Quantum Well State Induced Oscillation of Pure Spin Currents in Fe/Au/Pd(001) Systems

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Spin pumping at the ferromagnetic metal (Fe)/normal metal (Au) interface and the subsequent spin transport in Au/Pd heterostructures is studied using ferromagnetic resonance. The spin pumping induced damping in the Fe/Pd structure is greatly suppressed by the addition of a Au spacer layer in the structure Fe/Au/Pd. The rapid decrease in the interface damping with an increasing Au layer thickness does not correspond to an expectation based on a simple spin diffusion theory in the Au layer. It is possible to account for this behavior by introducing a partial reflection of spin current at the Au/Pd interface. Furthermore, oscillations in the amplitude of spin pumping damping are observed in the Fe/Au/Pd structure as a function of Au thickness for thicknesses less than half the electron mean free path of bulk Au. This new effect indicates a formation of quantum well states in the accumulated spin density in the Au spacer that affect the time irreversible process of spin pumping.

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The generation and transport of pure spin currents is an important topic in spintronics [1,2]. It allows one to transport spin current information without the presence of a net electric charge current (as opposed to spin-polarized currents), thus avoiding problems with capacitances, electromigration, and Joule heating. Spin pumping using microwave excitations or thermal gradients across a ferromagnetic layer (FM) allows one to create pure spin currents. For spintronics applications, understanding the propagation of pure spin currents in heterostructures involving both FMs and normal metal layers (NMs), and their associated interconnects, is vital. Furthermore, with ever shrinking device sizes, it is important to consider how quantum size effects may affect pure spin currents. For the remainder of this Letter, such pure spin currents will be referred to as spin currents.

The generation and transport of spin currents in simple NM systems, such as Au, Ag, and Cu, [3–6] has been extensively studied by ferromagnetic resonance (FMR) in FM/NM and FM/NM/FM structures and is well described by the standard spin pumping and spin diffusion model. Spin pumping leads to interface damping at the FM/NM interface that can be described by Gilbert phenomenology. There are two alternative (and agreeing) theories of spin pumping using a spin dependent scattering potential at the FM/NM interface: (a) the theory by Tserkovnyak *et al.* [7] based on the time dependent scattering matrix formalism [8], and (b) the theory by Šimánek and Heinrich [9] based on the time retarded response of spin dependent scattering. The theory of case (b) points out that the instantaneous response of NM electrons at the FM/NM interface leads to static accumulated spin density in the NM, which results in a time reversible static interlayer exchange coupling that exhibits oscillations with the NM thickness. The oscillation length scales are given by the Fermi surface spanning k vectors in the NM [10]. The time retarded response leads to a time irreversible process resulting in interface damping.

Considering that the interlayer exchange coupling and spin pumping are generated by the same mechanism and spin pumping also generates an accumulated spin density in the NM [11,12], one might expect that in some structures the spin pumping contribution to the interface damping can in principle show some degree of oscillatory behavior as a function of the NM thickness. This has not been reported, which is not that surprising for FM/NM/FM systems because in FMR with a small angle of precession, the spin current is fully absorbed at the FM/NM interfaces as the FMs act as spin sinks [11–13]. However, oscillatory behavior has not been previously found in FM/NM systems, on which we comment later.

It is not obvious that the confined geometry of an ultrathin NM must lead to quantum size effects in spin pumping, considering that this is an irreversible effect. This behavior was predicted in FM/NM structures with changing FM thickness by Mills [14] and Šimánek [15]. However, Zwierzycki *et al.* [16] showed how this effect would only be notable for an extremely thin FM. No oscillations were so far predicted with changing NM thickness, nor have they been experimentally observed. Therefore, it is challenging to explore a possibility to observe an oscillatory dependence of spin pumping with changing NM thickness. Since in our system the quantum confinement is in 1D, we refer to these effects as quantum well effects in the language of Refs. [14,15]. Since no oscillations have been so far reported in simple FM/NM structures, we have decided to investigate spin pumping in

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heterostructures FM/NM1/NM2 where NM2 differs significantly in spin behavior from NM1.

In Pd, thermally excited fluctuations of local spin moment known as paramagnons [17,18] lead to the absorption of spin current in Fe/Pd structures in a different manner than expected using a simple diffusion model. Fe/Pd structures have shown a large increase in the Fe interface damping that saturates with Pd thickness around 10 nm, which is very close to the electron mean free path (~9 nm) in the studied Fe/Pd structures. For these reasons, spin current propagation in Pd was interpreted using a concept of spin decoherence [19,20]. In this respect the structure Fe/Au/Pd represents an interesting FM/NM1/NM2 heterostructure in which the Au behaves as a simple NM with spin relaxation provided by the spin orbit interaction and Pd acts a spin dephaser with spin relaxation provided by the interaction with fluctuating paramagnons. Therefore, Pd represents a spin system that behaves between a FM and a NM and will be referred to as a spin fluctuating normal metal (SFNM).

Spin pumping and spin transport was investigated in Fe/Au(NM)/Pd(SFNM)/Au heterostructures. The thickness of the Pd was larger than the spin decoherence length in Pd, and therefore the returning spin current from the Pd/Au interface was only a small fraction of that entering the Pd. In this case our study was primarily directed towards the effectiveness of spin pumping as a function of the Au interlayer thickness, allowing one to determine the role of the Au(NM)/Pd(SFNM) interface in spin pumping and spin current transport.

The spin moment transfer from a FM to an adjacent NM is governed by the spin mixing conductance [9]

$$g_{\uparrow\downarrow} = \frac{1}{2}\sum_i [|r_{\uparrow,i} - r_{\downarrow,i}|^2 + |t_{\uparrow,i} - t_{\downarrow,i}|^2], \tag{1}$$

where $r_{\uparrow(\downarrow),i}$ and $t_{\uparrow(\downarrow),i}$ are the spin majority (minority) reflection and transmission parameters of a NM electron at the Fermi surface impinging on the FM/NM interface. The pumped spin current (expressed as magnetic current) from the FM/NM interface in a system with diffuse interface scattering of electrons is given by [16]

$$\boldsymbol{I}_{\rm sp} = -\frac{g\mu_B}{4\pi M_s}\mathrm{Re}(\tilde{g}_{\uparrow\downarrow})\left[\boldsymbol{M}\times\frac{\partial \boldsymbol{n}}{\partial t}\right], \tag{2}$$

where the enhanced spin-mixing parameter obtained by subtracting the Sharvin resistance is $\tilde{g}_{\uparrow\downarrow} = 2g_{\uparrow\downarrow}$, g is the Landé factor, M_s is the saturation magnetization, and $\boldsymbol{M}$ is the instantaneous magnetization vector with magnitude M_s and direction $\boldsymbol{n}$. Due to conservation of total magnetic moment, the spin current generated at the FM/NM interface leads to an increased interface Gilbert damping $\alpha_{\rm sp}$ in the FM that is inversely proportional to the film thickness $d_{\rm FM}$ [12,21].

Single crystal GaAs(001)/17Fe/$d_{\rm Au}$Au/50Pd/20Au samples were prepared by means of molecular beam epitaxy, where the integers and $d_{\rm Au}$ refer to the layer thickness in atomic layers (AL) (1Fe = 0.143 nm, 1Au = 0.204 nm, 1Pd = 0.195 nm). The 4×6 surface reconstruction GaAs(001) substrates were prepared as in Montoya *et al.* [22]. The layer thicknesses were monitored by means of oscillations in the intensity of the specular spot at an anti-Bragg RHEED reflection in conjunction with a quartz crystal thickness monitor.

These materials were chosen because (a) they are well lattice matched and therefore their growth is epitaxial and crystalline, which leads to sharp interfaces and reduces lattice defects, and (b) spin pumping and spin transport in GaAs/Fe/Au [6], GaAs/Fe/Ag/Fe [12], and GaAs/Fe/Pd [19] structures have been well studied and understood. The Fe thickness of $d_{\rm Fe} = 17$ AL (2.44 nm) was chosen because $\alpha_{\rm sp}$ is inversely proportional to $d_{\rm FM}$, so the film must be thin and at the same time its Gilbert damping is given by the Fe intrinsic contribution [23].

FMR measurements were carried out in a multimode microwave cavity in a field swept, field modulated configuration, as detailed in Montoya *et al.* [24]. The cavity allowed four resonance frequencies at $f \simeq 27.2$, 31.2, 35.7, and 40.4 GHz. The FMR linewidth was described by

$$\Delta H(\omega) = \alpha\frac{\omega}{\gamma} + \Delta H(0), \tag{3}$$

where α is the dimensionless Gilbert damping parameter, ω is the microwave angular frequency, γ is the absolute value of the gyromagnetic ratio, and $\Delta H(0)$ is the zero frequency line broadening due to long range magnetic inhomogeneities [25], see Fig. 1. The contribution to the damping due to spin pumping is given by $\alpha_{\rm sp} = \alpha_{\rm tot} - \alpha_{\rm 17Fe}$, where $\alpha_{\rm tot}$ is the value of the measured Gilbert damping parameter α in

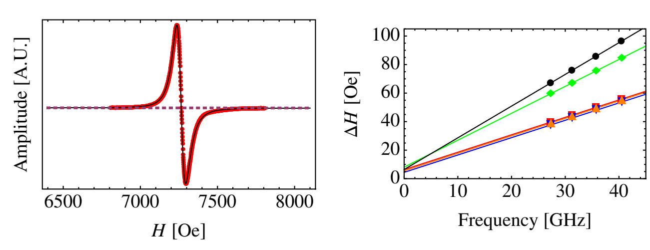

FIG. 1 (color online). Left: 35.6439 GHz FMR data for GaAs/17Fe/20Au. The relevant fit parameters are $H_{\rm FMR} = 7263.1$ Oe and $\Delta H_{\rm FMR} = 49.9$ Oe. Right: FMR linewidth as a function of frequency for (squares, inverted triangles, and triangles) three different growths of GaAs/17Fe/20Au with $\alpha_{\rm tot} = 0.003\,56$. These measurement have been used to define $\alpha_{\rm 17Fe} \equiv 0.003\,56$. (diamonds) GaAs/17Fe/250Au/50Pd/20Au with $\alpha_{\rm tot} = 0.005\,52$ and (circles) GaAs/17Fe/50Pd/20Au with $\alpha_{\rm tot} = 0.006\,50$ showing the increase in damping due to spin pumping $\alpha_{\rm tot} = \alpha_{\rm 17Fe} + \alpha_{\rm sp}$.

Eq. (3) and $\alpha_{17\mathrm{Fe}}$ is the bulk Gilbert damping in the 17Fe layer as determined from measuring GaAs/17Fe/20Au.

Figure 2 illustrates the two main results of this Letter. (A) α_{sp} is found to rapidly decrease from the 17Fe/50Pd/20Au value with the insertion of the Au layer in the 17Fe/d_{Au}Au/50Pd/20Au structures. The 50Pd (9.7 nm) layer was thick enough that the SFNM might be expected to behave as a spin sink [19]. The suppression in α_{sp} saturates around $d_{\mathrm{Au}} = 100$ AL (20 nm). If a SFNM behaved as a spin sink, then a FM/NM/SFNM structure would be analogous to FM1/NM/FM2. Surprisingly however, a rapid decrease in damping was observed in the thickness range of Au where d_{Au} was substantially smaller than the spin diffusion length (290 AL (59 nm) [22]). In this thickness range, one could expect that the spin current generated at the Fe/Au interface is only affected by the spin relaxation process in the Au and one would expect a more gradual decrease in α_{sp} as shown by the red (upper) line in Fig. 2. (B) α_{sp} exhibited an oscillatorylike behavior as a function of d_{Au}. The range $d_{\mathrm{Au}} < 50$ AL where the oscillatory behavior is prominent is significantly less than the electron mean free path in bulk Au [190 AL (38 nm) [26]]. It is interesting to note that these oscillations include two length scales: 8–10 and 2–3 AL, which are very close to those found for the quantum well oscillations of interlayer exchange coupling in Au(001) associated with the spanning k vectors along the belly and neck of the Au Fermi surface [27,28]. This is strong evidence that spin pumping is affected by quantum well states in the ultrathin Au.

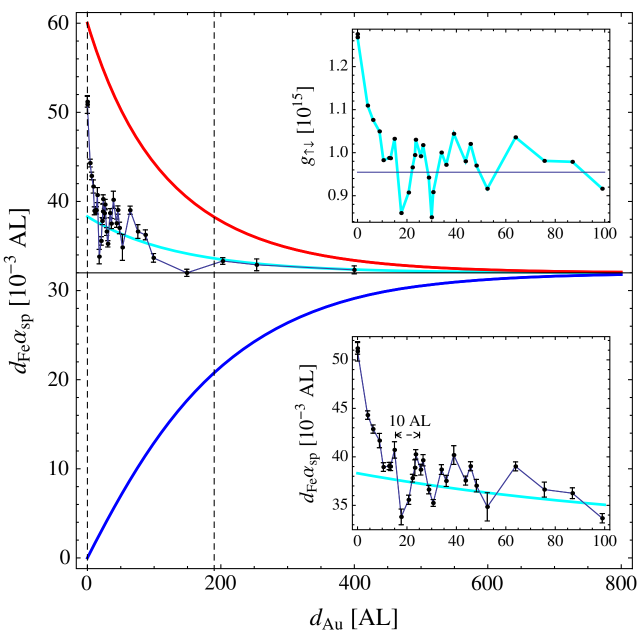

FIG. 2 (color online). Main: spin pumping induced damping α_{sp} multiplied by the Fe thickness in GaAs/17Fe/d_{Au}Au/50Pd structures as a function of Au spacer thickness d_{Au}. The error bars include errors for both α_{tot} and d_{Fe}. Two important effects can be seen. (a) α_{sp} rapidly reached a constant value by $d_{Au} \simeq 100$ AL. (b) An oscillatory behavior in α_{sp} was observed for $d_{\mathrm{Au}} <$ 100 AL. The red line (upper) was calculated using spin diffusion theory for 17Fe/Au/FM (perfect sink) and the blue line (lower) for 17Fe/Au (perfect reflection). The cyan line (central) was obtained by using Eq. (9). The vertical dashed lines represent the electron mean free path in bulk Au. Further details including the spin transport parameters are described in the text. Bottom inset: expanded view of the main figure. Top inset: the evaluated changes in $g_{\uparrow\downarrow}$ by employing Eq. (9).

We present a model to account for the dependence of α_{sp} on d_{Au} described in case (A) above. This model is based on backflow due to accumulated spin density in Pd at the Au/Pd interface. This can be seen as a partial reflection of spin current at the Au/Pd interface. The propagation of spin currents in the structure Fe/Au/Pd is treated as follows.

(a) The spin current propagation in the Au layer is based on standard spin diffusion theory. In the Au layer the spin diffusion equation is given by [29]

$$\frac{\partial \boldsymbol{m}_{\mathrm{Au}}}{\partial t} = D\frac{\partial^2 \boldsymbol{m}_{\mathrm{Au}}}{\partial x^2} - \frac{1}{\tau_{\mathrm{sf}}}\boldsymbol{m}_{\mathrm{Au}}, \tag{4}$$

where $\boldsymbol{m}_{\mathrm{Au}}$ is the accumulated spin density (expressed as magnetic density) in the Au spacer, $D = v_{F,\mathrm{Au}}^2\tau_{\mathrm{m}}/3$ is the spin diffusion constant, $v_{F,\mathrm{Au}}$ is the Fermi velocity, τ_{m} is the electron momentum scattering time, and τ_{sf} is the spin flip scattering time. Since our microwave frequencies are much smaller than the spin flip relaxation rates, Eq. (4) can be approximated as time independent.

The precessing magnetization in the FM leads to a spin current $\boldsymbol{I}_{\mathrm{sp}}$ pumped at the Fe/Au interface. The first boundary condition to Eq. (4) is given by

$$-D\frac{\partial \boldsymbol{m}_{\mathrm{Au}}(x)}{\partial x} = \boldsymbol{I}_{\mathrm{sp}} - \frac{1}{2}v_{F,\mathrm{Au}}\boldsymbol{m}_{\mathrm{Au}}(x)|_{x=0}, \tag{5}$$

where the second term on the right-hand side (rhs) represents a backflow of $\boldsymbol{m}_{\mathrm{Au}}$ at the Fe/Au interface [12].

(b) The spin current propagation in Pd follows a model of effective spin decoherence length described by Foros *et al.* [19]. At the Au/Pd interface there is the forward flow of spin current that is absorbed by the Pd layer

$$\boldsymbol{I}_{\mathrm{abs}} = \frac{1}{2}v_{F,\mathrm{Pd}}\boldsymbol{m}_{\mathrm{Pd}}(x)(1 - e^{-(2d_{\mathrm{Pd}}/\lambda_{\mathrm{dec}})})|_{x=d_{\mathrm{Au}}}, \tag{6}$$

where the term in parentheses accounts for the spin current returned to the Au spacer after reflection from the outer Pd/Au interface, as for finite d_{Pd} the Pd is not a perfect sink [19,20]. Here d_{Pd} is the thickness and λ_{dec} is the spin decoherence length in the Pd (47 AL (9.1 nm) [20]). $\boldsymbol{m}_{\mathrm{Pd}}(d_{\mathrm{Au}})$ is the accumulated spin density in Pd at the Au/Pd interface. The boundary condition concerning the net forward flow of spin current in Au at the Au/Pd interface is given by

$$-D\frac{\partial \boldsymbol{m}_{\rm Au}(x)}{\partial x} = \frac{1}{2}v_{F,\rm Au}\boldsymbol{m}_{\rm Au}(x) - \frac{1}{2}v_{F,\rm Pd}\boldsymbol{m}_{\rm Pd}(x)|_{x=d_{\rm Au}}, \tag{7}$$

where the second term on the rhs represents the backflow spin current of $\boldsymbol{m}_{\rm Pd}$ at the Au/Pd interface. Noting that the net absorbed spin current in Pd, $\boldsymbol{I}_{\rm abs}$, is equal to the net forward flow in Au at Au/Pd, one can set Eq. (6) equal to Eq. (7), and rewrite Eq. (7) in terms of $v_{F,\rm Au}$ and $\boldsymbol{m}_{\rm Au}$

$$-D\frac{\partial \boldsymbol{m}_{\rm Au}(x)}{\partial x} = \frac{\eta}{2}v_{F,\rm Au}\boldsymbol{m}_{\rm Au}(x)|_{x=d_{\rm Au}}, \tag{8}$$

where $\eta = 1 - 1/(2 - e^{-(2d_{\rm Pd}/\lambda_{\rm dec})})$. The solution of Eq. (4) using boundary conditions (5) and (8) leads to the interface damping

$$\alpha_{\rm sp} = \frac{g\mu_{\rm B}}{4\pi M_{\rm s}}\frac{2g_{\uparrow\downarrow}}{d_{\rm Fe}}\left[1 - \frac{v_F}{2} \times \frac{(Dk+\frac{\eta v_F}{2}) + e^{-2kd_{\rm Au}}(Dk-\frac{\eta v_F}{2})}{(Dk+\frac{v_F}{2})(Dk+\frac{\eta v_F}{2}) - e^{-2kd_{\rm Au}}(Dk-\frac{v_F}{2})(Dk-\frac{\eta v_F}{2})}\right], \tag{9}$$

where one can set $v_F = v_{F,\rm Au}$ since the equation is independent of the Fermi velocity in Pd and $k = 1/v_F\sqrt{\tau_{\rm m}\tau_{\rm sf}/3}$ is the inverse spin diffusion length.

The rapid approach of $\alpha_{\rm sp}$ to the asymptotic value, see Fig. 2, presents an intriguing result. For this reason, in addition to the measured spin pumping damping parameter $\alpha_{\rm sp}$, the expected $\alpha_{\rm sp}$ from simple spin diffusion theory is presented using the 17Fe/$d_{\rm Au}$Au (full spin current reflection, lower blue line) and 17Fe/$d_{\rm Au}$Au/40Fe structures (full spin current sink, upper red line), see Fig. 2. The red and blue lines were calculated using $g_{\uparrow\downarrow} = 0.95 \times 10^{15}$ cm^{-2} obtained from the 17Fe/20Au/40Fe(001) structure. In order to get the asymptotic behavior of the measured $\alpha_{\rm sp}$ between the red and blue lines in Fig. 2, $\tau_m = 2.3 \times 10^{-14}$ s and $\tau_{\rm sf} = 23.2 \times 10^{-14}$ s have been used in agreement with the values measured by Montoya *et al.* [22]. This is indeed a rewarding conclusion. The seemingly fast approach to the asymptotic value of $\alpha_{\rm sp}$ can be explained by a simple consequence of the spin backflow from the Pd at the Au/Pd interface, see the cyan line in Fig. 2.

The oscillatory dependence of $\alpha_{\rm sp}$ for $d_{\rm Au} < 100$ AL indicates that in this thickness range the net spin flow across the Fe/Au interface was affected by the presence of spin current collective excitations. Since our model does not include collective excitations in an explicit manner we propose that $g_{\uparrow\downarrow}$ is nonmonotonic and oscillatory in this thickness range. Using the above Pd backflow model (9), one can evaluate $g_{\uparrow\downarrow}$ as a function of $d_{\rm Au}$, see the top inset in Fig. 2.

In the $d_{\rm Au} < 50$ AL region, where the oscillatory behavior is prominent, the total modulation in $\alpha_{\rm sp}$ is ∼20% around the backflow background (cyan line). In comparison for Fe/Au structures in this region, $\alpha_{\rm sp}$ (blue line) is 10 times smaller than that of the Fe/Au/Pd structure (cyan line). Assuming a similar modulation of $\alpha_{\rm sp}$ (∼20%) to that in the Fe/Au/Pd case, the oscillations would be 10 times smaller than those observed in our studies; the effect would be comparable to our error bars. This illustrates why such behavior has not been observed in the Fe/Au structure where there is a perfect spin current reflection at the Au/vacuum interface. It would require very precise measurements of $\alpha_{\rm tot}$ and control of both the Fe and Au thickness, which has not yet been done; therefore, it seems that the partial reflection at the Au/Pd interface was not the only factor allowing us to observe oscillations in $\alpha_{\rm sp}$. In addition, one needs a large enough spin current absorption in the corresponding spin pumping structure. For this reason it may also be challenging to use FM/NM1/NM2 structures where both NM1 and NM2 are simple NMs.

Conclusions.—GaAs/17Fe/$d_{\rm Au}$Au/50Pd/20Au structures were studied by FMR, where $d_{\rm Au}$ is the thickness of the Au spacer in atomic layers. The thickness where spin dephasing nearly saturates is 50Pd and the Pd acts as an efficient spin sink. The spin pumping contribution to the interface damping $\alpha_{\rm sp}$ exhibits new features that were not observed in previous work. (A) $\alpha_{\rm sp}$ rapidly reaches an asymptotic value equivalent to both the 17Fe/$d_{\rm Au}$Au and 17Fe/$d_{\rm Au}$Au/40Fe values. (B) In the thickness range where $d_{\rm Au}$ was smaller than half the mean free path inside the Au layer, $\alpha_{\rm sp}$ exhibited oscillations.

We have presented a phenomenological model based on backflow of the spin density at the Au/Pd interface that was described by well known forward and backward terms in magnetoelectronics. The presence of the oscillatory behavior of $\alpha_{\rm sp}$ represents the fundamentally new results of this work. We have observed the presence of quantum size effects for the time irreversible spin pumping induced damping process for the first time.

This oscillatory behavior is possible considering that the accumulated spin density in spin pumping involves electron transitions from $-\mathbf{k}_F$ to $\mathbf{k}_F$ (required by zero net electron transport) with the spin flip, which is similar to that in interlayer exchange coupling. It looks like the quantum confinement in the accumulated spin density, created by spin pumping in Au, enhances these transitions along the belly and neck areas of the Fermi surface of Au. One should not expect the same oscillatory dependence as observed in interlayer exchange coupling, because one compares this behavior for time reversible and time irreversible processes.

The oscillations in the spin pumping contribution to the interface damping are not within our phenomenological model. A proper theoretical model has to include collective modes of the accumulated spin density in the NM spacer generated by spin pumping, which to our knowledge does not yet exist. The presented data in this Letter show that such modes do exist and their understanding would further advance the understanding of spin pumping and spin transport in heterogeneous structures.

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