Enhanced ferromagnetic damping in Permalloy/Cu bilayers

Th. Gerrits,[a] M. L. Schneider, and T. J. Silva
National Institute of Standards and Technology, Boulder, Colorado 80305

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We have investigated the enhancement of ferromagnetic damping for thin Permalloy ($Ni_{80}Fe_{20}$) films grown with Cu capping layers of variable thickness (5–1000 nm). The measurements were performed with a pulsed inductive microwave magnetometer in the frequency range between 2.3 and 2.7 GHz. The damping was enhanced if the Cu layers were thicker than the spin-diffusion length of $l_s \approx 250$ nm. For example, the damping was enhanced by 30% for a Permalloy (3 nm)/Cu (1000 nm) bilayer relative to the damping for Permalloy with a 5-nm-thick Cu capping layer. Existing theory for spin pumping from the Permalloy layer into the Cu layer was used to model the additional contribution to damping for these bilayer systems. Additional experiments on Permalloy (5 nm)/Cu(x nm)/Ta (5 nm) provided indirect evidence for spin accumulation inside the Cu layer.
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I. INTRODUCTION

We investigated how the thickness of the Cu layers affects the damping of gyromagnetic precession in $Ni_{80}Fe_{20}$(Permalloy)/Cu bilayer films excited by magnetic-field pulses and measured with a pulsed inductive microwave magnetometer (PIMM).[1] These experiments were performed to test the prediction that spin accumulation occurs in adjacent Cu layers due to the transmission of spin-polarized electrons caused by precession of the ferromagnetic moment.[2–4]

Recently, Kiselev *et al.*[5] and Rippard *et al.*[6] have demonstrated that spin-momentum transfer can be used to compensate damping by passing a spin-polarized current through a thin magnetic layer. There, the spin-polarized currents were generated by the accumulation of transverse spin components in the Cu spacer layer separating two ferromagnetic layers. At a given critical current, the accumulation is sufficiently large to drive spin precession in the thinner of the two ferromagnetic layers. Therefore, studies of the spin accumulation in ferromagnet/normal-metal (FM/NM) multilayers may lead to a better understanding of the critical current in spin-momentum transport experiments. In particular, the study of spin pumping in FM/NM bilayers provides an excellent opportunity to elucidate the important parameters that affect spin transport at the interface.

The reactive effect on the ferromagnetic damping due to spin pumping at a FM/NM interface and the resulting spin accumulation were theoretically described by Brataas *et al.*[2] The enhancement of ferromagnetic damping due to spin pumping has also been confirmed experimentally.[7–11] However, there has not yet been any systematic study of damping in simple Permalloy/Cu bilayer samples. Due to the *s*-like character of carriers in Cu, the spin lifetime is relatively long. This results in long spin-diffusion lengths, even at room temperature.[12] The aim of this study was to measure the increase in damping in the adjacent ferromagnet due to a spin current that is injected into the normal metal via ferromagnetic precession. The spin current decreases with distance z from the normal metal/ferromagnet interface, with a characteristic decay length given by the spin-diffusion length ℓ_s. Spin-flip events within the Cu layer cause a reduction in the total precession amplitude of the ferromagnet, thereby resulting in increased damping.

II. THEORY

According to Brataas *et al.*,[2] the injected spin current flows perpendicular to the normal metal/ferromagnet (NM/FM) interface and is polarized in the direction of $\hat{m} \times (d\hat{m}/dt)$, where $\hat{m}$ is the unit vector in the instantaneous direction of magnetization. The spin-pumping torque $\mathbf{T}_S^{\text{pump}}$ due to the injected spin current is[2]

$$\mathbf{T}_S^{\text{pump}} = \frac{h}{4\pi} g_{\uparrow\downarrow}\left(\hat{m} \times \frac{d\hat{m}}{dt}\right), \tag{1}$$

where $g_{\uparrow\downarrow} = g_{\downarrow\uparrow}$ is the dimensionless interfacial spin-mixing component in the spin-conductance tensor. The torque $\mathbf{T}_S^{\text{pump}}$ in Eq. (1) also represents the rate at which angular momentum quanta are injected from the ferromagnet into the normal metal. The mixing component may be approximated as $g^{\uparrow\downarrow} \approx \kappa k_F^2 A/4\pi$,[2] where k_F is the Fermi wave number for the normal metal, κ represents the number of conducting channels per surface atom (a value on the order of unity), and A is the total surface area of the interface. This spin current results in a quasi-instantaneous spin accumulation in the NM layer. The resultant spin accumulation decays at a rate given by the spin-flip time in the NM.

The torque term in Eq. (1) has the same form as the damping term in the Landau-Lifshitz-Gilbert equation.[13]

$$\frac{d\hat{m}}{dt} = -|\gamma|\mu_0[\hat{m} \times \mathbf{H}] + \eta\left[\hat{m} \times \frac{d\hat{m}}{dt}\right], \tag{2}$$

where η is the effective damping parameter, γ is the gyromagnetic ratio, μ_0 is the permeability of free space, and $\mathbf{H}$ is the total effective magnetic field. In Eq. (2) η can be represented in terms of the Gilbert damping parameter α and the spin-pumping contribution α' such that $\eta = \alpha + \alpha'$.

[a]Electronic mail: gerrits@boulder.nist.gov

In general, some angular momentum that is transmitted into the normal metal according to Eq. (1) diffuses back into the ferromagnet due to the nonequilibrium spin accumulation in the normal metal. In that case, the net spin-current torque acting on the ferromagnet is $\mathbf{T}_S = \mathbf{T}_S^{\text{pump}} - \mathbf{T}_S^{\text{back}}$. For high-$Z$ metals with a strong spin-orbit coupling, the spin-flip rate is large. Thus, the probability that a spin-polarized electron will lose its spin information in the normal metal is large compared with the probability that it reencounters the FM/NM interface during the random-walk process. A large spin-flip rate in turn reduces the backflow of spin angular momentum into the ferromagnet such that $\mathbf{T}_S^{\text{back}} \approx 0$. In a low-$Z$ metal with an s-like conductivity band such as Cu, the spin-flip rate is relatively small. If the Cu thickness δ_{NM} is much smaller than the spin-diffusion length, the probability that a carrier will return to the NM/FM interface is significant and $\mathbf{T}_S^{\text{back}}$ is no longer negligible. This accumulated spin distribution in the normal metal forms rapidly due to the high Fermi velocity of the electrons. Therefore, we can assume that the spin polarization reaches steady state instantaneously relative to the precessional rate of $\hat{m}$ in the ferromagnet. However, a sufficiently thick Cu layer increases the probability for a spin-flip event to occur during spin diffusion in the Cu. Hence, theory predicts an enhancement of the damping of the FM layer when $\delta_{\text{NM}} > \ell_s$. The spin-pumping component of damping is given by[14]

$$\alpha' = g^{\uparrow\downarrow} A^{-1} \cdot \frac{\gamma h}{M_S \delta_{\text{FM}}} \left[\frac{1}{1 + \beta \cdot g^{\uparrow\downarrow} A^{-1}} \right], \tag{3}$$

where β is the backflow factor, given by $\beta = (\ell_s e^2 / \sigma_{\text{NM}} h) \times [1/\tanh(\delta_{\text{NM}}/\ell_s)]$, δ_{FM} is the thickness of the ferromagnetic layer, and σ_{NM} is the conductivity of the normal metal. For $\ell_s \gg \delta_{\text{NM}}$ such that $\mathbf{T}_S^{\text{pump}} \approx \mathbf{T}_S^{\text{back}}$, the damping is unaffected, i.e., $\alpha' \to 0$ as $\beta g^{\uparrow\downarrow} A^{-1} \to \infty$. However, as $\delta_{\text{NM}} \to \ell_s$ then $\alpha' > 0$, and α' asymptotically approaches a maximum value of $\alpha' = g^{\uparrow\downarrow} \gamma \hbar / M_S A \delta_{\text{FM}}$ for $\delta_{\text{NM}}/\ell_s \gg 1$ and $(g^{\uparrow\downarrow} A^{-1}) \times (\ell_s \sigma_{\text{NM}}^{-1}) \ll h e^{-2}$. The latter condition essentially means that the resistance of a slab of the NM with thickness l_s must be much smaller than twice the unit of resistance per conducting channel.

We note that Eq. (3) is valid only if the precession frequency is much smaller than the spin-flip rate inside the normal metal. If the precession frequency is higher than the spin-flip rate, the instantaneous spin accumulation in the normal metal is increased, enhancing the spin-current backflow and, therefore, decreasing the net spin current into the normal metal. For Cu, the spin-flip rate is approximately 10^{11} s^{-1}, corresponding to a frequency of less than 15 GHz. Thus, operation at frequencies of just a few gigahertz is necessary to allow observation-enhanced damping due to spin pumping with Cu.

A significant increase in damping was observed in earlier experiments using Pt, Pd, and Ta capping layers.[8,9] For the case of a Pt capping layer the enhanced damping was a factor of 2 greater than the damping inferred from samples without a capping layer. However, for a Cu capping layer, the effect upon damping was inconclusive as measured to within the signal-to-noise ratio of the experiment.

III. EXPERIMENT

We investigated the validity of Eq. (3) by varying δ_{NM} over a wide range using a PIMM. The PIMM is capable of measuring precessional dynamics in Permalloy films between frequencies of 0.8 and 3 GHz. Since these frequencies are much lower than the spin-flip rate in Cu, the PIMM is ideally suited to observe enhanced damping as $\delta_{\text{NM}} \to \ell_s$. An external in-plane field is used to bias the sample magnetization perpendicular to the magnetic excitation pulse. A coplanar waveguide (CPW) structure was used to excite the sample. A pulse generator with a rise time of 50 ps and a duration of 10 ns generates a quasistep magnetic-field pulse in the structure. In response to the field pulse, the time-varying magnetization component along the field pulse direction induces a voltage in the CPW line that is measured with a sampling oscilloscope.[1] We extracted the damping values η by fitting the Fourier transform of the time-domain data to a simple damped harmonic oscillator resonance function in a frequency range where η is independent of frequency.[15] The reported value of η for each sample represents an average value for 11 different bias fields in the range from 5.6 kA/m (71 Oe) to 8.0 kA/m (101 Oe). This field range corresponds to precession frequencies between 2.3 and 2.7 GHz.

We studied samples consisting of Permalloy/Cu bilayers with 3, 5, and 10 nm Permalloy thicknesses and with $x = 5$ nm to $x = 1000$ nm Cu layer thicknesses. In addition, we also examined Permalloy (5 nm)/Cu(x nm)/Ta (5 nm) trilayers as control samples. All samples were dc magnetron sputtered and were deposited directly onto glass substrates. The glass substrates were Ar/O_2- and Ar-ion etched prior to the deposition of the Permalloy film to improve adhesion. We deposited both Permalloy and Cu at an Ar pressure of 4 mTorr. The Ta was sputtered at an Ar pressure of 5 mTorr. The deposited films had a roughness of less than 1 nm and the average grain size was about 30 nm as determined by atomic force microscopy.

IV. RESULTS

Figure 1 shows η as a function of Permalloy and Cu thicknesses. The values are plotted versus $g_L/(M_s \delta_{\text{FM}})$, where g_L is the Landé g factor and M_s is the saturation magnetization. The values for g_L and M_s for Permalloy films of varying thicknesses were obtained from an earlier study by Nibarger *et al.*[16] The inductive signal decreases with decreasing film thickness. Therefore, the uncertainty grows with decreasing film thickness. The graph shows the fits to the data using Eq. (3) for $\delta_{\text{NM}} = 100$, 200, and 500 nm and corresponding values of $\delta_{\text{FM}} = 3$, 5, and 10 nm. We used fixed values for α and ℓ_s of 0.0072 and 250 nm, respectively. The only fitting parameter used was the mixing component $g^{\uparrow\downarrow} A^{-1}$. The fits for all the samples independently yield $g^{\uparrow\downarrow} A^{-1} = 1.5 \times 10^{15}$ cm^{-2}. This agrees well with previously obtained values.[14] The inverse proportionality of η on δ_{FM} effectively eliminates eddy currents as a relevant effect; eddy currents scale in proportion to integrated magnetic flux and film conductance, resulting in a quadratic dependence upon δ_{FM}.

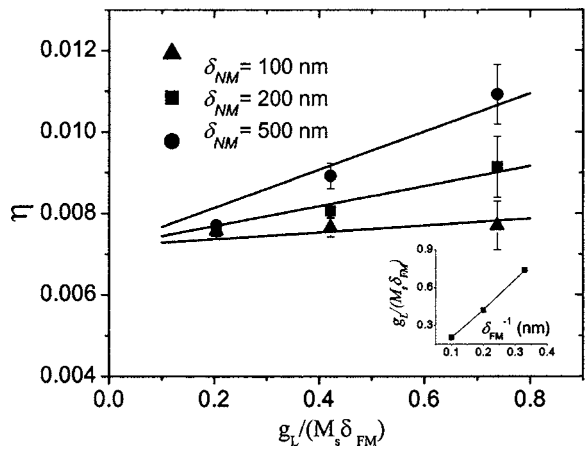

FIG. 1. Effective damping η as a function of three different Permalloy thicknesses δ_{FM} and three different Cu layer thicknesses δ_{NM}. The solid lines represent the fits from Eq. (3) for each δ_{NM} with a fitted mixing component $g^{\uparrow\downarrow}A^{-1}=1.5\times10^{15}$ cm^{-2} and a fixed diffusion length l_s=250 nm. The inset shows the dependence of $g_l(M_s\delta_{FM})^{-1}$ on $\delta_{FM}{}^{-1}$.

Figures 2(a)–2(c) show the damping values for three different Permalloy layers [(a) 3, (b) 5, and (c) 10 nm] as functions of δ_{Cu}. The solid lines are fits to Eq. (3), using a mixing component of 1.5×10^{15} cm^{-2} that was extracted from the fits in Fig. 1. The fits to Eq. (3) yielded a spin-diffusion length of $250\pm50, 300\pm13$ and 300 ± 70 nm for the 3, 5, and 10 nm Permalloy systems, respectively. Note that in Fig. 2(b) the data point for δ_{Cu}=1000 nm has been excluded from the fit. The theory is in good agreement with the data for varying Permalloy thicknesses. The value of α derived for each Permalloy film is insensitive to film thickness and varies between 0.0071 ± 0.0005 and 0.0072 ± 0.0001 and is only slightly lower than the commonly reported value for thick Permalloy films ($\alpha\sim0.008$). The arrows in Figs. 2(a)–2(c) labeled α_z, represent the damping values extracted for uncapped Permalloy films (0 nm Cu). The damping of these films is significantly increased compared with that of films that are capped with 5 nm of Cu. The 10 nm Permalloy film has a damping value of 0.0080 ± 0.0003, whereas that of the 3 nm Permalloy film reaches a value of 0.0136 ± 0.0015. This dramatic change in damping is most likely due to the native oxide layer on top of the uncapped Permalloy, which enhances the spin scattering at the Permalloy/oxide interface.

As an additional prediction of spin-pumping theory, we expect there to be spin accumulation in the Cu layer. To test this prediction we grew a series of Permalloy (5 nm)/Cu(x nm)/Ta (5 nm) multilayer samples. The filled circles in Fig. 3 show the extracted damping parameter for these samples with varying Cu layer thicknesses δ_{Cu}. The filled squares show the extracted damping parameter for the Permalloy (5 nm)/Cu(x nm) samples without a Ta cap layer. The data for samples with a Ta cap show enhanced damping relative to the samples without Ta, even for thin Cu layers. Since spin-orbit coupling is larger in Ta compared with Cu, a spin flip is more likely to occur at the Cu/Ta interface than at the Permalloy/Cu interface. We therefore conclude that the enhanced damping for the Ta capped samples is due to the proximity of the Ta to the Permalloy; the spins that are injected into the Cu diffuse and eventually flip at the Cu/Ta

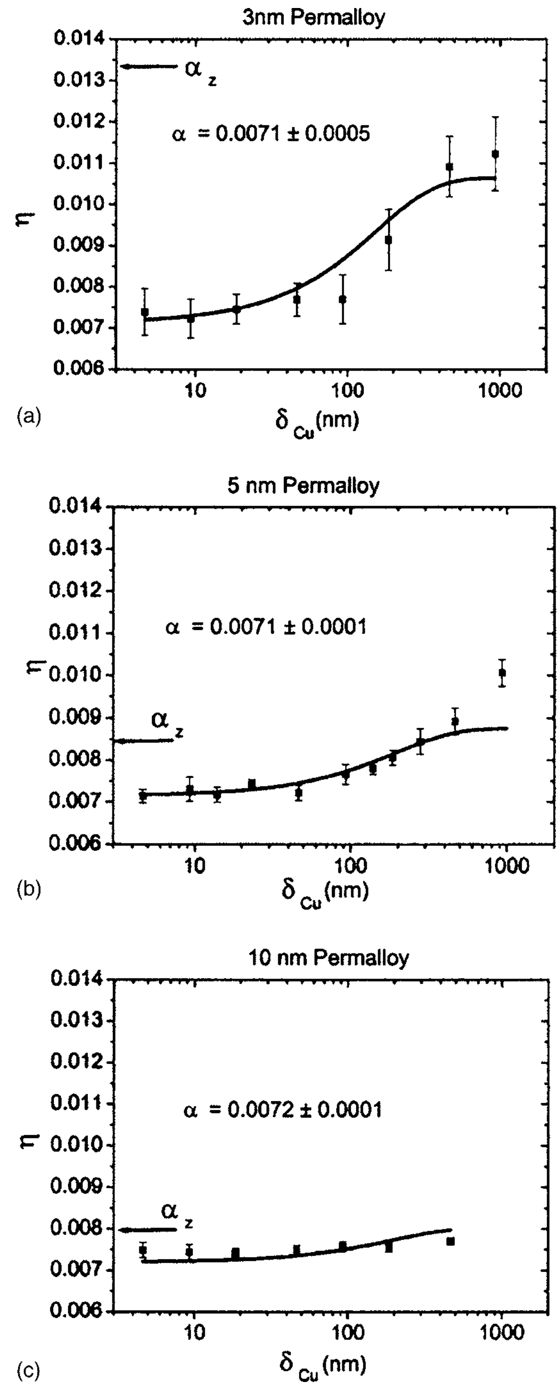

FIG. 2. Damping as a function of Cu layer thickness for three different Permalloy thicknesses: (a) 3, (b) 5, and (c) 10 nm. The solid lines represent a fit of Eq. (3) to the data using $g^{\uparrow\downarrow}A^{-1}=1.5\times10^{15}$ cm^{-2} as determined by the fit in Fig. 1. The intrinsic damping α and diffusion length were fitting parameters. The arrows indicate the damping of the Permalloy films without Cu cap, α_z.

interface. The dashed line represents the fit to a trilayer model that includes a NM/NM interface.[14] From that fit we obtained a mixing conductance for the Cu/Ta interface $g_{NN}A^{-1}=(0.62\pm0.08)\times10^{15}$ cm^{-2}. The damping value for the 5 nm Cu system was excluded from the fit. The data show a small decrease in damping for increasing Cu layer thickness. As the Cu layer thickness increases more spins scatter in the Cu layer due to the finite spin-diffusion length. For Cu layer thicknesses that are longer than the spin-

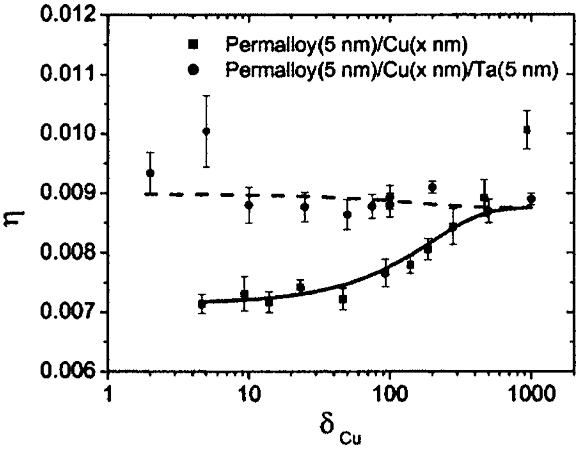

FIG. 3. Damping as a function of Cu thickness for the Permalloy (5 nm)/Cu(x nm) bilayer systems [squares, Fig. 2(b)] and a Permalloy (5 nm)/Cu(x nm)/Ta (5 nm) trilayer system (dots). The solid line represents the fit to the data using Eq. (3). The dashed line represents the fit to the data using a trilayer model with one FM/NM and one NM/NM interface (Ref. 14).

diffusion length, scattering inside the Cu dominates and the Ta cap no longer contributes to the spin-dephasing process.

V. DISCUSSION

The slight increase in damping due to the proximity of the Ta is much smaller than the damping increase observed by Mizukami *et al.* for Pt capping layers.[9] This is in qualitative agreement with Ref. 8, where the increase in damping for a Ta cap was so slight as to not be detectable to within the experimental uncertainty. However, the signal-to-noise ratio for our experiment is sufficient to observe a clear damping increase. Thus, we may conclude that Ta has a weaker spin-scattering efficiency than Pt. Nevertheless, a 5-nm-thin layer of Ta scatters spins with an efficiency as great as >500 nm Cu. Also, a layer of 5 nm Ta may not be enough to act as an efficient spin scatterer. Foros *et al.*[17] observed that a thickness of 10 nm Pd is needed to have Pd act as perfect spin sink. Hence, also a thicker Ta layer may be a more efficient spin scatterer.

If we consider the Permalloy/Cu/air system, we may ask whether the effect of spin-flip scattering at the Cu/air interface will have any effect on the relaxation rate of the Permalloy. As shown by the above experiments we see no damping enhancement due to thin Cu cap layers. However, for Ta capped Permalloy/Cu systems, the damping is significantly higher due to spin flips at the Cu/Ta interface. This leads to the conclusion that the Cu/air interface has a negligible contribution to spin relaxation in the Cu layer.

We can give an estimate for the spin accumulation in Cu layers based on the experimental observations presented. In the case of a very thin magnetic film, excited by a magnetic steplike field pulse, the vector cross product $\hat{m}\times d\hat{m}/dt$ in Eq. (1) is maximum for the out-of-plane component, $[\hat{m}\times d\hat{m}/dt]_z \approx \omega e^{-1/\tau}\sin \omega t$, if $\omega \gg \tau^{-1}$, where τ is the relaxation time of the ferromagnet. Thus, $T_{S,\max}^{\text{pump}} \propto \omega$. The torque acting on the ferromagnet due to the injection of the spin current into the normal metal is

$$T_S^{\text{pump}} \approx \omega \cdot \frac{h}{4\pi} \cdot g^{\uparrow\downarrow} = \frac{h}{2}\frac{\partial N}{\partial t}. \tag{4}$$

Equation (4) represents the number of polarized spins that are injected into the normal metal per unit time. If the backflow of spins from the normal metal to the ferromagnet is neglected, we can calculate the upper limit of the spatially averaged steady-state density of polarized spins in the Cu layer as

$$n = \frac{N}{V} = \frac{\partial N}{\partial t} \cdot \tau_{\text{sf}} \cdot \frac{1}{\delta_{\text{NM}} A} = f \cdot \frac{g^{\uparrow\downarrow}}{A}\frac{\tau_{\text{sf}}}{\delta_{\text{NM}}}, \tag{5}$$

where f is the precession frequency. τ_{sf} is the spin-flip time: $\tau_{\text{sf}}=\ell_s^2/D$. D is given by Einstein's diffusion relation: $D=\sigma/e^2 D_N$, where σ is the conductivity, e is the electron charge, and D_N is the density of states at the Fermi level of the normal metal. This results in a spin-flip time of 9.5 ps assuming a spin-diffusion length of 250 nm in Cu. Since the rate of spin injection is independent of δ_{NM}, the average spin polarization increases with decreasing normal metal layer thickness. If we choose a 10-nm-thick Cu layer, the density of polarized spins becomes 0.039 nm^{-3}. Compared with the total number of polarizable carriers at the Fermi level in Cu (84 nm^{-3}), this results in an upper limit for the estimated spin polarization of only 5×10^{-4}. To calculate the lower spin-polarization limit, we have to take into account the backflow of spins into the ferromagnet. Urban *et al.*[18] presented a model that allows for the calculation of the spin polarization at the FM/NM interface including the backflow of spins into the ferromagnet and the diffusion of polarized spins inside the normal metal. By combining Eqs. (7)–(9) and (13) from Ref. 18, we estimated the lower limit of the spin polarization inside the Cu layer to be 2×10^{-6}, approximately two orders of magnitude smaller than the polarization obtained without backflow. We therefore conclude that spin pumping is not an efficient means of driving spin accumulation in nonmagnetic materials.

VI. CONCLUSION

In conclusion, a systematic increase of the damping parameter η was shown to be the result of spin-flip processes in the Cu layer at thicknesses *larger* than the spin-diffusion length ℓ_s. However, η is not significantly enhanced up to a Cu thickness of 100 nm.

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