Spin-Hall Torques Generated by Rare-Earth (Lanthanide) Thin Films

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We report an initial experimental survey of spin-Hall torques generated by the rare-earth metals Gd, Dy, Ho, and Lu, along with comparisons to first-principles calculations of their spin Hall conductivities. Using spin torque ferromagnetic resonance (ST-FMR) measurements and DC-biased ST-FMR, we estimate lower bounds for the spin-Hall torque ratio, ξSH, of ≈ 0.04 for Gd, ≈ 0.05 for Dy, ≈ 0.14 for Ho, and ≈ 0.014 for Lu. The variations among these elements are qualitatively consistent with results from first principles (density functional theory, DFT, in the local density approximation with a Hubbard-U correction). The DFT calculations indicate that the spin Hall conductivity is enhanced by the presence of the partially-filled f orbitals in Dy and Ho, which suggests a strategy to further strengthen the contribution of the f orbitals to the spin Hall effect by shifting the electron chemical potential.

I. INTRODUCTION

In materials with strong spin-orbit coupling, the spin Hall effect (SHE) allows an applied charge current to generate a transverse spin current1–4. The SHE can be used to apply efficient spin-transfer torques in heavy-metal/ferromagnet bilayers for use in controlling magnetic memory devices5–8. The relative efficiency of different materials in generating spin-Hall torques is characterized by a figure of merit we will call the spin-Hall torque ratio: ξSH = TintθSH = Tint(h/2e)JS/Jc, where Tint is a spin transparency factor for the heavy-metal/ferromagnet interface, θSH is the spin Hall ratio describing the efficiency of spin-current generation within the heavy metal, JS is the spin current density generated within the heavy metal, and Jc is the applied charge current density9. Since in general Tint < 1, we expect ξSH < θSH. Values for ξSH and/or θSH have by now been measured for many materials. In pure elements, the strongest spin Hall effects observed10–12 have been found in the 5d transition metals, specifically Pt (with ξSH ≈ 0.06 for Pt/Permalloy samples9 and 0.15 for Pt/FeCoB)9,13, β-Ta (ξβ-Ta ≈ −0.12)5, and β-W (ξβ-W ≈ −0.30)14. The SHE in these pure metals is believed to be largely intrinsic in nature, arising from the Berry curvature of the metal’s band structure15,16. Work on alloys has shown that extrinsic scattering from defects can also produce a large SHE17–23. For example, for Cu(Bi)24 θCu(Bi) = −0.24 and for Cu(Ir)25 θCu(Ir) = 0.02126.

Here we investigate the strength of the spin-Hall torque generated by a class of elements that has been relatively unexplored, the f-electron lanthanide series or “rare-earth” (RE) metals. Our work is motivated by theoretical predictions that the spin Hall effect in metals with partially full f (l = 3) orbitals is potentially quite large27. In the most naive consideration of the SHE, one might expect that the combination of both large orbital angular momenta and large spins in f-electron atoms might lead to large spin-orbit coupling terms, scaling approximately as ⟨l·s⟩, and consistent with the qualitative trend observed in the 3d, 4d, and 5d transition metals11,28,29. Within this simple ansatz, rare-earths near 1/4 and 3/4 filling of the f orbitals are likely candidates for a strong SHE (Fig. 1). We caution, however, that the details of the material band structure and its associated Berry curvature can be critical, as well, so that one should not expect simply that ξSH ∝ ⟨l·s⟩.

We report both experimental measurements of spin-Hall torque ratios and theoretical calculations within density functional theory (DFT) of the spin Hall effect in four rare-earth metals: Gd, Dy, Ho, and Lu which have the f-level configurations f7 (half-full), f8, f10, and f14 (full), respectively30,31. These metals were chosen due to the range of orbital fillings that they cover, and because of their relative chemical stability and low vapor pressure (relative to the early lanthanides)32. To ensure that our measurements are robust, we report spin torque lead to large spin-orbit coupling terms, scaling approximately as ⟨l·s⟩, and consistent with the qualitative trend observed in the 3d, 4d, and 5d transition metals11,28,29. Within this simple ansatz, rare-earths near 1/4 and 3/4 filling of the f orbitals are likely candidates for a strong SHE (Fig. 1). We caution, however, that the details of the material band structure and its associated Berry curvature can be critical, as well, so that one should not expect simply that ξSH ∝ ⟨l·s⟩.

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ratios derived from two separate types of measurements: (i) based on the amplitude of spin-torque ferromagnetic resonance (ST-FMR) peaks\cite{5,6,33} and (ii) based on the change in the ST-FMR linewidth as a function of an applied DC current\cite{6,33,34}. Our results for Gd, Dy, and Ho should be considered lower bounds on $\xi_{SH}$ because the samples for these materials contained a thin Hf spacer layer between the rare-earth and the ferromagnet to reduce the magnetic damping enough for quantitative ST-FMR experiments. This spacer layer likely reduces the transparency factor $T_{int}$ for spin transmission from the rare-earth to the ferromagnet.

What we find experimentally is that the spin Hall effect is small, as expected, in half-filled ($\xi_{SD}^{Gd} \approx 0.04 \pm 0.01$) and fully-filled ($\xi_{LF}^{Dy} \approx 0.014 \pm 0.002$) f-level configurations, while for the materials with partially-filled f-levels the spin torque ratio can be enhanced, $\xi_{SH}^{Dy} \approx 0.05 \pm 0.01$ and $\xi_{SH}^{Ho} \approx 0.14 \pm 0.02$. The magnitude of the value we find for Ho is among the largest for any element (comparable to Pt and Ta and smaller only than $\beta$-W\textsuperscript{14}). The sign we measure for $\xi_{SH}$ is positive in all four cases (the same as Pt), as expected based on Hund’s rules given the orbital filling. Our results for Gd differ from a recent report by the Beach group in both sign and magnitude\textsuperscript{35}.

Our first-principles calculations suggest that the degree to which f orbitals affect the spin-Hall torque ratio depends on the proximity of the f levels to the Fermi level, $\epsilon_f$. In Lu the f orbitals are well below $\epsilon_f$ such that the spin Hall effect is determined entirely by the contributions from the d orbitals. In Dy and Ho, however, the f levels are sufficiently close to $\epsilon_f$ as to provide an enhanced spin Hall effect. Our calculations suggest that further f-orbital enhancement of $\xi_{SH}$ may be possible by artificially shifting the f-electron density of states closer to $\epsilon_f$, e.g., by alloying.

II. EXPERIMENT

A. Materials Growth and Device Fabrication

For our Lu devices, we used molecular-beam deposition (MBD) to grow multilayer samples consisting of substrate/Fe (5 nm)/Lu (10 nm)/Al (2.5 nm) using a sapphire(0001) substrate. The entire structure was deposited without heating the substrate, i.e., at room temperature. The Al protective cap was then oxidized on exposure to air. Fe was chosen as the magnetic layer because of its availability in the MBD system. This stack ordering is inverted compared to most studies of the SHE in heavy-metal/ferromagnet bilayers, because we found that it was necessary to grow the Fe layer first to produce smooth, continuous films. Attempts to to grow Lu by MBD directly on a sapphire substrate resulted in substantial islanding. For the other rare-earths, we were not able to grow any successful samples by MBD. Growth of Ho directly on sapphire yielded even worse island formation than Lu, and when we attempted to study substrate/Fe/Ho/Al samples grown by MBD the Al did not provide enough protection to keep the Ho sample from oxidizing. This could be directly observed as a progressive color change that propagated inward from the corners of the 10 mm x 10 mm chip to its center over the course of \( \approx 15 \) seconds as soon as the sample was removed from the loadlock of the MBD system.

To surmount this problem, for our Gd, Dy, and Ho samples we switched to DC magnetron sputtering using a high-vacuum sputter system (base pressure \(< 5 \times 10^{-9} \) Torr). The rare-earth layers were sputtered from a 1" target positioned directly beneath the center of the sample plate. Depositions were conducted without heating the substrate, i.e., at room temperature. Sputtering of the RE metals onto a sapphire substrate at room temperature yielded smooth films with RMS roughness < 500 pm by atomic force microscopy. We were then able to grow smooth, continuous films of Permalloy (Py = Ni\textsubscript{80}Fe\textsubscript{20}) on top of the RE followed by a 2 nm Al cap. Unfortunately, when we attempted ferromagnetic resonance measurements on the Dy and Ho devices no resonance could be detected, presumably because the damping was very large. Previous studies of Py films containing RE impurities have found a similar trend in the magnitude of damping as a function of varying the RE element – RE elements for which the orbital angular momentum contribution to the RE moment is non-zero (like Dy and Ho, but not Gd) can greatly increase spin relaxation to the lattice\textsuperscript{36,37}.

To counteract this increased damping, we grew samples with a Hf spacer layer inserted between the RE=Gd,Dy,Ho layer and the Py layer to minimize intermixing and reduce the magnetic damping\textsuperscript{38}. While not necessary, the Gd samples also received this Hf spacer layer to enable us to make robust statements about any observed trend in spin Hall effect. Ultimately, for RE=Gd, Dy, and Ho the multilayer stack we used for the measurements was RE (10 nm)/Hf (7 nm)/Py (5 nm)/Al (2.5 nm) grown at room temperature by sputtering onto (0001) sapphire, with the Al oxidized in air. Most of our measurements were performed with $t_{Hf} = 1.5$ nm, although we did perform some tests with other thicknesses as described below. X-ray diffraction experiments of the sputtered samples indicate that both the RE metal and the Py film are polycrystalline with randomly oriented grains. Control samples with thicker Hf layers (both Ho (10 nm)/Hf (4 nm)/Py (5 nm)/Al (2.5 nm) and Hf (5 nm)/Py (5 nm)/Al (2.5 nm)) exhibited Hf films with notable (0001) fiber texture. Atomic force microscopy indicated smooth films with RMS roughness < 500 pm for all of these multilayer samples.

We used optical lithography and Ar ion milling to define bars 30 \( \mu \)m long by 8 \( \mu \)m wide. To finish device fabrication, we made contact pads (Ti (30 nm)/Pt (250 nm)) via optical lithography and liftoff. There was no color change observed in any of the thin film samples upon exposure to air; all remained visibly shiny and metallic throughout fabrication and processing.
Figure 2. (Color online) Representative ST-FMR resonance curves at 9 GHz for (a) a Gd(10)/Hf(1.5)/Py(5)/AlOₓ sample (numbers in parentheses are thicknesses in nm), (b) a Dy(10)/Hf(1.5)/Py(5)/AlOₓ sample, and (c) a Ho(10)/Hf(1.5)/Py(5)/AlOₓ sample. (d) Representative trace for a Fe(5)/Lu(10)/AlOₓ sample at 11 GHz. The higher frequency used for Lu is due to the larger effective magnetization of the Fe layer in the Fe/Lu as compared to the Py used for all other samples. The sign of the signals generated by the Lu sample can be compared directly to the others despite the “inverted” order of the Lu sample (ferromagnet on the bottom, which would ordinarily cause a sign change), because the sign of the anisotropic magnetoresistance in Fe is also reversed relative to the Py in the other samples. The consistent signs of the symmetric components indicate that all four rare-earth systems exhibit a positive spin Hall effect (the same sign as Pt). The sign of the antisymmetric component of the resonance is reversed in the Ho sample relative to the others.

B. Measurement

We performed ST-FMR measurements using a microwave source power of 10 dBm and a fixed frequency in the range 6-12 GHz. The torques generated by the oscillating microwave-frequency current cause the magnetization orientation \( \hat{m} \) of the sample to precess, thereby producing microwave-frequency resistance oscillations on account of the anisotropic magnetoresistance (AMR) of the ferromagnetic layer (Py or Fe). We measured the DC voltage that results from mixing between this oscillating resistance and the oscillating applied current, while sweeping an applied external magnetic field \( \vec{B} \) in the sample plane at an angle approximately 45° from the current direction to scan through the resonance condition. The DC mixing voltage was detected through a bias tee by using lockin detection with amplitude modulation of the RF signal. The microwave power absorbed by the devices, and hence the amplitude of the microwave current in the sample, were calibrated using a vector network analyzer to measure the device \( S_{11} \) and cabling \( S_{21} \) parameters. Representative ST-FMR traces for each RE are shown in Fig. 2.

The magnetic dynamics during ST-FMR can be modeled using the Landau-Lifschitz-Gilbert-Slonczewski equation:\(^\text{39}\)

\[
\dot{\hat{m}} = -\gamma \hat{m} \times (\vec{B} - (\hat{m} \cdot \hat{z})\mu_0 M_{eff}\hat{z}) + \alpha \hat{m} \times \dot{\hat{m}} + \frac{\gamma h}{2\varepsilon M_s l_{mag} l_{REw}}(\xi_{SH}(\hat{m} \times \hat{\sigma} \times \hat{m}) - \xi_{\perp}(\hat{m} \times \hat{B}_{Oe}))
\]

(1)
where $\gamma$ is the absolute value of the gyromagnetic ratio, $M_{s\text{eff}}$ is the effective magnetization which characterizes the out of plane demagnetization field, $\alpha$ is the Gilbert damping parameter, $h$ is the reduced Planck's constant, $e$ is the magnitude of the charge of the electron, $M_s$ is the saturation magnetization of the ferromagnetic layer, $t_{\text{mag}}$ is the thickness of the ferromagnetic layer, $t_{\text{RE}}$ is the thickness of the RE layer, $w$ is the width of the device, $X$ is the fraction of current that flows through the RE layer determined by measuring the resistivities of the individual layers, $I_{\text{RF}}$ is the RF current through the device as calibrated using a vector network analyzer, $\sigma$ is spin-moment orientation of the spin current generated by the spin Hall effect, $B_{\text{Oe}}$ is the direction of the Oersted field for positive current, $\xi_{\text{SH}}$ is the in-plane spin torque ratio, and $\xi_{\perp}$ is an out-of-plane (field-like) torque ratio due the Oersted field generated by the RF current and potentially other spin-orbit torques. The oscillating resistance mixes with the applied RF current to give rise to an output voltage signal with components at DC and at twice the applied RF frequency. For an in-plane directed external magnetic field the resulting DC component has the form\textsuperscript{5,6,33}\n
$$
V_{\text{mix}} = -\frac{I_{\text{RF}}^2}{4eM_s\omega_t_{\text{mag}}t_{\text{RE}}} \left( \frac{dR}{d\phi} \right) \times 
(\xi_{\text{SH}}F_S(B) + \xi_{\perp}F_A(B))
$$

(2)

$$
F_S = \left( \frac{\omega^2\alpha(2B + \mu_0M_{s\text{eff}})}{(\omega^2 - \omega_0^2)^2 + \alpha^2\gamma^2\omega^2(2B + \mu_0M_{s\text{eff}})^2} \right)
$$

(3)

$$
F_A = \left( \frac{\gamma^2B(2B + \mu_0M_{s\text{eff}})^2 - \omega^2\alpha(2B + \mu_0M_{s\text{eff}})}{(\omega^2 - \omega_0^2)^2 + \alpha^2\gamma^2\omega^2(2B + \mu_0M_{s\text{eff}})^2} \right)
$$

(4)

where $\phi$ is the angle of the magnetic field relative to the current, $\frac{dR}{d\phi}$ is the amplitude of the resistance oscillation of the device from the AMR of the ferromagnetic layer, and $\omega_0$ is the resonance frequency defined as $\omega_0 \equiv \gamma \sqrt{B(2B + \mu_0M_{s\text{eff}})}$.

Our first method to determine the spin torque ratios is based on the amplitude of the ST-FMR signal: $\xi_{\text{SH}}$ is proportional to the amplitude of the symmetric component of the resonance and $\xi_{\perp}$ is proportional to the antisymmetric part. As noted above, we calibrate the microwave current in the sample using vector network analyzer measurements, to enable a quantitative determination of the individual torque ratios. The other materials parameters required to calculate the spin torque ratios from the ST-FMR data are summarized in Table I.

As a second approach to measure the spin-Hall torque efficiencies, we also performed DC-biased ST-FMR measurements on the same devices for the Gd, Dy, and Ho multilayers. (This measurement was not successful for the Fe/Lu due to small signal levels associated with much smaller AMR signals in Fe compared to Py.) The presence of a constant DC bias modifies Eq. 1 by adding both a DC in-plane (spin Hall) torque and a DC out-of-plane torque. The effect of the in-plane torque is to modify the effective Gilbert damping of the system, increasing or decreasing the linewidth of the ST-FMR resonance depending on the sign of the applied DC current. Assuming that the DC bias is sufficiently small that the damping modification is in the linear regime,\textsuperscript{5,33}

$$
\xi_{\text{SH}} = \frac{d\alpha_{\text{eff}}}{dI_{\text{RF}}} \frac{h\omega t_{\text{RE}}M_st_{\text{mag}}(B_0 + \mu_0M_{s\text{eff}}/2)}{\sin\phi}
$$

(5)

where $\alpha_{\text{eff}}$ is the effective damping determined by fitting to the resonance linewidth, $B_0$ is the resonant field, $w$ is the device width, $t_{\text{mag}}$ is the thickness of the magnetic layer, and $I_{\text{RF}} \equiv XI_{\text{RF}}$ is the current through rare-earth layer.

We carried out the DC-biased ST-FMR measurements using field modulation provided by a Helmholtz coil to measure the derivative of the ST-FMR mixing voltage signal. This was necessary to eliminate a large background offset that arises when locking into an amplitude modulated signal due to heating from the RF current. The field modulation allows for high sensitivity even at large (10 mA) applied currents. A representative trace for the damping as a function of current for a Ho multilayer is shown in Fig. 3. In addition to a linear shift of the damping as a function of DC current, we also observe a

| Element | $\mu_0M_s$ (T) | $I_{\text{RF}}$ (mA) | $X_{\text{eff}}$ (mV/mA) |
|---------|----------------|----------------------|---------------------------|
| Gd      | 0.88 ± 0.08    | 6.0 ± 0.3            | 0.25                      |
| Dy      | 0.94 ± 0.09    | 6.3 ± 0.3            | 0.27                      |
| Ho      | 1.1 ± 0.1      | 2.8 ± 0.1            | 0.13                      |
| Lu      | 2.3 ± 0.1      | 9.6 ± 0.2            | 0.07                      |
We used control measurements on substrate/Fe/AlOx to extract ρr values of σr that represent a lower bound on the intrinsic spin Hall conductivity due to spin current attenuation in the Hf spacer or interface spin relaxation, so these values are likely to be useful for applications despite its relatively large spin-Hall torque ratio.

The amplitude of the antisymmetric component of the ST-FMR signals gives additional information about the strength of the “effective field” component of the current-induced torque oriented perpendicular to the sample plane (see Eq. (2)). The Oersted field produced by the applied current contributes to this torque, but we also find a significant contribution from a spin-orbit torque that is oriented opposite to the Oersted field torque. In the Ho samples, the spin-orbit contribution is actually larger than the Oersted field contribution, so that the sign of the antisymmetric ST-FMR component is reversed relative to the other samples [compare Fig. 2c with the other panels in Fig. 2]. We can calculate the Oersted field based on our applied current and the measured resistivities of the sample layers, and then estimate from the amplitude of the antisymmetric ST-FMR component the out-of-plane spin torque ratio, with the results shown in Table II. (The negative signs indicate that the direction is opposite to the torque from the Oersted field.) In all cases the magnitude of ξ⊥SO is at least comparable to the antidamping spin-orbit torque ratio ξSH.

For Ho, which provides the strongest spin-Hall torque contributions (σSH/ρRE) based on this work, along with the measured electrical resistivity (ρRE) of Ho is, however, only slightly enhanced relative to Gd and Dy. This is a consequence of the very high resistivity of our sputtered Ho films (see Table III). Due to this high resistivity, Ho is unlikely to be useful for applications despite its relatively large spin-Hall torque ratio.

Table III. Estimated lower bounds on the spin torque conductivities (σSH/ρRE) based on this work, along with the measured electrical resistivity (ρRE) of each RE film. A literature value for Pt measured in a Pt/Pt bilayer system is included for comparison. The measured value of resistivity for our Hf films is 72 µΩ cm, and for our Py films is 65 µΩ cm.

| Element | tHf (nm) | σSH (h/2e) | ρRE (µΩ cm) |
|---------|----------|-------------|--------------|
| Gd      | 1.5      | 110         | 350          |
| Dy      | 1.5      | 180         | 330          |
| Ho      | 1.5      | 210         | 780          |
| Lu      | 0        | 160         | 87           |
| Pt      | 0        | 3000        | 20           |

For convenience in comparing to theoretical calculations, the strengths of the spin-orbit torques can alternatively be expressed in terms of spin torque efficiencies per unit electric field (or “spin torque conductivity”), σSH/ρRE, where ρRE is the electrical resistivity of the rare-earth. We have included these values in Table III as well as the values of ρRE. For RE=Gd, Dy, and Ho, we determined ρRE via 4-point resistance measurements on control samples of substrate/RE/AlOx. For RE=Lu, we used control measurements on substrate/Fe/AlOx to extract ρLu from the full substrate/Fe/Lu/AlOx bilayer by treating the Fe and Lu layers as parallel resistors. These data are plotted at the conclusion of the paper in Fig. 10b as well. We note that given the factor h/2e in our definition of ξSH there is explicitly a factor of 2 in our definition of σSH that is not used universally in the literature. Just as for our determination of ξSH, our values of σSH will be diminished by any spin attenuation in the Hf spacer or interface spin relaxation, so these values represent a lower bound on the intrinsic spin Hall conductivity within each rare-earth.

We find that the spin-Hall torque ratios for Gd, Dy, and Lu are relatively small. This is to be expected for Gd and Lu because these materials have f-electron occupations f5 (half-full) and f14 (full), respectively, with the consequence that the f-levels are relatively far from the Fermi level and unlikely to contribute any significant Berry curvature. The spin-Hall torque ratio for Ho (f7) is significantly greater. We find for the Ho samples with the 1.5 nm Hf spacer layer (using the average value of the two measurement techniques) that ξSH=0.14±0.03. This is among the largest known values for any pure material, comparable to β-Ta5 and Pt (in Pt/CoFeB samples9,13), and less than only β-W14. This large value is despite the likelihood, as we will discuss below, of significant spin relaxation at the interfaces in the RE samples. The spin-Hall torque conductivity σSH of Ho is, however, only slightly enhanced relative to Gd and Dy. This is a consequence of the very high resistivity of our sputtered Ho films (see Table III). Due to this high resistivity, Ho is unlikely to be useful for applications despite its relatively large spin-Hall torque ratio.

Table II. Measured spin torque ratios for Gd(10)/Hf(1.5)/Py(5)/AlOx, Dy(10)/Hf(1.5)/Py(5)/AlOx, Ho(10)/Hf(1.5)/Py(5)/AlOx and Fe(5)/Lu(10)/AlOx. ξ⊥SO refers to the spin-orbit-generated part of the field-like torque, with the contribution from the Oersted field subtracted.

| Element | tHf (nm) | ξSH | ξ⊥SO |
|---------|----------|-----|-------|
| ST-FMR amplitude |
| Gd      | 1.5      | 0.04±0.01 | -0.04 |
| Dy      | 1.5      | 0.06±0.01 | -0.03 |
| Ho      | 1.5      | 0.16±0.04 | -0.07 |
| Lu      | 0        | 0.014±0.009 | -0.03 |
| DC-biased ST-FMR |
| Gd      | 1.5      | 0.04±0.01 | |
| Dy      | 1.5      | 0.05±0.01 | |
| Ho      | 1.5      | 0.12±0.02 | |

quadratic background, which is likely due to Joule heating. For determination of ξSH, we use only the linear contribution to the current dependence, determined by a least-squares fit.

C. Analysis of Experimental Results

Our results for ξSH from both the ST-FMR amplitude measurements and the DC-biased ST-FMR measurements are shown in Table II for the Gd, Dy, and Ho samples with a 1.5 nm Hf spacer layer, and for the Lu/Fe samples with no Hf spacer. These data are also plotted at the conclusion of the paper in Fig. 10a. We find good agreement between the two types of measurements in the samples for which both measurements could be performed. It is important to note that these results do not correspond to intrinsic values of θSH for each rare-earth due to spin current attenuation in the Hf spacer layer and also due to the likelihood of additional spin relaxation at the interfaces.

For convenience in comparing to theoretical calculations, the strengths of the spin-orbit torques can alternatively be expressed in terms of spin torque efficiencies per unit electric field (or “spin torque conductivity”), σSH/ρRE, where ρRE is the electrical resistivity of the rare-earth. We have included these values in Table III as well as the values of ρRE. For RE=Gd, Dy, and Ho, we determined ρRE via 4-point resistance measurements on control samples of substrate/RE/AlOx. For RE=Lu, we used control measurements on substrate/Fe/AlOx to extract ρLu from the full substrate/Fe/Lu/AlOx bilayer by treating the Fe and Lu layers as parallel resistors. These data are plotted at the conclusion of the paper in Fig. 10b as well. We note that given the factor h/2e in our definition of ξSH there is explicitly a factor of 2 in our definition of σSH that is not used universally in the literature. Just as for our determination of ξSH, our values of σSH will be diminished by any spin attenuation in the Hf spacer or interface spin relaxation, so these values represent a lower bound on the intrinsic spin Hall conductivity within each rare-earth.

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| Pt      | 0        | 3000        | 20           |
from among the four elements, we fabricated a series of devices with Hf spacer thicknesses of 0.5, 1, 1.5, 2, and 4 nm, with the results for $\xi_{SH}$ and $\xi_{SO}$ shown in Fig. 4. Previous experiments from our research group have indicated that the spin Hall effect in Hf is very weak, so we anticipated that both $\xi_{SH}$ and $\xi_{SO}$ would decay to small values as a function of increasing Hf thickness, over a characteristic scale determined by the spin diffusion length in Hf, $\approx 1$–1.5 nm. (We note that other research groups have reported larger in-plane spin-torque ratios from Hf, presumably due to films with different crystal structures or different impurities, but our sputter chamber has always yielded negligible spin-torque ratios for Hf films.) As a function of increasing Hf thickness we find that $\xi_{SO}$ behaves consistently with our expectation but, surprisingly, $\xi_{SH}$ does not. Instead, $\xi_{SH}$ decreases only slightly (by about 30%) as the Hf thickness is increased from 0.5 nm to 4 nm. In contrast, control samples consisting of Hf(10 nm)/Py(5 nm)/AlO$_x$ with no RE layer and with $t_{HF} = 1.5$ nm and 5 nm did confirm a small value of $\xi_{SH}$ for Hf (Fig. 4), consistent with previous results from our group.

We checked using x-ray diffraction whether there was any significant structural change for the Hf within the Ho(10 nm)/Hf(4 nm)/Py(5 nm)/AlO$_x$ samples compared to the Hf(5 nm)/Py(5 nm)/AlO$_x$ control, and could observe no difference. We therefore suggest that the most likely explanation of of the unexpected behavior shown in Fig. 4, the relatively large values of $\xi_{SH}$ for thick Hf spacers in the Ho/Hf/Py/AlO$_x$ samples, is that the Ho and Hf layers may not be well-separated. For example, a thin layer of Ho may migrate preferentially to the Py interface even for relatively thick Hf, or Ho impurities may exist within the Hf and contribute an extrinsic spin Hall effect. Given the weak dependence of $\xi_{SH}$ as the Hf thickness is reduced from 1.5 nm to 0.5 nm (which should be well below the spin diffusion length), we still expect the contribution from within the RE layer to dominate the spin Hall torque for the data in Fig. II, even if there is some additional extrinsic spin Hall contribution. Still, given this uncertainty with respect to the effect of the Hf spacer layer, we will not attempt in this initial survey to extrapolate our results to zero Hf thickness, but will simply state the measured spin-torque ratios for each of our samples. It is important to note that we are able to rule out any significant contribution to the spin-orbit torques from an interfacial effect at the Py/AlO$_x$ or the Hf/Py interfaces given the absence of any measurable spin-orbit torque in the Hf(5 nm)/Py(5 nm)/AlO$_x$ control sample.

The Ho samples with different Hf spacer thickness can also be used to analyze how the magnetic damping of the ferromagnetic layer is affected by the spacer. We discuss these data in an appendix.

![Figure 4](image.png)

**Figure 4.** (Color online) Experimental estimates of the in-plane spin Hall (blue circles) and out-of-plane effective field (red squares) torque ratios in the Ho (10 nm)/Hf(t$_{HF}$ nm)/Py (5 nm)/AlO$_x$ multilayer structure using ST-FMR. For comparison, we also plot the in-plane torque ratios for measurements of a Hf (t$_{HF}$ nm)/Py (5 nm)/AlO$_x$ multilayer for t$_{HF}$ = 1.5 nm and 5 nm.

### III. THEORY CALCULATIONS FROM FIRST-PRINCIPLES

#### A. Theory

The intrinsic spin Hall conductivity matrix elements, $\sigma_{\alpha,\beta}$, can be calculated using the Kubo formula as:

$$\sigma_{\alpha,\beta}^\alpha = -\frac{2e}{h} \int_{BZ} \frac{d^3k}{(2\pi)^3} \Omega_{\alpha,\beta}^\alpha(k)$$

(6)

$$\Omega_{\alpha,\beta}^\alpha(k) = -2\text{Im} \left[ \sum_n f_n(k) \sum_{m \neq n} \frac{j_{\alpha,mn}^z v_{\beta,mn}}{(\omega_{mn} - \omega_n)^2} \right]$$

(7)

$$\hat{j}_{\alpha}^z = \frac{1}{2} \{ \hat{s}, \hat{v}_{\alpha} \}$$

(8)

where $\hat{j}_{\alpha}^z$, $\hat{s}$ and $\hat{v}$ are the spin current, spin, and velocity operators, respectively. We calculate these quantities for Gd, Dy, Ho, and Lu using first-principles, density-functional theory (DFT) in both the local density approximation (LDA) and the local density plus Hubbard U approximation (LDA+U) to the exchange-correlation functional. We considered only the intrinsic contribution to spin Hall conductivity, thereby ignoring possible contributions from extrinsic impurity scattering.

Because of the hexagonal close packed (HCP) crystal structure of the the rare-earths studied, the intrinsic spin Hall conductivity is predicted to be anisotropic. Since the samples used in experiment are polycrystalline with random orientation, we expect that the experimentally measured spin Hall conductivities are averaged over an isotropic distribution of all orientations of the crystal. To capture this theoretically, we present the average of $\sigma_{\perp,\perp}^{x,y}$, $\sigma_{\perp,z}^{x,y}$, and $\sigma_{\perp,\perp}^{x,z}$, which we call $\sigma_{\text{DFT}}^{x,y}$. Furthermore, we note that the quantity calculated theoretically is not...
identical to the spin Hall torque conductivity $\sigma_{\text{exp}}^{\text{SH}}$ measured experimentally. As mentioned above, the experimentally determined value for $\sigma_{\text{exp}}^{\text{SH}}$ is a lower bound as it will be reduced by any less-than ideal spin transmission through the sample interfaces and the Hf spacer layer in the real samples. The calculated spin Hall conductivity $\sigma_{\text{DFT}}^{\text{SH}}$ does not include any details about transmission of spin current into a ferromagnet and represents the case of perfect spin transmission.

B. DFT details and approach

Rare-earth metals pose a significant challenge for first-principles theories due to the degree of localization of the $4f$ electrons. To investigate this issue, we constructed two different sets of norm conserving, fully-relativistic pseudopotentials (PsP’s) in the local density approximation (LDA), following Perdew and Wang\textsuperscript{43,44}. These pseudopotentials were constructed using the Atomic Pseudopotential Engine (APE)\textsuperscript{45}. The pseudopotentials generated were benchmarked against the fully-relativistic all-electron potential.

One set of PsP’s posits that $4f$ electrons are highly localized, and as such do not affect the electronic properties of rare-earths\textsuperscript{46,47}. In this case, the $4f$ states are frozen in the core of the PsP and the valence only contains the $6s$, $5d$ and $5p$ electrons. We refer to calculations using these PsP’s, which were performed within the LDA, as ‘f-in-core’. The second set of PsP’s are constructed with the $4f$ states in the valence such that they can in principle affect the electronic properties\textsuperscript{48}. We refer to calculations using these PsP’s as ‘f-in-band’. These were performed within the LDA+U. Since the appropriate values of the Hubbard $U_f$ parameter are uncertain\textsuperscript{49,50}, we perform such calculations for a range of $U_f$ values.

We performed the calculations using Quantum ESPRESSO\textsuperscript{51}. Convergence of total energy of our structures required a plane-wave energy cut off of 50 Ry for the ‘f-in-core’ PsP’s and 290 Ry for the ‘f-in-band’ PsP’s. A $k$ mesh of $15 \times 15 \times 10$ was used for the self-consistent part of our DFT calculations to obtain electronic ground states. The crystal structures for Gd, Dy, Ho and Lu are in all cases hexagonal close-packed (hcp)\textsuperscript{52}. We fully relax all structures.

Calculating the spin Hall conductivity accurately requires a very dense $k$ mesh. To circumvent this issue we mapped our DFT ground state wavefunctions onto a maximally localized Wannier Function basis using WANNIER90\textsuperscript{53}. We extracted the relevant matrices for calculation of spin Hall conductivity, including the velocity matrix $\hat{v}$ and spin matrix $\hat{s}$ of the system. Finally, we employed an adaptive $k$ mesh strategy, inspired by the anomalous Hall functionality in WANNIER90\textsuperscript{54}, to calculate the spin Hall conductivity matrix.

![Figure 5. (Color online) For the ‘f-in-core’ picture: (a) Band structure for Lu (with Fermi energy $\epsilon_F$ at 0 eV) and spin Berry curvature $\Omega_{\alpha,\beta}(k)$ (b) calculated spin Hall conductivities.](image)

Results of the spin Hall conductivity calculations within the ‘f-in-core’ picture for Gd, Dy, Ho, and Lu are shown in Fig. 5b, which shows a monotonic increase in $\sigma_{\text{DFT}}^{\text{SH}}$ with atomic number $Z$. This trend can be understood by first noting that all four elements have the same crystal structure (hcp) and, within the ‘f-in-core’ picture, have the same electron filling, i.e., $6s^25d^1$, resulting in a very similar electronic structure. Therefore, the monotonic increase in spin Hall conductivity from Gd to Lu is consistent with an increase in the strength of spin-orbit coupling, a result shown previously by Ref. 55 for Pt.

To understand the origin of the spin Hall conductivity in the ‘f-in-core’ picture we focus on the electronic structure of Lu, shown in Fig. 5a, as representative. We find that there are $5d$, $6s$ bands near the Fermi level $\epsilon_F$, with avoided band crossings between the $\Gamma$ and $K$ points. These avoided crossings are the primary source of the
spin Berry curvature $\Omega_{\alpha,\beta}^s(k)$, also plotted in Fig. 5a, which upon integration over all occupied $k$-states gives the spin Hall conductivity $\sigma_{DFT}^s$. These features of the electronic structure do not vary for the different elements within the ‘f-in-core’ picture.

D. Spin Hall conductivity in the ‘f-in-band’ picture

For ‘f-in-band’ calculations, we included the $4f$ electrons in the variational density function and applied a Hubbard potential $U_f$ to account for correlation effects. The values of the Hubbard parameter for Gd, Dy, Ho and Lu were taken to be 4.6, 5.0, 4.9, and 5.5 respectively. (We discuss the consequences of assuming different values of $U_f$ below.) To highlight the effect of $4f$ electrons on the electronic structure we will compare and contrast Lu and Ho. This is because, from among the four elements we examined, Lu with its full $4f$-shell is expected to display the least contribution of $4f$ electrons to the band at the Fermi level, while Ho is expected to display the most. Dy has a larger $U_f$ than Ho, and Gd has a more stable $f^{14}$ shell filling, reducing the expected presence of $4f$ bands near $\epsilon_F$ as compared to Ho.

We first focus on Lu, for which the projected density of states and electronic structure are shown in Figs. 5a and 5b, respectively. The electronic structure of Lu in this ‘f-in-band’ picture (Fig. 6b) is very similar to the one obtained in the ‘f-in-core’ picture (Fig. 5a). We restrict the range of the ‘f-in-band’ figures to 1 eV above and below the Fermi level $\epsilon_F$ to limit the computational complexity. The complete and stable $f^{14}$ filling of the $4f$-level in Lu and the large Hubbard term ($U_f = 5.5$ eV) help place the $4f$ bands deep below $\epsilon_F$, i.e. $\sim 5$ eV below. This leaves the bands near the Fermi level to originate from $5d, 6s$ electrons alone and makes them very similar to the bands obtained in the earlier ‘f-in-core’ calculations. As a result, the spin Hall conductivity of Lu from the ‘f-in-band’ calculations for $U_f = 5.5$ eV, 410 ($h/2e$) $\Omega^{-1}$ cm$^{-1}$, is similar to the result 580 ($h/2e$) $\Omega^{-1}$ cm$^{-1}$ from the ‘f-in-core’ calculation. The difference between these values arises from the difference in the two pseudopotentials.

The influence of the $f$ electrons in Ho is very different than in Lu. The projected density of states and electronic structure for Ho are shown in Figs. 7a and 7b, respectively. While we can identify the signatures of bands originating in $6s$ and $5d$ electrons, similar to those calculated in ‘f-in-core’ calculations, these bands are now pushed about 1.0 eV higher in energy. In addition, a host of flat bands originating from the $4f$ levels are present centered near -0.6 eV, giving rise to a large peak in the density of states (DOS) arising from $4f$-electrons ranging in energy from -0.6 to 0.3 eV, with a significant presence at the Fermi level. The largest contribution to spin Berry curvature, shown in Fig. 7b, comes from the avoided band crossings around the K point. The calculated spin Hall conductivity of Ho, in the ‘f-in-band’ framework is 1260 ($h/2e$) $\Omega^{-1}$ cm$^{-1}$, which is almost
double the value obtained in the ‘f-in-core’ calculation, 
640 (ℏ/2e) Ω−1 cm−1. Such a large difference between 
σs,DFT−in-core and σs,DFT−in-band cannot be explained by a 
difference in the pseudopotentials alone. The central 
result of this calculation is therefore that, within our theo-
retical framework, 4f-electrons do contribute to the spin 
Hall conductivity of Ho in a significant way.

Differences in σs,DFT between the ‘f-in-band’ and ‘f-in-
core’ calculations can arise, in principle, not only from a 
direct contribution of the f-electron bands, but also from 
5d-4f interactions that will modify the band structure. 
Since the 4f electrons are largely localized and the 5d, 6s 
bands structure is altered only to a small extent by the 
presence of 4f-electrons, we conclude that these interac-
tions are small. This suggests that most of the difference 
in σs,DFT arises from the 4f electron contribution, mean-
ning that this 4f contribution is large and positive for Ho. 
This theoretical result agrees with our intuitive discus-
sion of the spin Hall effect from ‘LS’ coupling detailed 
earlier in Fig. 7 that in the late lanthanides the 4f bands 
should make a positive contribution to the total spin Hall 
conductivity.

Our DFT calculations for Dy and Gd also show the sig-
natures of shifted 5d, 6s band-structure features and flat 
4f bands. In the ‘f-in-band’ picture, the f-DOS at the 
Fermi level increases in the order Gd, Dy, and Ho. Ad-
ditionally, the atomic number Z and thus the spin orbit 
coupling constant also increases in the same order. This 
results in an increasing trend of spin Hall conductivity 
from Gd to Dy to Ho. Despite the larger atomic num-
ber Z and spin orbit coupling constant displayed by Lu 
as compared to Ho, the complete absence of 4f DOS at 
εf leads to a significantly smaller spin Hall conductivity 
for Lu compared to Ho. Altogether, the spin Hall con-
ductivity trend predicted by the ‘f-in-band’ calculations 
is σs,DFT,Ho < σs,DFT,Dy < σs,DFT,Gd, which is in con-
trast with the prediction of the ‘f-in-core’ framework for 
a monotonic increase with atomic number Z. The former 
goes against the trend we observe in our experiments.

Given that there is some uncertainty in the literature 
about the most appropriate values to use for Uf, we also 
investigated how changes in Uf affect the calculated spin 
Hall conductivity, shown in Fig. 8. We find that gen-
erally σs,DFT decreases with increasing Uf. In light of 
the above discussion, this reflects that a higher Uf will 
promote greater localization, pushing the 4f levels well 
below εf and lessening their participation in the conduc-
tion bands. This diminishes any enhancement to σs,DFT 
coming from the 4f levels so that in the limit of large 
Uf the ‘f-in-band’ σs,DFT reduces approximately to the 
‘f-in-core’ σs,DFT values (the region of the yellow band in 
Fig. 8). We find that in the cases of Dy and Ho, σs,DFT 
depends quite sensitively upon the choice of Uf. For Dy 
in particular, decreasing Uf from 5 eV to 3 eV doubles 
the calculated spin Hall conductivity. This sensitivity to 
Uf, which depends on the details of electronic structure, 
highlights how potently the 4f bands can enhance σs,DFT. 
For our main results, we take values of Hubbard Uf for
For Lu, the $4f$ occupancy for Ho in the energy range we have examined is 1350 eV between them. The largest value of spin Hall conductivity of $4f$-DOS is less clear for Ho. This is likely due to the proximity of $4f$ to $5d$ bands and the subsequent hybridization between them. The largest value of spin Hall conductivity for Ho in the energy range we have examined is 1350 eV, which is close to the value for the native Fermi level. For Lu, the $4f$ bands lie outside of our scanning region of 0.8 eV around the Fermi level and the variation in spin Hall conductivity in our plotting range arises from the $5d$-electrons alone. The spin Hall conductivity peak for Lu at -0.4 eV and for Dy and Ho near 0.6 eV originates from a $5d$ band feature that is shifted to different energies in the two elements because of different interactions with the $f$ levels. In the case of Gd, there is a small but finite presence of $f$-DOS at the Fermi level. This coincides with the presence of the $5d$ band feature which is responsible for the spin Hall conductivity peak at -0.4 eV in Lu. Together the two contribute to the small spin Hall conductivity value predicted by our calculations. This analysis suggests that engineering the placement of the Fermi level, possibly by alloying, could enhance the spin Hall conductivity significantly if $\epsilon_F$ were tuned to place it in better proximity to either a peak in the $4f$ DOS or to an avoided crossing of the $5d$ and $6s$ bands. The largest value of $\sigma_s^{DFT}$ we have found by tuning $\epsilon_F$ in our four elements is 2070 eV, for Dy at $\epsilon_F = -0.5$ eV.

IV. COMPARISON BETWEEN EXPERIMENTS AND THEORY

We compare in Fig. 10 our experimental results of spin torque ratio and spin torque per unit electric field $\sigma_{SH}^{exp}$ with our theoretical predictions for spin Hall conductivity $\sigma_s^{DFT}$. For best consistency in the experimental values,
we have used the results for Gd, Dy, and Ho with a 1.5 nm Hf spacer. The Lu sample had no Hf spacer. As discussed above and shown in Fig. 5b, if there were no f orbital participation in the spin Hall effect, we should expect to see only a small monotonic increase in the spin Hall conductivity from Gd to Lu owing to the increasing spin orbit coupling with Z. Instead, in the experiment we observe a quite sizable increase in measured spin Hall effect going from Gd to Dy to Ho, and then a drop to a much smaller value in Lu that cannot be captured by the ‘f-in-core’ picture. In contrast, the ‘f-in-band’ calculations are in good qualitative agreement with this tell-tale drop. The ‘f-in-band’ calculations can explain the strong variations in the spin Hall conductivity from Gd to Lu owing to the increasing spin orbit coupling with Z. Instead, in the spin current generated by the spin Hall effect. Another indication of a large amount of spin scattering at the rare-earth interfaces may cause significant attenuation of the spin currents generated by the spin Hall effect. Another mechanism of enhancing SHE in the rare-earths could be to promote the delocalization of the 4f electrons.

Quantitatively, the ‘f-in-band’ calculations predict values of $\sigma^{\text{DFT}}$ that are significantly larger than the experimentally-measured quantities $\sigma^{\text{exp}}$, by a factor of 3-6 depending on the element. That $\sigma^{\text{DFT}}$ is larger than $\sigma^{\text{exp}}$ is not surprising, because the experimentally-measured torques will be reduced by less-than-ideal spin transmission through sample interfaces and by the presence of the Hf spacer in the Gd, Dy, and Ho samples. The large value of the difference suggests that spin scattering at the rare-earth interfaces may cause significant attenuation of the spin currents generated by the spin Hall effect. Another indication of a large amount of spin scattering at interfaces is the fact that the magnetic damping in the RE/Py and RE/Hf/Py samples is generally much larger than can be explained by the standard theory of spin pumping from the Py layer (see the Appendix). Improvement of the rare-earth interfaces might therefore be able to reduce this spin scattering and enable stronger spin-orbit torques.

We conclude, based on both the measurements and the ‘f-in-band’ calculations, that the 4f electrons do indeed contribute to an enhancement of the spin Hall effect in Ho, and perhaps to a lesser extent in Dy. The amount of enhancement depends on the proximity of the 4f-electrons to the Fermi level, so that the spin Hall effect in Gd and Lu remains small. We suggest that the spin Hall conductivity might be further enhanced by tuning the Fermi level closer to either the peak in the 4f DOS or to a 5d avoided band crossing present in the lanthanides. Another mechanism of enhancing SHE in rare-earths could be to promote the delocalization of the 4f electrons.

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**V. APPENDIX**

**A. Analysis of Magnetic Damping**

As noted in the main text, the magnetic damping is increased for heterostructures in which a RE layer is placed in proximity to the ferromagnetic layer, even if there is a thin Hf spacer in between. This increase reflects the...
Table IV. A summary of the damping enhancement $\Delta \alpha$, the calculated effective spin mixing conductance for each RE (10 nm)/Hf (1.5 nm)/Py (5 nm)/AlO$_x$ heterostructure as described in the Appendix, and an estimated effective spin conductivity $2\rho_{RE}/\lambda_{RE}$ for each rare-earth. $\alpha = 0.0077$ as measured for a sapphire/Hf (1.5 nm)/Py (5 nm)/AlO$_x$ control sample. $\lambda_{RE}$ is assumed to be $\approx 1$ nm.

| Element | $\Delta \alpha$ (10$^{-3}$) | $g_{eff}^{\gamma}(\text{nm}^{-2})$ | $2\rho_{RE}/\lambda_{RE}$ (nm$^{-2}$) |
|---------|----------------------------|-------------------------------|---------------------------------|
| Gd      | 1.2                        | 2.9                           | 9.6                             |
| Dy      | 9.3                        | 18.6                          | 10.6                            |
| Ho      | 2.4                        | 6.2                           | 4.4                             |

contributions to magnetic dissipation from spin pumping into the RE layer and any spin relaxation that may occur at the Hf/RE interface. For Gd, Dy, and Ho, we compared the value of the Gilbert damping ($\alpha_C$) determined by ST-FMR for RE (10 nm)/Hf (1.5 nm)/Py (5 nm)/AlO$_x$ multilayers with the value ($\alpha_C - \Delta \alpha$) for control samples without the RE layer, but still containing the Hf spacer layer. Assuming diffusive spin flow, we can define an effective spin mixing conductance for spin transport from the Py/Hf to the RE as

$$g_{eff}^{\gamma} = \frac{M_s^{eff} E_{ss}^{eff}}{\gamma_h^2} \Delta \alpha.$$  

(9)

The results are listed in Table IV. The values determined by this technique should be less than the true effective mixing conductance of the interface between Py and Hf because of the spin current attenuation within the Hf. In spite of this, the values for Dy and Ho (in particular) are surprisingly large. In the absence of interfacial spin relaxation mechanisms, the maximum effective spin mixing conductance should be no more than $2\rho_{RE}/\lambda_{RE}$, where $\rho_{RE}$ is the electrical resistivity of the rare-earth material and $\lambda_{RE}$ is its spin diffusion length, because this quantity determines the ability of the RE layer to absorb a spin current from the precessing ferromagnet. If we assume $\lambda_{RE} \approx 1$ nm, we find for Dy and Ho that $g_{eff}^{\gamma}$ is much larger than $2\rho_{RE}/\lambda_{RE}$ (see Table IV). We therefore conclude that the large values of damping in the Dy and Ho samples are not dominated by conventional spin pumping from the ferromagnet into the RE layer, but rather are more likely the result of interfacial intermixing, and the ability of Dy and Ho impurities to greatly increase spin relaxation.$^{36,37}$

We also measured the effective Gilbert damping coefficient as a function of Hf spacer thickness in a set of Ho (10 nm)/Hf ($t_{Hf}$)/Py (5 nm)/AlO$_X$ samples for $t_{Hf} = 0.5, 1, 2,$ and 4 nm (Fig. 11). The results fit well to a simple exponential function plus an offset, with an effective spin diffusion length in the Hf of 0.81 nm. The offset of 0.0071 gives the limit in which the Hf is so thick that the Ho layer is not relevant to the damping enhancement. This value agrees with the damping for the control sample of sapphire/Hf (1.5 nm)/Py (5 nm)/AlO$_X$ of $\alpha_0 = 0.0077$ that was used to determine the values of $\Delta \alpha$ in Table IV.

Figure 11. (Color online) Damping of Ho (10 nm)/Hf ($t_{Hf}$/Py (5 nm)/AlO$_X$ stacks for $t_{Hf} = 0.5, 1, 1.5, 2,$ and 4 nm (red circles). Fitting to $\alpha = \text{offset} + Ae^{-t/\lambda_{sd}}$ (blue line) yields offset $= 0.0071$, $A = 0.0087$, and $\lambda_{sd} = 0.81$ nm.

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