ohmic contact to the back of the Si collector.

The right panel in Fig. 1 shows the typical bias voltage dependence of the collector hole current (I_C). We observe an abrupt increase of I_C at an onset voltage of around 0.3 eV, corresponding to the Schottky barrier height of the *p*-type Si/Au contact. Figure 2 shows the MC of a MTT with a 3.6 nm thick Co base for different emitter biases ranging from 0.6 to 1.4 V. The MC is defined by $(I_C^P - I_C^{\text{AP}})/I_C^{\text{AP}}$, where I_C^P (I_C^{AP}) is the collector current in the parallel (antiparallel) alignment of the magnetization of the two FM layers. The curves show only the switching of the Co base layer as the magnetization of the $\text{Ni}_{80}\text{Fe}_{20}$ emitter is pinned by antiferromagnetic CoO with an exchange field larger than -100 Oe. The negative magnetic field corresponds to the parallel state. We obtain a negative MC of -18% at 0.6 V, which means a larger hole current in the antiparallel state than in the parallel state. This is unexpected and is the first observation of a negative mag-

FIG. 2. Magnetic field dependence of the collector current in a *p*-type MTT with a 3.6 nm thick Co base at different emitter bias, as indicated in each panel. Labels P and AP denote parallel and antiparallel alignment, respectively, of the emitter and base magnetizations. $T = 82$ K.

netic response of a MTT. As V_E is increased, the magnitude of the MC decreases and becomes almost zero at 0.9 V. With further increase in the V_E , surprisingly, the MC changes its sign, resulting in a positive MC of $+5\%$ at 1.4 V.

The MC versus the V_E of MTTs with various Co base thicknesses between 1.5 and 12 nm is shown in Fig. 3. A MTT with a thin Co base of 1.5 nm shows a negative MC (-25%) at 0.5 V and preserves the negative MC for bias voltages up to 1.4 V. A MTT with a Co base thicker than 8 nm shows positive MC for all bias voltages. For a Co base of intermediate thickness, the MC is negative at small V_E and becomes positive with increasing V_E as also shown in Fig. 2. At large V_E , the MC tends to zero irrespective of the Co thickness, which is due to the reduction of the tunnel spin polarization of the emitter interface at larger bias [18]. Apart from this, there is a clear trend of negative MC for a thin Co base and a small V_E and positive MC for a thick Co and a large V_E . Since the tunnel spin polarization of the $\text{Ni}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3$ emitter interface is known to be positive [19], the base transmission is responsible for the negative MC. While the base transmission is dominated by the volume contribution for a thick base, a possible interfacial contribution may become important as the Co thickness is decreased. Thus, the negative MC with a thin base might be due to an interface with a larger transmission of holes in the minority spin bands. Below, we will first rule out this explanation.

There are two interfaces in the base: the $\text{Al}_2\text{O}_3/\text{Co}$ tunnel interface and the Co/Au interface. To investigate the contribution of these interfaces to the spin asymmetry in the hole transmission, we have modified the base structure by inserting a Au layer either in the middle of the Co or at the tunnel interface. The upper panel in Fig. 4 shows the MC of a MTT with a base of Co(3.6 nm)/Au(2 nm)/Co(4.4 nm) compared with a MTT with a single Co base

FIG. 3. MC versus V_E in *p*-type MTTs with different thickness of Co base ranging from 1.5 to 12 nm. $T = 82$ K.

FIG. 4. MC versus V_E in p -type MTTs with an extra Au layer inserted into the base. Top panel: Co(8 nm) compared to Co(3.6 nm)/Au(2 nm)/Co(4.4 nm). Bottom panel: $\text{Al}_2\text{O}_3/\text{Co}$ (3.6 nm) compared to $\text{Al}_2\text{O}_3/\text{Au}$ (2 nm)/Co(3.6 nm). $T = 82$ K. The solid lines are a guide to the eyes.

of the same total Co thickness (8 nm). The insertion of the Au layer adds two additional Co/Au interfaces in the base, which makes the MC more positive at an emitter bias lower than 0.8 V. There are no changes at higher bias. This indicates that the contribution of the Co/Au interface is not negative but even slightly positive. This is expected because the band structure of Au is better matched to that of the majority spin bands of Co. Thus, spin-dependent transmission across the Co/Au interface cannot be responsible for the negative MC observed.

Next we examine the $\text{Al}_2\text{O}_3/\text{Co}$ interface. The tunnel spin polarization for $\text{Al}_2\text{O}_3/\text{Co}$ is known to be positive from spin-polarized tunneling measurements [20]. However, the p -type MTT utilizes energy states of 0.3 eV or more below E_F whose spin polarization can be different from that of the states near E_F . We inserted a Au layer of 2 nm between the Al_2O_3 tunnel barrier and the Co base, which should essentially quench the tunnel spin polarization of the $\text{Al}_2\text{O}_3/\text{Co}$, while leaving unchanged the tunnel spin polarization of the $\text{Ni}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3$ emitter interface. Therefore, spin-polarized holes are still injected into the base. The result is shown in the lower panel in Fig. 4, where the Co base thickness is 3.6 nm. The MC of a MTT with the Au layer becomes more negative as compared to that of a MTT without Au, for all biases. This implies that the tunnel spin polarization of the interface was initially positive. Thus, the $\text{Al}_2\text{O}_3/\text{Co}$ interface is also not responsible for the negative MC. Note that the TMR of the $\text{Ni}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3/\text{Au}/\text{Co}$ emitter-base tunnel junction is found to be almost zero (less than 0.1%), confirming the zero tunnel spin polarization of the $\text{Al}_2\text{O}_3/\text{Au}/\text{Co}$ interface.

Since the negative MC and the transition to positive MC at larger thickness cannot be explained by the interfaces, we study the hole transmission of the Co base. The top panel in Fig. 5 shows the transmitted hole current (I_C/I_E) as a function of the Co layer thickness (t_{Co}), where I_E is the injected emitter current. The I_C/I_E decays drastically for small thicknesses up to ~ 3 nm and rather slowly for larger thicknesses. We can extract the attenuation length λ from the slope of the I_C/I_E versus t_{Co} curve, using exponential decay $\propto \exp(-t_{\text{Co}}/\lambda)$. Clearly, there are two slopes corresponding to two attenuation lengths in different thickness regimes. The attenuation lengths at 0.6 V (1.4 V) are 0.8 ± 0.1 (0.9 ± 0.1) and 3.6 ± 0.2 nm (4.0 ± 0.2 nm) for the small and large thickness regimes, respectively. This demonstrates that there are two scattering processes governing the hole transmission. One dominates in the small thickness regime with a strong attenuation, while the other dominates for large thickness with a weaker attenuation. We attribute this to elastic and inelastic scattering, which is rationalized in the following way. Since the tunneling process favors states with small momentum component parallel to the interface, the injected current predominantly involves holes traveling in the direction perpendicular to the Co film. Hence, the hole current initially is very sensitive to elastic scattering, which produces a strong decay of the collector current (recall that transmission across the Au/Si Schottky barrier depends on the energy as well as on the momentum [7]). As the holes travel through the base

FIG. 5. Transmitted hole current I_C/I_E for parallel state (top panel) and MC (bottom panel) as a function of Co thickness for an emitter bias of 0.6 (circles), 0.8 (open squares), and 1.4 V (triangles).

layer, their momentum distribution gradually broadens due to the elastic scattering. After a certain distance from the injection interface, the momentum distribution has become completely isotropic such that the elastic scattering no longer leads to attenuation of the hole current. Consequently, inelastic scattering with a different (longer) attenuation length dominates in the large thickness regime.

The transition in the dominating scattering mechanism is directly related to the sign reversal of the MC with Co base thickness, as shown in the bottom panel in Fig. 5 for different emitter bias of 0.6, 0.8, and 1.4 V. The MC is negative in the small thickness regime where the elastic scattering dominates and becomes more positive with increasing Co thickness, corresponding to the increase of the inelastic scattering contribution. The negative MC is therefore attributed to the elastic scattering. This implies that elastic scattering is stronger for holes in the majority (M) spin bands, thus producing a shorter attenuation length compared to holes in the minority (m) spin bands ($\lambda_M^{\text{el}} < \lambda_m^{\text{el}}$). Moreover, the positive MC at large Co thickness implies that inelastic scattering has the opposite spin asymmetry ($\lambda_M^{\text{inel}} > \lambda_m^{\text{inel}}$). This explains the sign reversal of the MC with t_{Co} .

For the inelastic scattering, we found that holes in the minority spin bands are preferentially attenuated. This is similar to the spin asymmetry for hot electrons [6,7,11]. However, the origin of the spin asymmetry of the inelastic scattering of holes is not clear yet. As argued before [15], the phase space available for inelastic decay of the hot holes in the majority and minority spin bands is not hugely different (as it is for hot electrons) since a large density of filled d states below E_F exists for both spins. Therefore, a spin-dependent group velocity was considered [15]. Detailed calculations of the inelastic lifetimes and scattering lengths, such as recently presented for hot electrons [21], may shed more light on this issue.

With respect to the elastic scattering, it shows the opposite spin asymmetry, with preferential attenuation of holes in the majority spin bands. This may be due to spontaneous emission of spin waves with a long wavelength (small energy) for which the dominant effect is a change of the momentum of the hot holes (quasielastic). For hot electrons, spontaneous spin-wave emission is accompanied by a spin flip of the primary hot electron [22,23] such that only minority spin electrons can emit a spin wave. Therefore, for hot electrons the spin asymmetry due to spin-wave emission and e - h pair excitation is the same. For holes, the spin asymmetry due to spontaneous spin-wave emission is opposite, since emission is allowed only for hot holes in the majority spin bands. This would yield $\lambda_M < \lambda_m$. Thus, the observed spin asymmetry of the elastic scattering is consistent with spontaneous emission of spin waves by the injected hot holes. Note that the spin asymmetry of the elastic scattering almost disappears at 1.4 V (Fig. 5, bottom panel). It was shown before [21,22] that the cross section for hot-electron scattering by spontaneous

spin-wave emission is significant only at low energy (below 1.2 eV). In a similar fashion, one may explain the small spin asymmetry for elastic scattering of holes at higher energy.

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