Decoherence of Transverse Spin Current in Ferrimagnetic Alloys

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It has been predicted that transverse spin current can propagate coherently over a long distance in antiferromagnetically ordered metals. Here, we determine the coherence length $\lambda_c$ of transverse spin current in ferrimagnetic CoGd alloys across magnetic compensation. Spin pumping measurements reveal that nearly compensated CoGd spin sinks exhibit $\lambda_c \approx 4$ nm, approximately a factor of 3-4 greater than $\lambda_c$ for ferromagnetic metals. Our results confirm partial mitigation of spin dephasing in antiferromagnetically ordered metals, analogous to spin echo rephasing for nuclear and qubit spin systems.

A spin current is said to be coherent when the spin polarization of its carriers (e.g., electrons) is locked in a uniform orientation or precessional phase. The length scale over which spin currents remain coherent underpins various phenomena in magnetic materials [1,2]. For instance, the coherence length $\lambda_c$ of electronic spin current polarized transverse to the magnetization fundamentally impacts spin-transfer torque [3–5]. In ferromagnetic metals (FMs) that are often used in spintronic devices, transverse spin current typically exhibits a short $\lambda_c$ of $\approx 1$ nm [6–8]. As illustrated in Fig. 1(a), transverse spins propagating forward with different wavevectors (sampled over half the Fermi surface) precess by different angles about the uniform exchange field. Within just a few atomic monolayers in the FM, the transverse spin polarization averages to zero, i.e., the spin current dephases [6–9]. The transverse spin angular
momentum lost by the spin current is transferred to the magnetization [3–5], thereby giving rise to spin-transfer torque within a ≈1-nm-thick region (corresponding to λc) near the FM interface [6–8]. Spin-transfer torque in FMs is therefore essentially an interfacial effect, such that its efficiency scales inversely with magnetic thickness [3–5].

Transverse spin currents in antiferromagnetically ordered metals – including antiferromagnetic metals (AFMs) and compensated ferrimagnetic metals (FIMs) – have been predicted to exhibit longer λc [10–13]. As illustrated in Fig. 1(b), the spin current interacts with the staggered antiferromagnetic exchange field whose direction alternates at the atomic length scale. The propagating spins precess in alternating directions as they move from one magnetic sublattice to the next, such that spin dephasing is suppressed over multiple monolayers. This cancellation of dephasing in AFM/FIMs is analogous to spin rephasing by π-pulses (Hahn spin echo method) in nuclear magnetic resonance [14], which has recently inspired several approaches of mitigating decoherence of qubit spin systems [15–17]. In contrast to the short λc in FMs that restricts efficient spin-transfer torque to ultrathin media, the longer λc in AFM/FIMs may enable efficient spin-transfer torque in thicker, more thermally stable magnetic media for high-density nonvolatile memory devices [18,19].

The above idealized picture for extended λc in antiferromagnetically ordered metals (Fig. 1(b)) assumes spin current without any scattering and simple layer-by-layer alternating collinear magnetic order. In real materials, finite scattering and complex magnetic order may reduce λc significantly [18,20,21]. Most experiments on AFMs (e.g., polycrystalline IrMn) indeed show short λc of typically ≈1 nm [8,22–25]. Nevertheless, a recent experimental study utilizing a spin galvanic detection method [26,27] has reported λc >10 nm at room temperature in FIMs of CoTb [18], consisting of antiferromagnetically coupled transition-metal (TM) and rare-earth-metal (RE) magnetic sublattices. The report in Ref. [18] is quite surprising considering the strong spin-orbit coupling of CoTb, primarily from RE Tb with a large orbital angular momentum, which can result in increased spin-flip scattering [28–30] and noncollinear sperimagnetic
order [31–33]. TM and RE elements also tend to form amorphous alloys [31–34], whose structural disorder may result in further spin scattering and deviation from layer-by-layer antiferromagnetic order. It therefore remains a critical issue to confirm whether extended $\lambda_c$ emerges in antiferromagnetically ordered metals, particularly in structurally disordered FIMs.

Here, we experimentally determine $\lambda_c$ at room temperature in a series of amorphous FIM CoGd alloys, which exhibit significantly weaker spin-orbit coupling than CoTb due to the nominally zero orbital angular momentum of RE Gd. We perform broadband resonant spin pumping measurements [8] on NiFe/Cu/CoGd trilayers: a coherent spin current generated by ferromagnetic resonance (FMR) [35,36] in NiFe propagates through the diamagnetic Cu spacer and decoheres in the CoGd spin sink. From the spin sink thickness dependence of the nonlocal Gilbert damping, $\lambda_c$ is quantified in a straightforward fashion [8,23] for CoGd of various compositions. We observe a maximum of $\lambda_c \approx 4$ nm for CoGd close to the magnetic compensation composition. This finding confirms that, even in the presence of substantial structural disorder, the antiferromagnetic order in FIMs can partially mitigate the decoherence of transverse spin current, enabling a factor of 3-4 enhancement in $\lambda_c$ compared to FMs (e.g., pure Co). On the other hand, the observed maximum $\lambda_c$ in CoGd – despite its weaker spin-orbit coupling – is much shorter than $\lambda_c > 10$ nm reported in Ref. [18] for CoTb. This seeming contradiction suggests that the apparent value of $\lambda_c$ may be dependent on experimental details.

Overall, our work takes a crucial step towards understanding the interplay between spin current and antiferromagnetic order.

We deposited spin-valve-like stacks of Ti(3)/Cu(3)/Ni$_{80}$Fe$_{20}$(7)/Cu(4)/Co$_{100-x}$Gd$_x$(t)/Ti(3) (unit: nm) by dc magnetron sputtering on Si/SiO$_2$ substrates. The deposition rate of each layer was calibrated by x-ray reflectivity. The Ti(3)/Cu(3) seed layer promotes the growth of NiFe with low Gilbert damping and minimal inhomogeneous linewidth broadening, whereas the Ti(3) capping layer protects the stack from oxidation. The Cu(4) spacer layer suppresses static exchange coupling between the NiFe and CoGd layers [37]. The diamagnetic Cu spacer also
accommodates spin transport mediated solely by conduction electrons, such that interlayer magnon coupling [13,51,52] does not play a role here.

FIM Co$_{100-x}$Gd$_x$ films with various Gd concentrations ($x$ in atomic %) were deposited by co-sputtering Co and Gd targets at different Gd sputtering powers (resulting in an uncertainty in composition of $\approx \pm 0.5$ at. % Gd), except for Co$_{80}$Gd$_{20}$ and Co$_{70}$Gd$_{30}$ films that were deposited by compositional alloy targets. Our vibrating sample magnetometry results (shown in the Supplemental Material [37]) reveal the magnetic compensation composition for Co$_{100-x}$Gd$_x$ thin films to be $x \approx 22-26$, consistent with prior reports [38,39]. The angular momentum compensation composition is only $\approx 1$ Gd at. % below the magnetic compensation composition, since the $g$-factors of Co and Gd are similar ($g_{\text{Co}} \approx 2.15$, $g_{\text{Gd}} = 2.0$) [40]. We further note that, in contrast to some TM-RE FIMs reported recently [18,19,41–49], CoGd layers in our stack structures do not show perpendicular magnetic anisotropy, i.e., CoGd films here are in-plane magnetized [38,39,50].

To determine how the transverse spin coherence length $\lambda_c$ evolves with CoGd composition across magnetic compensation, we performed broadband FMR spin pumping measurements [8,23,25] on NiFe/Cu/Co$_{100-x}$Gd$_x$ stacks with $x = 0, 20, 22, 23, 25, 28, 30, \text{ and } 100$. The half-width-at-half-maximum linewidth $\Delta H$ of the NiFe layer was measured via field-sweep measurements at microwave frequencies $f = 2-22$ GHz. We note that the FMR response of the NiFe layer is readily deconvoluted from that of pure Co ($x = 0$) [37], and that CoGd did not yield FMR signals above our instrumental background. Thus, as shown in Fig. 2, the Gilbert damping parameter $\alpha$ for the NiFe layer is quantified from the $f$ dependence of $\Delta H$ through the standard linear fit,

$$\mu_0 \Delta H = \mu_0 \Delta H_0 + \frac{h}{g \mu_B} \alpha f,$$

where $g \approx 2.1$ is the Landé $g$-factor of Ni$_{80}$Fe$_{20}$, $\mu_0$ is the permeability of free space, $h$ is Planck’s constant, $\mu_B$ is the Bohr magneton, $\mu_0 \Delta H_0 (< 0.2 \text{ mT})$ is the zero-frequency linewidth attributed
to magnetic inhomogeneity. For NiFe without a spin sink, we obtain $\alpha_{\text{no-sink}} \approx 0.0067$, similar to typically reported values for Ni$_{80}$Fe$_{20}$ [53,54].

A finite thickness of spin sink results in a damping parameter $\alpha_{\text{w/sink}}$ that is greater than $\alpha_{\text{no-sink}}$. For example, the damping increases significantly with just 1 nm of Co (Fig. 2), suggesting substantial spin absorption by the spin sink. By contrast, a stack structure that includes an insulating layer of Ti-oxide before the spin sink does not show the enhanced damping (Fig. 2). This observation is consistent with the Ti-oxide layer blocking the spin current [55,56] between the spin source and spin sink layers. Thus, the enhanced damping $\Delta \alpha = \alpha_{\text{w/sink}} - \alpha_{\text{no-sink}}$ is nonlocal in origin, i.e., due to the spin current propagating through the Cu spacer and decohering in the magnetic spin sink [8,35,36,57]. The decoherence of transverse spin current in the spin sink is then proportional to $\Delta \alpha$.

For a given composition of spin sink, the spin sink thickness dependence of $\Delta \alpha$ allows for quantifying $\lambda_c$. As shown in Fig. 3, $\Delta \alpha$ first increases with spin sink thickness, corresponding to increasing decoherence of transverse spin current. Above a critical spin sink thickness equivalent to $\lambda_c$, $\Delta \alpha$ reaches a saturation value $\Delta \alpha_{\text{sat}}$, as the transverse spin current completely decoheres within the depth defined by $\lambda_c$. We quantify $\lambda_c$ by fitting our experimental results with the linear cutoff model, where the transverse spin current decoheres via dephasing in magnetically ordered spin sinks [6,8,23,58]. The details of our fitting procedure is explained in the Supplementary Material [37]. While pure Gd is paramagnetic at room temperature, we apply the same linear cutoff model to keep the analysis protocol consistent for all spin sink compositions.

As shown in Fig. 3(a), pure Co exhibits a short $\lambda_c$ of 1.6±0.5 nm consistent with previous results on FM spin sinks [8]. By contrast, a significantly longer $\lambda_c$ of 4.3±0.8 nm is obtained for the Co$_{75}$Gd$_{25}$ spin sink (Fig. 3(b)) whose composition is within the magnetic compensation window of $x \approx 22$-26 [37]. With further increase in Gd content, $\lambda_c$ decreases to 2.7±0.7 nm for
Co\textsubscript{70}Gd\textsubscript{30} (Fig. 3(c)) and 2.7±0.3 nm for pure Gd (Fig. 3(d)). These results indicate that \( \lambda_c \) has a nonmonotonic dependence on the Gd concentration.

Indeed, the plot of \( \lambda_c \) versus Gd content (Fig. 4(a)) reveals a peak coinciding with magnetic compensation. This maximum \( \lambda_c \) corresponds to a factor of \( \approx 3 \)-4 enhancement over \( \lambda_c \) in pure FM spin sinks. We have also employed other models to fit the spin sink thickness dependence of \( \Delta \alpha \) (shown in the Supplemental Material [37]); irrespective of the fitting model, we observe a maximum in the characteristic transverse spin coherence length near the compensation composition of FIM CoGd.

Our results are qualitatively consistent with the theoretical prediction that antiferromagnetic order mitigates the decoherence of transverse spin current [10–13,18]. In a nearly compensated FIM CoGd spin sink, the alternating Co and Gd moments of approximately equal magnitude (as qualitatively illustrated by the alternating blue and green vertical arrows in Fig. 1(b)) partially cancel the dephasing of the propagating spins. Transverse spin current in compensated CoGd is therefore able to remain coherent over a longer distance than in FMs, although it does decohere within a finite length scale due to the imperfect suppression of dephasing and the presence of electronic momentum and spin scattering [20].

It is important to note that CoGd alloys – and FIM TM-RE systems in general – are amorphous with no long-range structural order. Instead of the simple layer-by-layer alternating order illustrated in Fig. 1(b), the TM and RE atoms are expected to be arranged in a rather disordered fashion. Considering that disorder and electronic scattering tend to quench transverse spin coherence in antiferromagnetically ordered metals [18,20], it is remarkable that such amorphous FIMs permit extended \( \lambda_c \) at all. We speculate the observed enhancement of transverse spin coherence is enabled by short-range ordering of Co and Gd atoms, e.g., finite TM-TM and RE-RE pair correlations in the film plane (and TM-RE pair correlation out of the film plane) as suggested by prior reports [18,59].
Although our results (Figs. 3 and 4(a)) indicate partial mitigation of spin decoherence in nearly compensated FIM CoGd, we do not observe $\lambda_c$ in excess of 10 nm recently reported for CoTb [18]. One potential explanation is that the CoTb films in Ref. [18] exhibit a higher degree of layer-by-layer ordering than our CoGd films. However, even if that were the case, the strong spin-orbit coupling in CoTb [28–33,60] might induce spin scattering that decoheres transverse spin current within a short length scale. Another consideration is the difference in experimental method for deducing $\lambda_c$. Reference [18] utilizes spin galvanic measurements on Co/Cu/CoTb/Pt stacks: FMR in the Co layer pumps a spin current presumably through the CoTb spacer and generates a lateral dc voltage from the inverse spin-Hall effect in the Pt detector [61]. A finite dc voltage is detected for a range of CoTb alloy spacer thicknesses up to 12 nm, interpreted as evidence that the spin current propagates from Co to Pt even with >10 nm of CoTb in between. However, there could be coexisting voltage contributions besides spin current propagation through CoTb. For example, spin scattering in the CoTb layer could yield an inverse spin-Hall effect, i.e., the reciprocal of the strong spin-orbit torque reported in CoTb [60]. Furthermore, the FMR-driven spin-galvanic measurement could also pick up spin rectification [62–64] and thermoelectric voltages [65,66] from the dynamics of the FM Co layer, which might be challenging to disentangle from the inverse spin-Hall effect in Pt. Figures 4(d) and 5(c) in Ref. [18] indeed show that the spin-galvanic signal drops sharply with the inclusion of a CoTb spacer, independent of its thickness, which might be explained by other mechanisms [62–66] unrelated to spin transport through CoTb.

By contrast, our spin pumping method measures the nonlocal damping that is directly attributed to the decoherence of spin current (Fig. 2). The maximum $\lambda_c$ of $\approx 4$ nm in our work is a conservative measure for how far transverse spin current can remain coherent in amorphous TM-RE FIMs at room temperature. This relatively modest enhancement of $\lambda_c$ means that spin-transfer torque can be equally as efficient for, for example, $\approx 1$-nm-thick and $\approx 4$-nm-thick FIMs;
however, in FIMs much thicker than $\lambda_c$, the spin-transfer torque efficiency scales inversely with the FIM thickness, as confirmed by a recent study [19].

In addition to the length scale for spin decoherence, our spin pumping results also provide insight into the magnitude of transverse spin current absorption by the spin sink. Specifically, the saturated value of the Gilbert damping enhancement $\Delta\alpha_{\text{sat}}$ is related to the efficiency of transverse spin absorption in the spin sink thicker than $\lambda_c$, i.e., the effective spin-mixing conductance $g_{\text{eff}}^{\uparrow\downarrow}$ across the Cu/Co$_{100-x}$Gd$_x$ interface,

$$\Delta\alpha_{\text{sat}} = \frac{g_{\text{eff}}^{\uparrow\downarrow} g_B}{4\pi M_s t_F},$$

where $M_s = 8.0 \times 10^5$ A/m and $t_F = 7.0$ nm for the NiFe spin source. As shown in Figs. 3 and 4(b), the ferrimagnetic CoGd spin sinks examined here ($x = 20$-$30$) exhibit $g_{\text{eff}}^{\uparrow\downarrow}$ slightly lower than that of pure Co and higher than that of pure Gd. Evidently, the magnitude of interfacial transverse spin current absorption (Fig. 4(b)) is uncorrelated with magnetic compensation and, rather, $g_{\text{eff}}^{\uparrow\downarrow}$ appears to decrease with decreasing Co content. The latter observation implies that the available channels for transverse spin current are dominated by the $3d$ bands of the TM Co sublattice.

More generally, it might be expected that transverse spin current interacts more strongly with the Co magnetization (from the spin-split itinerant $3d$ bands near the Fermi level) [42,67] than the Gd magnetization (primarily from the localized $4f$ levels $\approx 7$-$8$ eV below the Fermi level [68,69]) – analogous to magnetotransport phenomena dominated by itinerant $3d$ band magnetism in TM-RE FIMs [39,70]. Our results in Fig. 4(b) indeed point to the $3d$ band electrons of the TM playing a larger role in transverse spin injection across the Cu/CoGd interface. However, we are unable to conclude whether transverse spin current within CoGd interacts more strongly with the TM Co sublattice than the RE Gd sublattice [67]. If the interaction with the Co sublattice is stronger, more Gd would be needed to compensate spin dephasing about the Co magnetization; then, $\lambda_c$ would be maximized at a more Gd-rich composition than the
magnetic compensation composition. Here, any offset between the maximum-\(\lambda_c\) composition and the magnetic compensation composition is obscured by our experimental uncertainty [37]. Much higher precision in film composition and magnetometry would be required to determine the subtle distinct roles of the TM and RE sublattices in spin decoherence.

In summary, we have utilized broadband FMR spin pumping to quantify the coherence length \(\lambda_c\) of transverse spin current in ferrimagnetic CoGd alloys. We obtain a maximum of \(\lambda_c \approx 4\) nm in nearly compensated CoGd, consistent with the antiferromagnetic order mitigating the decoherence (dephasing) of transverse spin current. The observed maximum \(\lambda_c\) constitutes a factor of \(\approx 3-4\) enhancement compared to that for ferromagnetic metals. Such partial spin rephasing by antiferromagnetic order – even in disordered ferrimagnetic alloys at room temperature – demonstrates a spin-echo-like scheme built into the solid to counter spin decoherence. Our finding also points to the possibility of further extending transverse spin coherence in structurally pristine antiferromagnetic metals, thus opening a new avenue for fundamental studies of spin transport in magnetic media.

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FIG. 1. (Color online) Dephasing of coherent transverse spin current excited by ferromagnetic resonance (FMR) in the spin source. The spin current is coherent in the normal metal (NM) spacer layer (indicated by the aligned black arrows), but enters the spin sink with different incident wavevectors (dashed gray lines). (a) In the FM spin sink, the propagating spins accumulate different precessional phases in the ferromagnetic exchange field (red vertical arrows) and completely dephase within a short distance. (b) In the AFM/FIM, the spin current does not dephase completely in the alternating antiferromagnetic exchange field (blue and green vertical arrows), as any precession at one sublattice is compensated by the opposite precession at the other sublattice.
FIG. 2. (Color online) Half-width-at-half-maximum (HWHM) FMR linewidth vs frequency for a stack with a spin sink (NiFe/Cu/Co), stack without a spin sink (NiFe/Cu), and stack with an insulating Ti-oxide spin blocker before the spin sink (NiFe/Cu/TiOx/Co).
FIG. 3. (Color online) Nonlocal damping enhancement $\Delta \alpha$ vs $\text{Co}_{100-x}\text{Gd}_x$ thickness for $x = 0, 25, 30, \text{ and } 100$ Gd at.% spin sink layers. The lower bound, midpoint, and upper bound values of the transverse spin coherence length $\lambda_c$ (saturated damping enhancement $\Delta \alpha_{\text{sat}}$) are indicated by vertical (horizontal) red, black, and blue dashed lines, respectively.
FIG. 4. (Color online) (a) Transverse spin coherence length $\lambda_c$ vs CoGd spin sink composition. (b) Saturated damping enhancement $\Delta \alpha_{sat}$ (proportional to the effective spin-mixing conductance $g_{eff}^{↑↓}$ of the Cu/CoGd interface) vs CoGd spin sink composition. The shaded region indicates the window of composition corresponding to magnetic compensation.
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