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Spin pumping damping and magnetic proximity effect in Pd and Pt spin-sink layers

M. Caminale,^{1,2,*} A. Ghosh,^{2,†} S. Auffret,² U. Ebels,² K. Ollefs,³ F. Wilhelm,⁴ A. Rogalev,⁴ and W.E. Bailey^{1,5,‡}

¹Fondation Nanosciences, F-38000 Grenoble, France

²SPINTEC, Univ. Grenoble Alpes / CEA / CNRS, F-38000 Grenoble, France

³Fakultät für Physik and Center for Nanointegration (CENIDE),

Universität Duisburg-Essen, 47057 Duisburg, Germany

⁴European Synchrotron Radiation Facility (ESRF), 38054 Grenoble Cedex, France

⁵Dept. of Applied Physics & Applied Mathematics,

Columbia University, New York NY 10027, USA

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We investigated the spin pumping damping contributed by paramagnetic layers (Pd, Pt) in both direct and indirect contact with ferromagnetic Ni₈₁Fe₁₉ films. We find a nearly linear dependence of the interface-related Gilbert damping enhancement $\Delta\alpha$ on the heavy-metal spin-sink layer thicknesses t_N in direct-contact Ni₈₁Fe₁₉/(Pd, Pt) junctions, whereas an exponential dependence is observed when Ni₈₁Fe₁₉ and (Pd, Pt) are separated by 3 nm Cu. We attribute the quasi-linear thickness dependence to the presence of induced moments in Pt, Pd near the interface with Ni₈₁Fe₁₉, quantified using X-ray magnetic circular dichroism (XMCD) measurements. Our results show that the scattering of pure spin current is configuration-dependent in these systems and cannot be described by a single characteristic length.

I. INTRODUCTION

As a novel means of conversion between charge- and spin-currents, spin Hall phenomena have recently opened up new possibilities in magneto-electronics, with potential applications in mesoscale spin torques and electrical manipulation of domain walls¹⁻⁹. However, several aspects of the scattering mechanisms involved in spin current flow across thin films and interfaces are not entirely understood. Fundamental studies of spin current flow in ferromagnet/non-magnetic-metal (F/N) heterostructures in the form of continuous films have attempted to isolate the contributions of interface roughness, microstructure and impurities¹⁰⁻¹². ~~magnet/non-magnetic-metal (F/N) heterostructures in the form of continuous films have attempted to isolate the contributions of interface roughness, microstructure and impurities.~~ Prototypical systems in this class of studies are Ni₈₁Fe₁₉/Pt (Py/Pt)^{3,5-7,13-18} and Ni₈₁Fe₁₉/Pd (Py/Pd)^{8,11,14,16,19,20} bilayers. In these systems, Pt and Pd are employed either as efficient spin-sinks or spin/charge current transformers, in spin pumping and spin Hall experiments, respectively. Pd and Pt are metals with high paramagnetic susceptibility and when placed in contact with a ferromagnetic layer (eg. Py, Ni, Co or Fe) a finite magnetic moment is induced at the interface by direct exchange coupling²¹⁻²⁴.

The role of the magnetic proximity effect on interface spin transport properties is still under debate. Zhang *et al.*²⁵ have reported that induced magnetic moments in Pt and Pd films in direct contact with Py correlate strongly reduced spin Hall conductivities. This is ascribed to a spin splitting of the chemical potential and on the energy dependence of the intrinsic spin Hall effect. In standard spin pumping theory²⁶, possible induced moments in N are supposed to be *a priori* included in calculations of the spin-mixing conductance $g^{\uparrow\downarrow}$ of a F|N interface^{27,28}, which tends to be insensitive to their presence.

Recent theoretical works, on the other hand, propose ~~the need of a~~ generalized spin pumping formalisms including spin flip and spin orbit interaction at the F|N interface, in order to justify discrepancies between experimental and calculated values of mixing conductance^{29,30}. At present, it is still an open issue whether and how proximity-induced magnetic moments in F/N junctions are linked to the variety of the spin-transport phenomena reported in literature^{10,17,31}.

Here, we present an experimental study of the prototypical ~~heterostructure system Py/(Pd, Pt) and Py/Cu/(Pd, Pt)~~. The objective of our study is to address the role of proximity-induced magnetic moments in spin pumping damping. To this end, we employed two complementary experimental techniques. X-ray magnetic circular dichroism (XMCD) is an element sensitive technique which allows us to quantify any static proximity-induced magnetic moments in Pt and Pd. Ferromagnetic resonance (FMR) measurements provide indirect information on the spin currents pumped out the Py layer by the precessing magnetization, through the characterization of the Pd, Pt thickness dependence of the interface-related Gilbert damping α . In Fig. 3 (Sec. III B), comparative measurements in Py/Cu/N and Py/N structures show a change of the N thickness dependence of $\Delta\alpha(t_N)$ from an exponential to a linear-like behavior. A change in $\Delta\alpha(t_N)$ indicates a transformation in the spin scattering mechanism occurring at the interface, ascribed here to the presence of induced moments in directly exchange coupled F/N systems. Theoretical works predicted a deviation from a conventional N-thickness dependence when interface spin-flip scattering is considered in the pumping model^{29,30}, ~~but no~~ functional form was provided. For Py/N systems, we find that the experimental thickness dependence cannot be described ~~well~~ by standard models^{16,26,32}, but rather a linear function reproduces the data to a better degree of accuracy, ~~by~~ introducing

84 a different characteristic length. We speculate that the
 85 spatial extent of spin current absorption in F/N systems
 86 shows an inverse proportionality to interfacial exchange
 87 coupling energy, obtained from XMCD, as proposed be-
 88 fore for spin polarized, decoupled interfaces in $F_1/Cu/F_2$
 89 heterostructures¹⁴.

90 II. EXPERIMENT

91 The heterostructures were fabricated by DC mag-
 92 netron sputtering on ion-cleaned Si/SiO₂ substrates in
 93 the form of *substrate/seed/multilayer/cap* stacks, where
 94 Ta(5 nm)/Cu(5 nm) bilayer was employed as *seed*. Ta/Cu
 95 is employed to promote $\langle 111 \rangle$ growth in Py and subse-
 96 quent fcc layers (Pd, Pt), and Ta is known to not affect
 97 the damping strongly^{17,32,33}. Different stacks were grown
 98 as *multilayer* for each measurement.

99 For FMR measurements, we have *multilayer* =
 100 $Py(t_F)/N(t_N)$, $Py(t_F)/Cu(3\text{ nm})/N(t_N)$ with $N = Pd,$
 101 Pt ; an Al(3 nm) film, oxidized in air, was used as
 102 *cap*. The smallest N layer thickness t_N deposited is
 103 0.4 nm, the maximum interdiffusion length observed
 104 for similar multilayers³⁴. Samples with *multilayer* =
 105 $Py(t_N)/Cu(3\text{ nm})$ and no sink layer were also fabricated
 106 as reference for evaluation of the Gilbert damping en-
 107 hancement due to the Pd or Pt layer. The t_N -dependence
 108 measurements of FMR were taken for Py thicknesses $t_F =$
 109 5 and 10 nm. Results from the $t_F = 10\text{ nm}$ data set are
 110 shown in Appendix B. Measurements of the FMR were
 111 carried out at fixed frequency ω in the 4-24 GHz range,
 112 by means of an in-house apparatus featuring an external
 113 magnetic field ~~up to~~ 0.9 T, applied parallel to a copla-
 114 nar waveguide with a broad center conductor width of
 115 350 μm .

116 For XMCD measurements, given the low X-ray absorp-
 117 tion cross-section presented by Pt and Pd absorption
 118 edges, a special set of samples was prepared, consisting
 119 of **Y repeats—with $Y = 20$ or 15 , as specified in the**
 120 **figure captions of the data presented—**per structure in
 121 order to obtain sufficiently high signal-to-noise ratio. In
 122 this case, we have *multilayer* = $[Py(5\text{ nm})/Cu(0, 0.5, 1,$
 123 $3\text{ nm})/N/Cu(0, 0.5, 1, 3\text{ nm})]_Y$, with $N = Pd(2.5\text{ nm})$ and
 124 $Pt(1\text{ nm})$; $Py(5\text{ nm})/Cu(5\text{ nm})/Al(3\text{ nm})$ was deposited
 125 as *cap*. The Pt and Pd thicknesses were chosen to yield
 126 a damping enhancement equal about to half of the res-
 127 pective saturation value (as it will be shown later), i.e.
 128 a thicknesses for which the F/N interface is formed but
 129 the damping enhancement is still increasing. XMCD ex-
 130 periments were carried out at the Circular Polarization
 131 Beamline ID-12 of the European Synchrotron Radiation
 132 Facility (ESRF)³⁵. Measurements were taken in total
 133 fluorescence yield detection mode, at grazing incidence
 134 of 10° , with either left or right circular helicity of the
 135 photon beam, switching a 0.9 T static magnetic field at
 136 each photon energy value (further details on the method
 137 are in Ref.²²). No correction for self-absorption effects
 138 is needed; however XMCD spectra measured at the $L_{2,3}$

139 edges of Pd have to be corrected for incomplete circu-
 140 lar polarization ~~rate of monochromatic X-rays which is,~~
 141 12% and 22% at L_3 and L_2 , respectively. The circular
 142 polarization rate is in excess of 95 % at the $L_{2,3}$ edges of
 143 Pt.

144 III. RESULTS AND ANALYSIS

145 In order to study how the proximity-induced mag-
 146 netic moments ~~may~~ affect the absorption of spin-currents
 147 through interfaces, the static moment induced in Pt, Pd
 148 layers in direct contact with ferromagnetic Py is char-
 149 acterized first, by means of XMCD. The value of the
 150 induced moment extracted for the two Py/N systems is
 151 used to estimate the interfacial exchange energy acting
 152 on the two paramagnets (as described in Sec. III A). Af-
 153 terwards, the dynamic response of the magnetization is
 154 addressed by FMR measurements in Py/N (direct con-
 155 tact) and Py/Cu/N (indirect contact) heterostructures.
 156 From FMR measurements carried out on both configura-
 157 tions as a function of N thickness, the damping en-
 158 hancement due to the presence of the spin-sink layers Pt
 159 and Pd is obtained from the frequency-dependence of the
 160 FMR linewidth. The relation between the static induced
 161 moment and the spin pumping damping is discussed by
 162 comparing the results of the **direct- and indirect-contact**
 163 systems.

164 A. XMCD: Probing the induced magnetic moment

165 In Fig. 1 we show X-ray absorption (XAS) and mag-
 166 netic circular dichroism (XMCD) spectra at the $L_{2,3}$
 167 edges of Pt (top panel) and Pd (bottom panel), taken
 168 on $[Py(5\text{ nm})/Pt(1\text{ nm})]_{20}$ and $[Py(5\text{ nm})/Pd(2.5\text{ nm})]_{20}$,
 169 respectively. Rather intense XMCD signals have been de-
 170 tected at both Pt and Pd $L_{2,3}$ edges, showing unambigu-
 171 ously that a strong magnetic moment is induced by di-
 172 rect exchange coupling at the Py|N interface. The static
 173 induced moment is expected to be ferromagnetically cou-
 174 pled with the magnetization in Py²¹. From the integrals
 175 of XMCD spectra, the induced magnetic moment on the
 176 Pt, Pd sites is determined by applying the sum rules as
 177 in Ref.²² (and references therein). In $[Py/Pd(2.5\text{ nm})]_{20}$,
 178 Pd atoms bear a moment of $0.12\ \mu_B/\text{at}$, averaged over
 179 the ~~full Pd film thickness the whole volume of the vol-~~
 180 ~~ume~~, with an orbital-to-spin ratio $m_L/m_S = 0.05$. In
 181 $[Py/Pt(1\text{ nm})]_{20}$, a magnetic moment $0.27\ \mu_B/\text{at}$ is found
 182 on Pt, comparable to that reported in Ni/Pt epitax-
 183 ial multilayers²³, with a relatively high orbital character
 184 $m_L/m_S = 0.18$, as compared with Pd induced moment.
 185 The large difference in volume-averaged induced moment
 186 per atom comes from the different film thickness, hence
 187 volume, for Pt and Pd. Assuming that the induced mag-
 188 netic moment is confined to the first atomic layers at the
 189 interface with Py^{23,24}, one could estimate $0.32\ \mu_B/\text{at}$ for
 190 Pd and $0.30\ \mu_B/\text{at}$ for Pt³⁶.

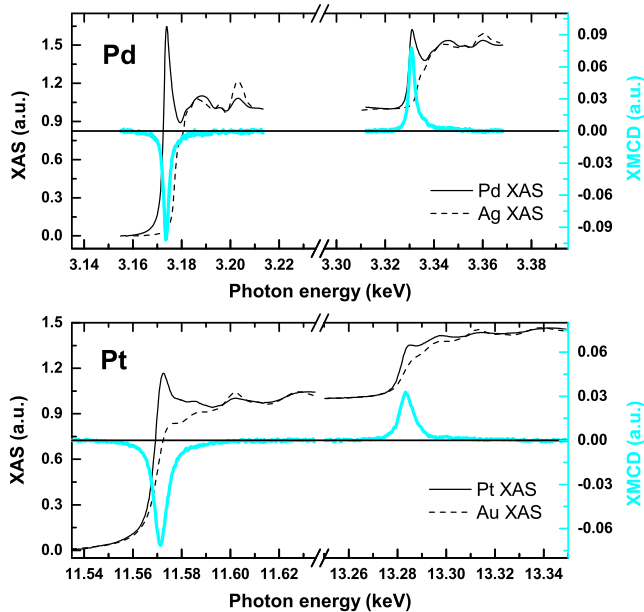


FIG. 1. (Color online) X-ray absorption (XAS, left axis) and magnetic circular dichroism (XMCD, right axis) spectra at the L-edges of Pd (top panel) and Pt (bottom panel) for [Py(5nm)/Pt(1 nm)]₂₀ and [Py(5nm)/Pd(2.5 nm)]₂₀ multilayers. The dashed traces represent XAS spectra at L-edge of Ag and Au used as background of Pd and Pt, respectively, to extract the values of induced magnetic moment reported in Tab. I.

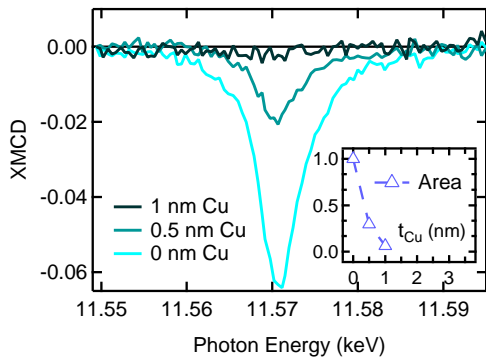


FIG. 2. (Color online) XMCD spectra at the L₃ edge of Pt for [Py(5nm)/Cu(t_{Cu})/Pt(1 nm)]₁₅, with t_{Cu} = 0, 0.5 and 1 nm. *Inset*: the area of the L₃ XMCD peak is plot as a function of Cu thickness.

When a 3 nm thick Cu interlayer is introduced between Py and N, a two orders of magnitude smaller magnetic moment ($0.0036 \mu_B/\text{at}$) was found for 2.5 nm Pd²², while Pt showed a XMCD signal of the order of the experimental sensitivity $\sim 0.5 \cdot 10^{-3} \mu_B/\text{at}$ (see spectra in Fig. 7, Appendix A). In Fig. 2 XMCD spectra at the L₃ edge of Pt are shown for Cu interlayer thicknesses 0, 0.5 and 1 nm. For 0.5 nm Cu the integral of XMCD signal at the L₃ edge shrinks to 30%, while for 1 nm it is reduced to zero within experimental error. This result could be ex-

N	χ_{mol}^{37} [cm ³ /mol] 10^{-4}	S^{37}	N_0 [1/eV·at]	a_{bulk} [nm]	t_N [nm]	$\langle M \rangle$ [μ_B/at]	M_i [μ_B/at]	J_{ex} [meV]
Pd	5.5 ± 0.2	9.3	0.83 ± 0.03	0.389	2.5	0.116	0.32	42
Pt	1.96 ± 0.1	3.7	0.74 ± 0.04	0.392	1.0	0.27	0.30	109

TABLE I. Spin-sink layer N properties in Py/N heterostructures: experimental molar susceptibility χ_{mol} at 20 °C; density of states N_0 calculated from tabulated χ_{mol} ; Stoner parameter S from Ref.³⁷; bulk lattice parameter a ; layer thicknesses t_N ; volume averaged induced magnetic moment $\langle M \rangle$ from XMCD measurement in Fig. 1; interface magnetic moment M_i ³⁶; Py|N interfacial exchange energy per interface atom J_{ex} (Eq. 1).

plained either by a 3d growth of the Cu layer, allowing a fraction of the Pt layer to be in direct contact with Py for Cu coverages of 0.5 nm, or by diffusion of magnetic Ni atoms in Cu on a scale shorter than 1 nm. The film then becomes continuous, and at 1 nm coverage, no direct exchange coupling takes place between Py and Pt layers. For FMR measurements presented in the following section, a 3 nm thick Cu interlayer is employed, reducing also any possible indirect exchange coupling. From the values of induced moments in Pd and Pt, we can make a step forward and estimate the interfacial exchange coupling energies for the two cases. Equating interatomic exchange energy J_{ex} and Zeeman energy for an interface paramagnetic atom, we have (see Appendix C1 for the derivation)

$$J_{ex} = \frac{1}{2} \frac{\langle M \rangle}{\mu_B N_0 S} \frac{t_N}{t_i} \quad (1)$$

where $\langle M \rangle$ is the thickness-averaged paramagnetic moment, N_0 is the single-spin density of states (in eV⁻¹at⁻¹), S is the Stoner factor and $t_i = 2 * a/\sqrt{3}$ is the polarized interface-layer thickness³⁶. The 1/2 factor accounts for the fact that in XMCD measurements the N layer has both interfaces in contact with F. Under the simplifying assumption that all the magnetic moment is confined to the interface N layer and assuming experimental bulk susceptibility parameters for χ_v , we obtain $J_{ex}^{Pd} = 42$ meV for Pd and $J_{ex}^{Pt} = 109$ meV for Pt (results and properties are summarized in Tab. I). Here the difference in estimated J_{ex} , despite roughly equal M_i , comes from the larger Stoner factor S for Pd. A stronger interfacial exchange energy in Pt denotes a stronger orbital hybridization, yielding possibly a higher orbital character of the interfacial magnetic moment in the ferromagnetic Py counterpart²¹. For comparison, we consider the interatomic exchange parameters J_{ex} in ferromagnetic Py and Co, investigated in Ref.¹⁴. J_{ex} is estimated from the respective Curie temperatures T_C , through $J_{ex} \simeq 6k_B T_C / (m/\mu_B)^2$, where m is the atomic moment in μ_B/at (see Appendix C2). Experimental Curie temperatures of 870 K and 1388 K give $J_{ex}^{Co} = 293$ meV for Co and $J_{ex}^{Py} = 393$ meV for Py, which

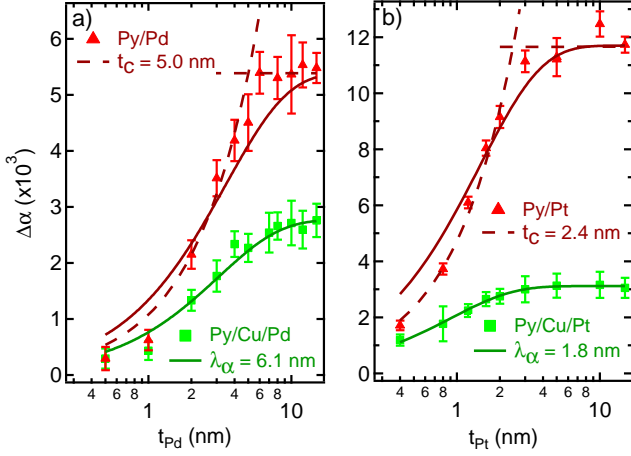


FIG. 3. (Color online) Damping enhancement $\Delta\alpha$, due to pumped spin current absorption, as a function of thickness t_N for Py(5 nm)/N and Py(5 nm)/Cu(3 nm)/N heterostructures, with N = Pd(t_N) (panel a), Pt(t_N) (panel b). Solid lines result from a fit with exponential function (Eq. 2) with decay length λ_α . Dashed lines represents instead a linear-cutoff behavior (Eq. 3) for $t_N < t_c$. Error bars show 95% confidence intervals. Please Notice in panels a, b that the thickness axis is logarithmic.

are of the same order of the value calculated for Pt (details about calculation in Appendix C 2).

In the following, the effect of these static induced moments on the spin pumping damping of the heterostructures characterized will be discussed.

B. FMR: damping enhancement

The main result of our work is now shown in Figure 3. In Fig. 3 the damping enhancement $\Delta\alpha$ is plotted as a function of the spin-sink layer thickness t_N , for Py/Pd, Py/Cu/Pd (panel a) and Py/Pt, Py/Cu/Pt (panel b). The enhancement $\Delta\alpha$ is compared with the damping α of a reference structure Py(5 nm)/Cu, excluding the sink layer N. Each value of α results from established analysis of the linewidth of 11 FMR traces^{13,14}, employing a g -factor equal to 2.09 as a constant fit parameter for all samples.

In Py/Cu/N systems (Fig. 3, green square markers), $\Delta\alpha$ rises with increasing t_N thickness to similar saturation values $\Delta\alpha_0 = 0.0028, 0.0031$ for Pd and Pt, but reached on different length scales, given the different characteristic spin relaxation lengths of the two materials. From the saturation value, an effective mixing conductance $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Cu}/\text{N}) = 7.5 - 8.3 \text{ nm}^{-2}$ is deduced in the framework of the standard spin pumping picture^{13,17,19}, with Py saturation magnetization $\mu_0 M_s = 1.04 \text{ T}$. The fact that the spin-mixing conductance is not material-dependent indicates that similar Cu|N interfaces are formed. The thickness dependence is well described by

N	$g_{\text{eff}}^{\uparrow\downarrow}(\text{Py} \text{Cu}/\text{N})$ [nm ⁻²]	λ_α [nm]	$g_{\text{eff}}^{\uparrow\downarrow}(\text{Py} \text{N})$ [nm ⁻²]	t_c [nm]
Pd	7.5±0.2	6.1±0.6	14±1	5.0±0.4
Pt	8.3±0.2	1.8±0.3	32±1	2.4±0.2

TABLE II. Mixing conductance values extracted from the damping enhancement $\Delta\alpha$ at saturation in Fig. 3, and respective length scales (see text for details).

the exponential function^{14,20}

$$\Delta\alpha(t_N) = \Delta\alpha_0(1 - \exp(-2t_N/\lambda_\alpha^N)) \quad (2)$$

as shown by the fit in Fig. 3a-b (continuous line). As a result, exponential decay lengths $\lambda_\alpha^{\text{Pt}} = 1.8 \text{ nm}$ and $\lambda_\alpha^{\text{Pd}} = 5.8 \text{ nm}$ are obtained for Pt and Pd, respectively.

When the Pt, Pd spin-sink layers come into direct contact with the ferromagnetic Py, the damping enhancement $\Delta\alpha(t_N)$ changes dramatically. In Py/N systems (Fig. 3a-b, triangle markers), the damping saturation values become $\Delta\alpha_0^{\text{Pt}} = 0.0119$ and $\Delta\alpha_0^{\text{Pd}} = 0.0054$ for Pt and Pd, respectively a factor ~ 2 and ~ 4 larger as compared to Py/Cu/N. Within the spin-pumping description, a larger damping enhancement implies a larger spin-current density pumped out of the ferromagnet across the interface and depolarized in the sink.

In Py/N heterostructures, because of the magnetic proximity effect, few atomic layers in N are ferromagnetically polarized, with a magnetic moment decaying with distance from the Py|N interface. The higher value of damping at saturation might therefore be interpreted as the result of a magnetic bi-layer structure, with a thin ferromagnetic N characterized by high damping $\alpha_{\text{high}}^{\text{N}}$ coupled to a low damping $\alpha_{\text{low}}^{\text{F}}$ ferromagnetic Py³⁸. To investigate whether damping is of bi-layer type, or truly interfacial, in Fig 4 we show the t_{F} thickness dependence of the damping enhancement $\Delta\alpha$, for a Py(t_{F})/Pt(4 nm) series of samples. The power law thickness dependence adheres very closely to t_{F}^{-1} , as shown in the logarithmic plot. The assumption of composite damping for synchronous precession, as $\Delta\alpha(t_1) = (\alpha_1 t_1 + \alpha_2 t_2)/(t_1 + t_2)$, shown here for $t_2 = 0.25 \text{ nm}$ and 1.0 nm , cannot follow an inverse thickness dependence over the decade of $\Delta\alpha$ observed. Damping is therefore observed to have a purely interfacial character.

In this case, the mixing conductances calculated from the saturation values are $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Pd}) = 14 \text{ nm}^{-2}$ and $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Pt}) = 32 \text{ nm}^{-2}$. From *ab initio* calculations within a standard spin-pumping formalism in diffusive films^{10,29}, it is found $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Pd}) = 23 \text{ nm}^{-2}$ for Pd and $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Pt}) = 22 \text{ nm}^{-2}$ for Pt. Theoretical spin mixing conductance from a standard picture does reproduce the experimental order of magnitude, but it misses the 2.3 factor between the Py|Pt and Py|Pd interfaces. Beyond a standard pumping picture, Liu and coworkers²⁹ introduce spin-flipping scattering at the interface and calculate from first principles, for ideal interfaces in finite diffusive films: $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Pd}) = 15 \text{ nm}^{-2}$, in excellent agree-

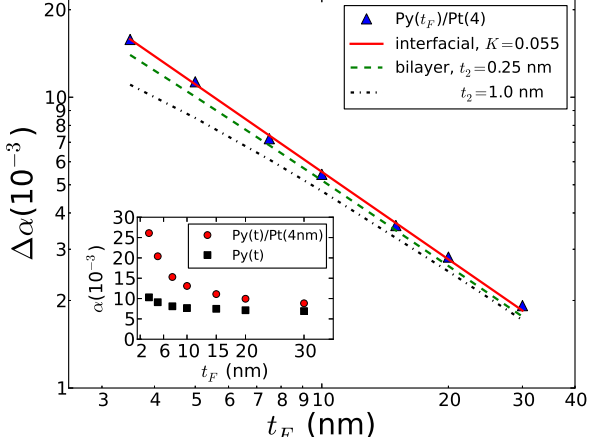


FIG. 4. (Color online) Logarithmic plot of the damping enhancement $\Delta\alpha$ (triangle markers) as a function of the Py layer thickness t_F , in $\text{Py}(t_F)/\text{Pt}(4 \text{ nm})$. Solid and dashed lines represent, respectively, fits according to the spin pumping (*interfacial*) model $\Delta\alpha = Kt_F^{-1}$ and to a $\alpha^{\text{low}}(t_F)/\alpha^{\text{high}}(t_2)$ *bilayer* model, with $t_2 = 0.25, 1.0 \text{ nm}$. *Inset*: Gilbert damping α for $\text{Py}(t_F)$ (square markers) and $\text{Py}(t_F)/\text{Pt}(4 \text{ nm})$ (round markers).

ment with the experimental value here reported for Pd (Tab. II), and $g_{\text{eff}}^{\uparrow\downarrow}(\text{Py}|\text{Pt}) = 25 \text{ nm}^{-2}$. Zhang *et al.*¹⁰ suggest an increase up to 25% of the mixing conductance can be obtained by introducing magnetic layers on the Pt side. The results here reported support the emerging idea that a generalized model of spin pumping including spin-orbit coupling and induced magnetic moments at F/N interfaces may be required to describe the response of heterostructures involving heavy elements.

The saturation value of **damping enhancement** $\Delta\alpha_0$ as a function of the Cu interlayer thickness is shown in Fig. 5 to follow the same trend of the XMCD signal (dashed line), **reported extracted** from Fig. 2. Indeed, it is found that the augmented $\Delta\alpha_0$ in Py/N junctions is drastically reduced by the insertion of 0.5 nm Cu at the $\text{Py}|\text{N}$ interface¹⁷, and the saturation of the $\text{Py}/\text{Cu}/\text{N}$ configuration is already reached for 1 nm of Cu interlayer. As soon as a continuous interlayer is formed and no magnetic moment is induced in N, $\Delta\alpha_0$ is substantially constant with increasing Cu thickness.

The N-thickness dependence of $\Delta\alpha(t_N)$ in Py/N systems before saturation is addressed in the following. In **contrast to** the $\text{Py}/\text{Cu}/\text{N}$ case, the thickness dependence of $\Delta\alpha$ is not anymore well-described by an exponential **saturation, as a fit to Eq. 2, with exponential decay length as only free fit-parameter**, fails to reproduce the increase of $\Delta\alpha$ towards saturation (solid lines in Fig. 3a-b). More rigorous fitting functions employed in spin pumping experiments, within standard spin transport theory^{16,26,32}, **also cannot as well reproduce the experimental data as well** (see Appendix. B). It is worth mentioning that the same change of trend between the two

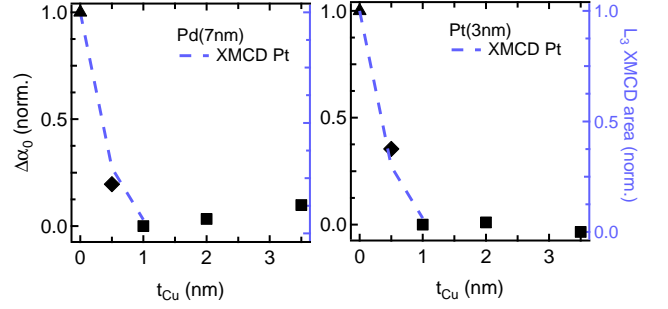


FIG. 5. (Color online) **Damping enhancement** $\Delta\alpha_0$ (left axis, **normalized as** $\Delta\alpha_0(t_{\text{Cu}}) \rightarrow \Delta\alpha_0(t_{\text{Cu}}) - \Delta\alpha_0(1 \text{ nm}) / \Delta\alpha_0(0 \text{ nm}) - \Delta\alpha_0(1 \text{ nm})$), due to spin pumping, as a function of interlayer thickness t_{Cu} for $\text{Py}(5 \text{ nm})/\text{Cu}(t_{\text{Cu}})/\text{N}$ heterostructures, with $\text{N} = \text{Pd}(7 \text{ nm})$, $\text{N} = \text{Pt}(3 \text{ nm})$. The dashed line represents the XMCD signal (right axis) reported from inset in Fig. 2.

configurations was observed for the same stacks with a 10 nm thick Py layer (data shown in Appendix B, Fig. 8). A change of the functional dependence of $\Delta\alpha$ on t_N reflects a change in the spin-depolarization processes undergone by the pumped spin-current, as shown for instance in Ref.³⁰, when interfacial spin-orbit coupling is introduced in the spin-pumping formalism. Experimentally, a *linear* thickness dependence with sharp cutoff has been shown to characterize spin-current absorption in spin-sink layers exhibiting ferromagnetic order at the interface, as reported for $\text{F}_1/\text{Cu}/\text{F}_2(t_{\text{F}_2})$ junctions with $\text{F} = \text{Py}, \text{Co}, \text{CoFeB}$ ¹⁴. Given the presence of ferromagnetic order in N at the interface of F/N structures, the data are tentatively fit with a linear function

$$\Delta\alpha = \Delta\alpha_0 t_N / t_c^N \quad (3)$$

This linear function better reproduce the sharp rise of $\Delta\alpha$ (dashed lines in Fig. 3a-b) and gives **cutoff thicknesses** $t_c^{\text{Pd}} = 5.0 \pm 0.3 \text{ nm}$ and $t_c^{\text{Pt}} = 2.4 \pm 0.2 \text{ nm}$ for **Pd and Pt**, respectively. The linearization is ascribed to the presence of ferromagnetic order in the paramagnetic Pd, Pt spin-sink layers at the interface with the ferromagnetic Py. The linear trend extends beyond the thickness for which a continuous layer is already formed (less than 1 nm), and, especially for Pd, far beyond the distance within the non-uniform, induced moment is confined (**up to 0.9 nm**²⁴). In Ref.¹⁴, the cutoff t_c in F/Cu/F heterostructures is proposed to be on the order of the transverse spin coherence length λ_J in ferromagnetically ordered layers. λ_J can be expressed in terms of the exchange splitting energy J_{ex} ,

$$\lambda_J = \frac{h v_g}{2 J_{ex}} \quad (4)$$

where v_g is the electronic group velocity at the Fermi level. This form, found from hot-electron Mott polarimetry¹, is expressed equivalently for free electrons as $\pi/|k^\uparrow - k^\downarrow|$, which is a scaling length for geometrical

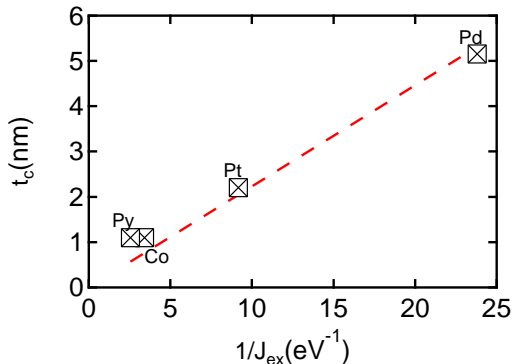


FIG. 6. (Color online) Effect of direct exchange strength on length scale of spin current absorption. Cutoff thickness t_c extracted from the $\Delta\alpha(t_N)$ data in Fig. 3 as a function of reciprocal interfacial exchange energy $1/J_{ex}$ extracted from XMCD in Fig. 1. Labels are given in terms of J_{ex} . The Co and Py points are from Ref. 14.

dephasing in spin momentum transfer². Electrons which enter the spin-sink at E_F do so at a distribution of angles with respect to the interface normal, traverse a distribution of path lengths, and precess by different angles (from minority to majority or *vice versa*) before being reflected back into the pumping ferromagnet. For a constant v_g , it is therefore predicted that t_c is inversely proportional to the exchange energy J_{ex} .

In Figure 6 we plot the dependence of the cutoff thickness t_c^N upon the inverse of the estimated exchange energy J_{ex} (Tab. I), as extracted from the XMCD measurements. A proportionality is roughly verified, as proposed for the transverse spin coherence length across spin polarized interfaces. Under the simplistic assumption that $t_c = \lambda_J$, from the slope of the line we extract a Fermi velocity of $\sim 0.1 \cdot 10^6$ m/s (Eq. 4), of the order of magnitude expected for the materials considered^{39,40}. These data show that, up to a certain extent, the length scale for spin-current scattering shares a common physical origin in ferromagnetic layers and paramagnetic heavy-metals, such as Pd and Pt, under the influence of magnetic proximity effect. This unexpected result is observed in spite of the fact that $F_1/Cu/F_2$ and F/N systems present fundamental differences. In F/N structures, the induced moment in N is expected to be directly exchange coupled with the ferromagnetic counterpart. In $F_1/Cu/F_2$, the magnetic moment in F_2 (off-resonance) are only weakly coupled with the precession occurring in F_1 (in-resonance), through spin-orbit torque and possible RKKY interaction. Magnetization dynamics in N might therefore be expected with its own pumped spin current, though, to the best of our knowledge, no experimental evidence of a dynamic response of proximity induced moments has been reported so far. From these considerations and the experimental findings, counter-intuitively the proximity-induced magnetic moments appear not to be involved in the production of spin current, but rather to contribute

exclusively with an additional spin-depolarization mechanism at the interface.

IV. CONCLUSIONS

We have investigated the effect of induced magnetic moments in heavy metals at Py/Pt and Py/Pd interfaces on the absorption of pumped spin currents, by analyzing ferromagnetic resonance spectra with varying Pt, Pd thicknesses. Static, proximity-induced magnetic moments amount to $\sim 0.3 \mu_B/\text{atom}$ in both Pd and Pt, at the interface with Py, as deduced from XMCD measurements taken at the $L_{2,3}$ edges. We have shown that when the proximity-induced moment in Pt and Pd is present, an onset of a linear-like thickness dependence of the damping is observed, in contrast with an exponential trend shown by Py/Cu/Pd and Py/Cu/Pt systems, for which no induced moment is measured. These results point to the presence of an additional spin-flip process occurring at the interface and to a change of the character of spin current absorption in the ultrathin Pd and Pt paramagnets because of the interfacial spin polarization. The range of linear increase is proposed to be inversely proportional to the interfacial exchange energy in Py/Pt and Py/Pd, inferred from XMCD data.

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Appendix A: XMCD – Pt

In Fig. 7 we report the XMCD spectra taken at the L -edges of Pt on the $[\text{Py}(5\text{nm})/\text{Cu}(t_{Cu})/\text{Pt}(1\text{nm})]_{20}$ multilayer, with $t_{Cu}=0, 3$ nm. The presence of 3 nm-thick Cu interlayer suppresses any proximity induced moment in Pt. The signal, at the margin of detectability, is ascribed to the paramagnetic response of the Pt film in 0.6 T magnetic field and perhaps RKKY coupling from the Py layers, at room temperature.

Appendix B: N-thickness dependence

In order to confirm the results presented in the manuscript, additional sample series with thicker Py layer were fabricated and measured. The experimental results for the 10 nm thick Py layer are shown in Figs 8 and 9 for Pd and Pt, respectively. We have presented the data here, rather than including them with the other

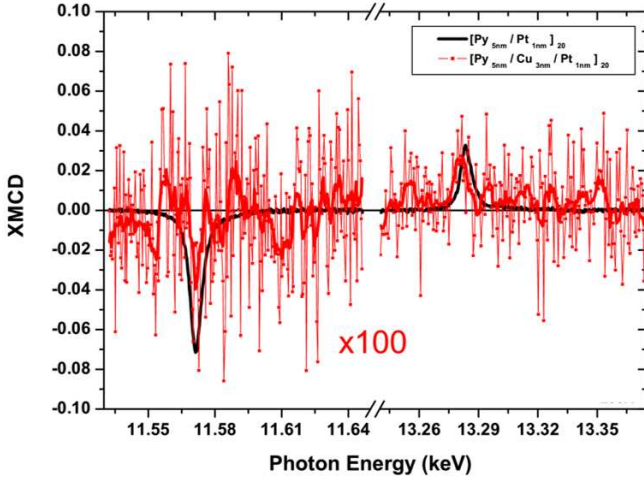


FIG. 7. (Color online) X-ray absorption magnetic circular dichroism (XMCD) spectra at $L_{3,2}$ absorption edges of Pt for $[\text{Py}(5\text{nm})/\text{Pt}(1\text{nm})]_{20}$ (black solid line) and $[\text{Py}(5\text{nm})/\text{Cu}(3\text{nm})/\text{Pt}(1\text{nm})]_{20}$ (red markers; enhanced by a factor 100) multilayers.

plots in Figure 3, to keep the figures from being over-
crowded. As expected when doubling the ferromagnet
thickness, the saturation values $\Delta\alpha_0$ are about half of
those measured for 5 nm Py (Fig. 3). Confirming the
data presented in the manuscript, we again observe a
change of thickness dependence of $\Delta\alpha(t_N)$, from *expo-*
ponential for Py/Cu/N (solid lines; Eq. 2, $\lambda_\alpha = 4.8\text{nm}$
and 1.4 nm for Pd and Pt respectively) to *linear-like* for
Py/N (dashed lines; Eq. 3, $t_c = 5.3\text{nm}$ and 2 nm for Pd
and Pt respectively).

Here we have also considered some alternate fitting
forms based on the standard theory of diffusive spin
transport^{16,26,32}, describing the dependence of $\Delta\alpha$ as

$$\Delta\alpha = \frac{\gamma\hbar}{4\pi M_s t_{FM}} \frac{g^{\uparrow\downarrow}}{1 + g^{\uparrow\downarrow}/g_{ext}} \quad (\text{B1})$$

with the g_{ext} functional form determined by the number
and properties of the adjacent metallic layers—either N
or Cu/N in our case (Eq. 7 in Ref.¹⁶, and Eq. 6 in Ref.³²)

$$g_{ext}^N = g_N \tanh(t_N/\lambda_{sd}^N) \quad (\text{B2a})$$

$$g_{ext}^{Cu/N} = g_{Cu} \frac{g_{Cu} \coth(t_N/\lambda_{sd}^N) + g_N \coth(t_{Cu}/\lambda_{sd}^{Cu})}{g_{Cu} \coth(t_N/\lambda_{sd}^N) \coth(t_{Cu}/\lambda_{sd}^{Cu}) + g_N} \quad (\text{B2b})$$

where $g_x = \sigma_x/\lambda_{sd}^x$, σ_x and λ_{sd}^x are the electrical conduc-
tivity and spin diffusion length of the non-magnetic layer
 x . For the thin Cu layer, we used a resistivity $\rho_{Cu} =$
 $1 \times 10^{-7} \Omega\text{m}$ and a spin diffusion length $\lambda_{sd}^{Cu} = 170\text{nm}$.
For the Pt and Pd layers, two fitting models in which the
conductivity of the films is either constant or thickness
dependent are considered, as recently proposed by Boone
and coworkers¹⁶. The values of conductivity, as taken di-
rectly from Ref.¹⁶, will influence the spin diffusion length
 λ_{sd}^N and spin mixing conductance $g^{\uparrow\downarrow}$ resulting from the

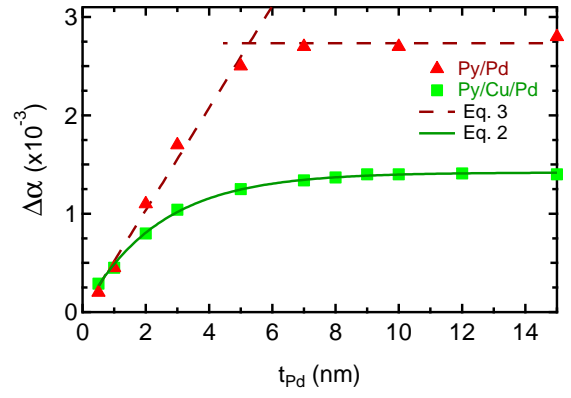
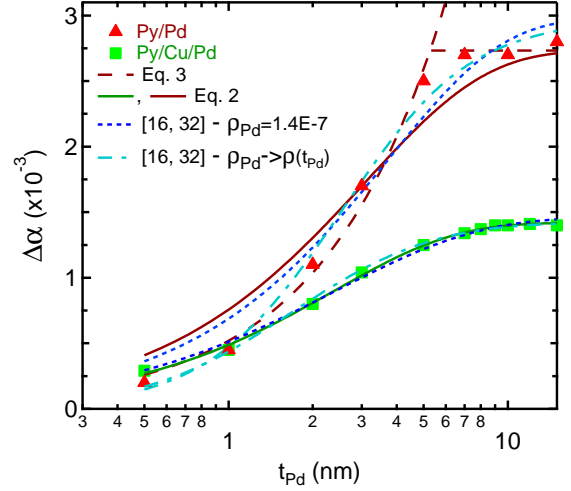


FIG. 8. (Color online) Damping enhancement $\Delta\alpha$, due to pumped spin current absorption, as a function of thickness t_{Pd} for $\text{Py}(10\text{nm})/\text{Pd}$ and $\text{Py}(10\text{nm})/\text{Cu}(3\text{nm})/\text{Pd}$ heterostructures. Solid lines result from a fit with exponential function (Eq. 2) with decay λ_α . Dashed lines represents instead a linear-cutoff behavior (Eq. 3) for $t_{Pd} < t_c$. Short-dash and point-dash traces are fit to the data, employing equations from standard spin transport theory (see text for details)^{16,32}. In the bottom panel, $\Delta\alpha$ is plotted in linear scale for completeness.

fit, but will not affect the conclusions drawn about the
overall trend. When a constant resistivity is used (short-
dash, blue lines), the model basically corresponds to the
simple exponential function in Eq. 2. It nicely repro-
duces the data in the indirect contact case (Py/Cu/N)
for both Pd (Fig. 8) and Pt (Fig. 9), but it fails to fit
the direct contact (Py/N) configuration. When a thick-
ness dependent resistivity of the form $\rho_N = \rho_N^b + \rho_N^s/t_N$
is used (dash-point, cyan lines)¹⁶, in Py/Cu/N systems, no
significant difference with the other functions is observed
for Pt, while for Pd a deviation from experimental trend
is observed below 1.5 nm. In Py/N systems, the fit better
describes the rise at thicknesses shorter than the charac-
teristic relaxation length, while deviates from the data
around the saturation range. It is worth mentioning that
inserting a fictitious layer in between Py and Pt, Pd to ac-

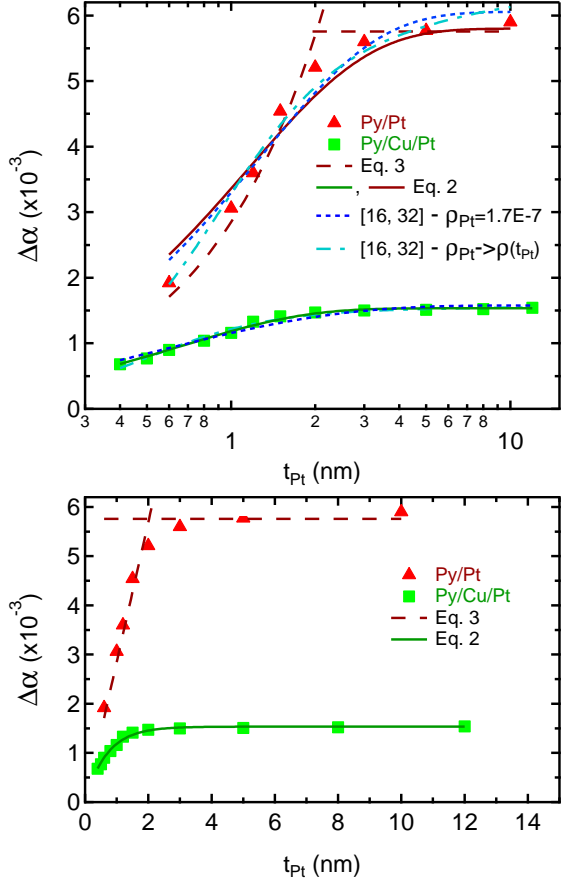


FIG. 9. (Color online) Damping enhancement $\Delta\alpha$, due to pumped spin current absorption, as a function of thickness t_{Pt} for Py(10 nm)/Pt and Py(10 nm)/Cu(3 nm)/Pt heterostructures. Solid lines result from a fit with exponential function (Eq. 2) with decay λ_α . Dashed lines represents instead a linear-cutoff behavior (Eq. 3) for $t_{Pt} < t_c$. Short-dash and point-dash traces are fit to the data, employing equations from standard spin transport theory (see text for details)^{16,32}. In the bottom panel, $\Delta\alpha$ is plotted in linear scale for completeness.

count for an additional interfacial spin-flip δ (as in Ref.¹⁶) did not lead to any improvement in the fit result.

Models from standard spin transport theory cannot satisfactorily describe the experimental data for the direct contact Py/N systems. For this reason a different mechanism for the spin depolarization processes has been proposed, considering the presence of induced magnetic moments in N in contact with the ferromagnetic layer.

Appendix C: Interfacial interatomic exchange

1. Paramagnets

We will show estimates for exchange energy based on XMCD-measured moments in [Py/(Pt, Pd)]_{repeat} su-

perlattices. Calculations of susceptibility are validated against experimental data for Pd and Pt. Bulk susceptibilities will be used to infer interfacial exchange parameters J_{ex}^i .

a. Pauli susceptibility For an itinerant electron system characterized by a density of states at the Fermi energy N_0 , if an energy ΔE splits the spin-up and spin-down electrons, the magnetization resulting from the (single-spin) exchange energy ΔE is

$$M = \mu_B (N^\uparrow - N^\downarrow) = 2\mu_B N_0 S \Delta E \quad (C1)$$

where N_0 is the density of states in $\#/eV/at$, S is the Stoner parameter, and $2\Delta E$ is the exchange *splitting* in eV. Moments are then given in μ_B/at . Solving for ΔE ,

$$\Delta E = \frac{M}{2\mu_B N_0 S} \quad (C2)$$

If the exchange splitting is generated through the application of a magnetic field, $\Delta E = \mu_B H$,

$$\mu_B H = \frac{M}{2\mu_B N_0 S} \quad (C3)$$

and the dimensionless volume magnetic susceptibility can be expressed

$$\chi_v \equiv \frac{M}{H} = 2\mu_B^2 N_0 S \quad (C4)$$

In this expression, the prefactor can be evaluated through

$$\mu_B^2 = 59.218 \text{ eV}\text{\AA}^3 \quad (C5)$$

so with $N_0 [=]/eV/at$, χ_v takes units of volume per atom, and is then also called an atomic susceptibility, in cm^3/at , as printed in Ref³⁷.

b. Molar susceptibility Experimental values are tabulated as molar susceptibilities. The atomic susceptibility χ_v can be contrasted with the mass susceptibility χ_m and molar susceptibility χ_{mol}

$$\chi_{mass} = \frac{\chi_v}{\rho} \quad \chi_{mol} = \frac{ATWT}{\rho} \chi_v \quad (C6)$$

where ATWT is the atomic weight (g/mol) and ρ is the density (g/cm^3). These have units of $\chi_{mass} [=]\text{cm}^3/\text{g}$ and $\chi_{mol} [=]\text{cm}^3/\text{mol}$. The molar susceptibility χ_{mol} is then

$$\chi_{mol} = 2\mu_B^2 N_0 N_A S \quad (C7)$$

in cm^3/mol , where μ_B is the Bohr magneton, and

$$2N_0S = \frac{\chi_{mol}}{N_A\mu_B^2} \quad (C8)$$

$$\langle M \rangle t_p = 2M_p t_i \quad (C14)$$

Eq. C8 provides a convenient method to estimate experimental unknowns, the density of states N_0 and Stoner parameter S , from measurements of χ_{mol} .

Example: for Pd, the low-temperature measurement (different from the room-temperature measurement in Table I) is $\chi_{mol} \sim 7.0 \times 10^{-4} \text{ cm}^3/\text{mol}$. In the denominator, $(N_A\mu_B^2) = 2.622 \times 10^{-6} \text{ Ry} \cdot \text{cm}^3/\text{mol}$, The value $2N_0S$ consistent with the experiment is $266/(\text{Ry-at})$ or $19.6/(\text{eV-at})$. For the tabulated measurement of $S = 9.3$, the inferred density of states is then $N_0 = 1.05/\text{eV-at}$.

c. Interfacial exchange We can assume that the Zeeman energy per interface atom is equal to its exchange energy, through the Heisenberg form

$$\frac{M_p^2}{\chi_v} V_{at} = 2J_{ex}^i s_f s_p \quad (C9)$$

where M_p is the magnetization of the paramagnet, with the atomic moment of the paramagnet m_p in terms of its per-atom spin s_p ,

$$M_p = \frac{m_p}{V_{at}} \quad m_p = 2\mu_B s_p \quad (C10)$$

V_{at} is the volume of the paramagnetic site, $s_{f,p}$ are the per-atom spin numbers for the ferromagnetic and paramagnetic sites, and J_{ex}^i is the (interatomic) exchange energy acting on the paramagnetic site from the ferromagnetic layers on the other side of the interface. Interatomic exchange energy has been distinguished from intraatomic (Stoner) exchange involved in flipping the spin of a single electron. Rewriting Eq C9,

$$\frac{M_p^2}{\chi_v} V_{at} = 2J_{ex}^i s_f \frac{M_p}{2\mu_B} V_{at} \quad (C11)$$

if $s_f = 1/2$, appropriate for $4\pi M_s \sim 10 \text{ kG}$,

$$J_{ex}^i = 2\mu_B \frac{M_p}{\chi_v} \quad (C12)$$

and substituting for χ_v through Eq C4,

$$J_{ex}^i = \frac{M_p}{\mu_B N_0 S} \quad (C13)$$

In the XMCD experiment, we measure the thickness-averaged magnetization as $\langle M \rangle$ in a $[F/N]_n$ superlattice. We make a simplifying assumption that the exchange acts only on nearest-neighbors and so only the near-interface atomic layer has a substantial magnetization. We can then estimate M_p from $\langle M \rangle$ through

where t_i is the polarized interface-layer thickness of N . Since the interface exists on both sides of the N layer, $2t_i$ is the thickness in contact with F . Finally,

$$J_{ex} = \frac{1}{2} \frac{\langle M \rangle t_p}{\mu_B N_0 S t_i} \quad (C15)$$

The exchange energy acting on each interface atom, from all neighbors, is $J_{ex}^{Pt} = 109 \text{ meV}$ for Pt and $J_{ex}^{Pd} = 42 \text{ meV}$ for Pd. Per nearest neighbor for an ideal F/N(111) interface, it is $J^{Py|Pt} = 36 \text{ meV}$ and $J^{Py|Pd} = 14 \text{ meV}$. Per nearest neighbor for an intermixed interface (6 nn), the values drop to 18 meV and 7 meV , respectively.

Since explicit calculations for these systems are not in the literature, we can compare indirectly with theoretical values. Drenner⁴¹ showed that at a (3d)F/(4d)N interface (e.g. Co/Rh), there is a geometrical enhancement in the moment induced in N per nearest-neighbor of F . The $4d$ N atoms near the F interface have larger induced magnetic moments per nn of F by a factor of four. Specific calculations exist of $J^{F|N}$ (per neighbor) for dilute Co impurities in Pt and dilute Fe impurities in Pd⁴². $J^{Fe-Pd} \sim 3 \text{ meV}$ is calculated, roughly independent of composition up to 20% Fe. If this value is scaled up by a factor of four, to be consistent with the interface geometry in the XMCD experiment, it is $\sim 12 \text{ meV}$, comparable with the value for Pd, assuming intermixing. Therefore the values calculated have the correct order of magnitude.

2. Ferromagnets

The Weiss molecular field,

$$H_W = \beta M_s \quad (C16)$$

where β is a constant of order 10^3 , can be used to give an estimate of the Curie temperature, as

$$T_C = \frac{\mu_B g J J (J+1)}{3k_B} H_W \quad (C17)$$

Density functional theory calculations have been used to estimate the molecular field recently^{42,43}, for spin type, the $J(J+1)$ term is substituted with $\langle s \rangle^2$, giving an estimate of

$$T_C = \frac{2 \langle s \rangle^2 J_0}{3k_B} \quad (C18)$$

where $\langle s \rangle$ is the number of spins on the atom as in Eq C10; see the text by Stöhr and Siegmann⁴⁴. $\langle s \rangle$

615 can be estimated from $m = 1.07\mu_B$ for Py and $1.7\mu_B$ for
616 Co, respectively. Then

$$J_0 \simeq \frac{6k_B T_C}{(m/\mu_B)^2} \quad (\text{C19})$$

617 with experimental Curie temperatures of 870 and 1388
618 K, respectively, gives estimates of $J_0 = 293$ meV for Co
619 and $J_0 = 393$ meV for Py.

620 Note that there is also a much older, simpler method.
621 Kikuchi⁴⁵ has related the exchange energies to the Curie
622 temperature for FCC lattices through

$$J = 0.247k_B T_C \quad (\text{C20})$$

623 Taking 12 NN, $12J$ gives a total energy of 222 meV for
624 Py (870 K) and 358 meV for FCC Co (1400K), not too
625 far off from the DFT estimates.

626 *d. Other estimates* The J_0 exchange parameter is
627 interatomic, describing the interaction between spin-
628 clusters located on atoms. Reversing the spin of one of
629 these clusters would change the energy J_0 . The Stoner

630 exchange Δ is different, since it is the energy involved in
631 reversing the spin of a single electron in the electron sea.
632 Generally Δ is understood to be greater than J_0 because
633 it involves more coulomb repulsion; interatomic exchange
634 can be screened more easily by sp electrons.

635 This exchange energy is that which is measured by
636 photoemission and inverse photoemission. Measurements
637 are quite different for Py and Co. Himpfel⁴⁰ finds an
638 exchange splitting of $\Delta = 270$ meV for Py, which is not
639 too far away from the Weiss J_0 value. For Co, however,
640 the value is between 0.9 and 1.2 eV, different by a factor
641 of four. For Co the splitting needs to be estimated by a
642 combination of photoemission and inverse photoemission
643 because the splitting straddles E_F .⁴⁶

644 For comparison with the paramagnetic values of J_{ex}^i ,
645 we use the J_0 estimates, since they both involve a bal-
646 ance between Zeeman energy (here in the Weiss field)
647 and Heisenberg interatomic exchange. Nevertheless the
648 exchange splitting Δ_{ex} is more relevant for the estimate
649 of $\lambda_c = hv_g/(2\Delta_{ex})$. For Py, the predicted value of λ_c
650 from the photoemission value (through $\lambda_c = \pi/|k^\uparrow - k^\downarrow|$)
651 is 1.9 nm, not far from the experimental value of 1.2 nm.

652 * michael.caminale@cea.fr

653 † Present address: Data Storage Institute, Agency for Sci-
654 ence, Technology and Research (A*STAR), Singapore
655 138634

656 ‡ web54@columbia.edu

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739 ³⁶ The values of interface moment M_i in Pt and Pd are calcu- 764 ⁴⁴
740 lated confining the volume-averaged moment to the first 2 765
741 atomic planes at both the Py|N|Py interfaces of the stack. 766
742 Given the bulk (111)-plane distance $a/\sqrt{3}$ (with a as in 767
743 Tab. I), the polarized interface-layer thickness $t_i = 2*a/\sqrt{3}$ 768
744 is therefore defined. Considering the decay of the moment 769
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