Linear relation between Heisenberg exchange and interfacial Dzyaloshinskii-Moriya interaction in metal films

Hans T. Nembach, Justin M. Shaw, Mathias Weiler*, Emilie Jué and Thomas J. Silva

Electromagnetics Division, National Institute of Standards and Technology,
Boulder, Colorado 80305, USA

*present address: Walther-Meißner-Institut, 85748 Garching, Germany
Determination of effective magnetic thickness for Ni$_{80}$Fe$_{20}$ films

The physical thickness of the sputtered (111)-textured Ni$_{80}$Fe$_{20}$ layers was calibrated by use of x-ray reflectometry. All samples were cut by a dicing saw into equal sized chips. The 0 K saturation magnetic moment $m_s$, obtained from the fit of $m_s$ vs. $T$ SQUID magnetometer data to the Bloch $T^{3/2}$ law, is plotted against the physical thickness $t_n$ to determine the effective magnetic thickness of the Ni$_{80}$Fe$_{20}$ layer, as shown in Fig. S1. The zero-moment intercept of a linear fit of the data indicates that there is a magnetic “dead” layer of thickness $t_D=0.77$ nm $\pm 0.05$ nm. The observation of such magnetic dead layers at the ferromagnet interface is not uncommon for sputtered metallic films$^1$. Recently reported data from spin-pumping measurements provide evidence that the Ni$_{80}$Fe$_{20}$/Pt interface does not exhibit a dead layer$^2$ suggestive that the dead layer in the present case is at the Ni$_{80}$Fe$_{20}$/SiN interface, perhaps due to the formation of a non-magnetic silicide such as Fe$_x$Si and/or Ni$_x$Si during SiN deposition$^3$. In the main text, we use the layer thickness $t=n-t_D$.

**Fig. S1:** The magnetic moment is plotted versus the nominal thickness $t_n$ of the Ni$_{80}$Fe$_{20}$ layer. The non-zero x-axis intercept indicates the existence of a dead layer with a thickness $t_D$. The error is smaller than the symbol size.
Determination of effective magnetic thickness for Ni80Fe20 films

The physical thickness of the sputtered (111)-textured Ni80Fe20 layers was calibrated by use of x-ray reflectometry. All samples were cut by a dicing saw into equal sized chips. The 0 K saturation magnetic moment $m_s$, obtained from the fit of $m_s$ vs. $T$ with SQUID magnetometer data to the Bloch $T^{3/2}$ law, is plotted against the physical thickness $t_n$ to determine the effective magnetic thickness of the Ni80Fe20 layer, as shown in Fig. S1. The zero-moment intercept of a linear fit of the data indicates that there is a magnetic “dead” layer of thickness $t_D = 0.77 \text{ nm} \pm 0.05 \text{ nm}$. The observation of such magnetic dead layers at the ferromagnet interface is not uncommon for sputtered metallic films. Recently reported data from spin-pumping measurements provide evidence that the Ni80Fe20/Pt interface does not exhibit a dead layer suggestive that the dead layer in the present case is at the Ni80Fe20/SiN interface, perhaps due to the formation of a non-magnetic silicide such as Fe$_n$Si and/or Ni$_n$Si during SiN deposition.

In the main text, we use the layer thickness $t = t_N - t_D$.

**Fig. S1:** The magnetic moment is plotted versus the nominal thickness $t_n$ of the Ni80Fe20 layer. The non-zero x-axis intercept indicates the existence of a dead layer with a thickness $t_D$. The error is smaller than the symbol size.

**Fig. S2:** Temperature-dependence of saturation moment $m_s$ for the $t_{N=10} = 2.7$ nm sample. The red line is a fit of the data to the Bloch $T^{3/2}$ Law.

**Determination of the symmetric (Heisenberg) exchange constant**

The Heisenberg Hamiltonian is

$$H = - \sum_{i \neq j} J_{ij} \left( \vec{S}_i \cdot \vec{S}_j \right),$$

(S1)

where $J_{ij}$ is the exchange energy between the two atomic sites $i$ and $j$, with spin $\vec{S}_i$ and $\vec{S}_j$, respectively. The symmetric-exchange contribution to the spin-wave energy $D_{spin}$ for a fcc lattice with lattice constant $a$ is given by

$$E_{spin} = \frac{1}{3} S \sum_{i \neq j} J_{ij} \left( 1 - \cos \left( k \cdot \vec{r}_{ij} \right) \right)$$

$$= \left( 2 J_{nn} S a^2 \right) k^2$$

$$= D_{spin} k^2$$

(S2)

where $\vec{r}_{ij}$ is the position vector connecting sites $i$ and $j$, $J_{nn}$ is the nearest-neighbor exchange integral, and $S$ is the total number of spins at each site.
In order to determine $D_{\text{spin}}$, we measured the temperature dependence of the magnetic moment $m_s(T, t)$ for Ni$_{80}$Fe$_{20}$ films of thickness $t$ for all samples by SQUID magnetometry with an applied field of $\mu_0H=10$ mT. The data were fit with the Bloch $T^{3/2}$ Law:

$$m_s(T, t) = m_s(T = 0 K, t) \cdot \left(1 - \frac{g\mu_B\eta}{M_s(T = 0 K, t)} \cdot \left(\frac{k_B T}{D_{\text{spin}}(T = 0 K, t)}\right)^{3/2}\right)$$  \hspace{1cm} (S3)

where $k_B$ is Boltzmann’s constant, $D_{\text{spin}}(T = 0 K, t)$ is the zero kelvin spin-wave stiffness with units of J m$^2$, and $\eta$ is a dimensionless constant that can depend on the sample dimensions. For a bulk sample, $\eta = 0.0587$. However, for the thin films of reduced dimensionality used here, $\eta$ must be explicitly calculated as detailed below. The prefactor $\frac{g\mu_B}{M_s(T = 0 K, t)}$ is equal to the volume per spin. A plot of $m_s$ vs. $T$ for the $t = 2.7$ nm sample, together with the fit to Eq. (S3), is shown in Fig. S2. We extrapolated the fits to $T=0$ K to obtain the magnetization for all samples at $T = 0 K$, which was approximately equal to the bulk magnetization $\mu_0 M_s \sim 1 T$ for all samples. These data imply that the spin-densities for all samples are the same and do not change throughout the thickness.

In order to determine the spin-wave stiffness $D_{\text{spin}}(T = 300 K, t)$ at room temperature from $D_{\text{spin}}(T = 0 K, t)$, renormalization of the exchange needs to be taken into account. To lowest order the temperature dependence can be approximated with the temperature dependence of the saturation magnetization raised to the power of $\gamma$

$$D(T, t) \approx D_{\text{spin}}(T = 0 K, t) \left[\frac{M_s(T = 300 K, t)}{M_s(T = 0 K, t)}\right]^\gamma.$$  \hspace{1cm} (S4)

According to mean field theory $^5$ $\gamma = 1$, for an itinerant ferromagnet with magnon-electron interaction $^6$ $\gamma = 4/3$ and $\gamma = 0.7$ was found experimentally for magnetite $^7$. In our manuscript we use $\gamma = 1$ from mean field theory and determine the error bars from the variation of $D_{\text{spin}}(T = 0 K, t)$ when using $\gamma = 4/3$ and $\gamma = 0.7$, respectively. (The particular choice of $\gamma$ does not alter the result, as indicated by the small size of the error bars for $A$ in the manuscript). We measured in-plane hysteresis curves by SQUID magnetometry at room temperature to determine the saturation magnetization $M_s(300 K)$ of our samples. We find that $M_s(300 K)$ decreases with decreasing Ni$_{80}$Fe$_{20}$ layer thickness (see Fig. 4a in the main text). We then calculate the exchange constant $A$ at room temperature (plotted in Fig. 4c in the main text) for each sample via

$$A(T = 300 K, t) = D_{\text{spin}}(T = 0 K, t) \left[\frac{M_s(T=300 K,t)}{M_s(T=0 K,t)}\right]^{\gamma} \cdot \left[\frac{M_s(T=300 K,t)}{2g\mu_B}\right].$$ \hspace{1cm} (S5)

**Calculation of the magnon density**

For the determination of the spin-wave stiffness constant from the Bloch $T^{3/2}$ law, the prefactor $\eta$ in thermal equilibrium is required. We used exact numerical summation of the Planck distribution for a finite volume with unpinned boundary conditions to calculate $\eta$. The magnon density at a temperature $T$ in thermal equilibrium is
\[ n_m(T) = \frac{1}{8V} \sum_{i,j,k} \frac{1}{\exp \left( \frac{D_{\text{spin}} k_{i,j,k}^2 + hf_{\text{FMR}}}{k_B T} \right) - 1}, \quad (S6) \]

where

\[ k_{i,j,k}^2 = \left( \frac{i\pi}{L_x} \right)^2 + \left( \frac{j\pi}{L_y} \right)^2 + \left( \frac{k\pi}{L_z} \right)^2. \quad (S7) \]

\( L_{x,y,z} \) are the Cartesian dimensions of the finite-sized sample, \( V = L_x L_y L_z \) is the volume of the magnetic sample, and \( f_{\text{FMR}} \) is the FMR frequency in the field at which the temperature-dependent magnetization is measured. (As was the case for the original derivation of the \( T^{1/2} \) law, only exchange-mode magnons are considered. Refinement of the calculation to include dipole field effects would greatly increase the complexity of the calculation, though it is not expected to have a larger impact on the final results, as is further explained below.) We transform Eq. (S6) into cylindrical coordinates and evaluate the sum numerically.

To determine \( \eta \), we then divide the magnon density by the argument of the Bloch law, i.e.

\[ \eta = \frac{n_m(T)}{k_B T^{3/2}}. \quad (S8) \]

In Fig. S3, we plot of \( \eta \) as a function of \( t \) for the case of Permalloy with a lattice constant \( a = 0.354 \text{ nm} \). We see that \( \eta \) is approximately equal to the classic bulk value of 0.0587 at the largest thickness calculated, but it deviates significantly from the bulk value for \( t < 10 \text{ nm} \).

The reason for the deviation can be easily understood in terms of how the Planck distribution is affected for spin waves with wavelengths comparable to or shorter than the film thickness. Spin waves with wavelengths less than 4 nm have an energy greater than \( k_B T \). Referring to the Planck distribution, such modes are populated according to a Boltzmann-like factor of \( \exp \left( -D k^2 / k_B T \right) \), such that they are only marginally occupied. When the sample size is reduced, the reduction in the number of thermally excited magnons is balanced by the reduced volume of the sample, such that \( n_m(T) \) is mostly unaffected. However, the lowest order excitation of the solid, i.e. the FMR mode, is only weakly affected by the reduced sample volume, such that its occupation remains approximately constant. In the spirit of the original derivation of the Bloch law, we are ignoring dipole field effects that would generally cause some modification of the lowest order excitation of the solid with reduced size. In particular, for the specific case considered here, the relative contribution of the exchange energy to the mode frequency for \( 4 \text{ nm} < \lambda < 20 \text{ nm} \) (8 meV > \( E > 0.3 \text{ meV} \)) always exceeds the contribution due to dipole fields (0.1 meV > \( E > 0.06 \text{ meV} \)) by a significant margin. Thus, reducing the sample dimension along a particular quantization axis freezes out all spin-wave modes in that direction, but the FMR mode continues to be highly occupied. This results in an enhancement of \( n_m(T) \), and therefore \( \eta \), with decrease sample thickness.
Fig. S3: Prefactor $\eta$ for the Bloch $T^{3/2}$ law. For thicknesses $t > 10$ nm, $\eta$ approaches the bulk value 0.0587.

**Ferromagnetic Resonance Measurements**

We determined the net perpendicular anisotropy and in-plane spectroscopic $g$-factor $g^\perp$, see Fig. S4 and S5, respectively, by use of broadband vector network analyzer ferromagnetic resonance (VNA-FMR) measurements for the cases of a saturating magnetic field both perpendicular-to and parallel-to the sample plane. Experimental details can be found in Nembach et al.\(^9\). The data for the resonance field $H_\perp$ vs. excitation frequency $f$ in the out-of-plane geometry are fitted to the Kittel equation for the perpendicular geometry,

$$f = \frac{\mu_0 \mu_B g^\perp}{h} \left( H_\perp - M_{\text{eff}} \right),$$  

(S12)

where $\mu_0$ is the permeability of free space, $\mu_B$ is the Bohr magneton, $h$ is Planck’s constant, $g^\perp$ is the spectroscopic splitting factor for the perpendicular geometry, and $M_{\text{eff}} = M_s - H_k$ is the effective magnetization, where $H_k$ is the interfacial perpendicular anisotropy field. For $H_k > 0$, the interfacial anisotropy easy axis is parallel to the sample normal. The fitting parameters are $g^\perp$ and $M_{\text{eff}}$. Together with the room temperature values for $M_s(T = 300 \text{ K}, t_n)$ obtained from SQUID magnetometry, we determine the thickness-dependence of $H_k$, shown in Fig. S4. The linear dependence of $H_k$ on $1/t$ is a clear signature of interfacial anisotropy.

The in-plane VNA-FMR resonance fields $H_\parallel$ are also fitted with the Kittel equation for this geometry.
Fig. S3: Prefactor \( \eta \) for the Bloch T3/2 law. For thicknesses \( t > 10 \) nm, \( \eta \) approaches the bulk value 0.0587.

Ferromagnetic Resonance Measurements

We determined the net perpendicular anisotropy and in-plane spectroscopic splitting factor \( g_\perp \), see Fig. S4 and S5, respectively, by use of broadband vector network analyzer ferromagnetic resonance (VNA-FMR) measurements for the cases of a saturating magnetic field both perpendicular-to and parallel-to the sample plane. Experimental details can be found in Nembach et al9. The data for the resonance field \( H_\perp \) vs. excitation frequency \( f \) in the out-of-plane geometry are fitted to the Kittel equation for the perpendicular geometry,

\[
f = \frac{\mu_0 \mu_B g_\parallel}{h} \sqrt{H_\parallel \cdot (H_\parallel + M_{eff})}.
\]  

(S13)

Since we have previously shown that the perpendicular and in-plane values of \( M_{eff} \) are identical10, \( g_\parallel \) shown in Fig. S5 is more precisely determined from Eq. (S13) by fixing the value of \( M_{eff} \) to that obtained in the perpendicular geometry.

Fig. S4: Anisotropy field \( H_K \) versus the reciprocal thickness \( t \). The error originated from the fit of the data to the Kittel equation and is smaller than the symbol size for all but the thinnest sample.
Fig. S5: In-plane spectroscopic splitting factor $g^\parallel$ versus the reciprocal thickness $t$. The error originated from the fit of the data to the Kittel equation and is smaller than symbol size for all but the thinnest sample.
Wavevector dependent frequency shift

We measured this wavevector dependent frequency shift with BLS for the 2.0 nm thick sample with an applied field of $\mu_0 H = 244$ mT, see Fig. S6. The measured frequency shift is proportional to the spin-wave frequency as it is expected from Eq. (2) in the main text.

**Fig. S6**: Wavevector dependence of the frequency shift for the 2.0 nm thick sample. The applied field for the measurements is $\mu_0 H = 244$ mT. The error bars originate from the fit of the spectra.

1. Lefakis, H. et al. Structure and magnetism of Ta/Co/Ta sandwiches. *J. Magn. Magn. Mater.* **154**, 17–23 (1996).
2. Boone, C., Shaw, J. M., Nembach, H. T. & Silva, T. J. *ArXiv14085921v1* 2014
3. Schlesinger, M. E. Thermodynamics of solid transition-metal silicides. *Chem. Rev.* **90**, 607–628 (1990).
4. Vaz, C. a. F., Bland, J. a. C. & Lauhoff, G. Magnetism in ultrathin film structures. *Rep. Prog. Phys.* **71**, 056501 (2008).

5. Atxitia, U. *et al.* Micromagnetic modeling of laser-induced magnetization dynamics using the Landau-Lifshitz-Bloch equation. *Appl. Phys. Lett.* **91**, 232507 (2007).

6. Lovesey, S. W. *Theory of neutron scattering from condensed matter*. (Clarendon Press, 1984).

7. Heider, F. & Williams, W. Note on temperature dependence of exchange constant in magnetite. *Geophys. Res. Lett.* **15**, 184–187 (1988).

8. Nembach, H. T. *et al.* Perpendicular ferromagnetic resonance measurements of damping and Landég-factor in sputtered (Co2Mn)1−xGex thin films. *Phys. Rev. B* **84**, 054424 (2011).

9. Shaw, J. M., Nembach, H. T., Silva, T. J. & Boone, C. T. Precise determination of the spectroscopic g-factor by use of broadband ferromagnetic resonance spectroscopy. *J. Appl. Phys.* **114**, 243906 (2013).