

Negative temperature coefficient of Gilbert damping in magnetic bilayers

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(Received 7 July 2025; revised 16 January 2026; accepted 30 April 2026; published 22 June 2026)

The Gilbert damping of magnetic materials is an important magnetic parameter that determines the switching speed and energy dissipation of spintronic devices. In simple metals, the intrinsic Gilbert damping increases with temperature and diverges near the Curie temperature as a result of spin fluctuations. Here we present atomistic simulations and experimental measurements showing surprising and opposite behavior in Py/Nd bilayers, where the Gilbert damping *decreases* with increasing temperature. The effect arises because of the enhanced damping at the interface as a result of spin pumping, where elevated temperatures cause a dynamic separation of the interfacial and bulk magnetization during relaxation. Furthermore, the temperature dependence of the damping can be controlled by varying the thickness of the Nd capping layer. Our findings present a spintronic effect that can be used to modify the dynamic properties of nanoscale materials and devices for enhanced energy efficiency or with improved switching dynamics.

DOI: [10.1103/physrevb.113.L220409](https://doi.org/10.1103/physrevb.113.L220409)

The Gilbert damping factor is an important magnetic parameter that determines the speed of magnetization dynamics in spintronics devices, such as magnetic sensors and magnetic random access memory. The intrinsic Gilbert damping arises at the electronic structure level due to magnon/electron [1–3] or magnon/phonon interactions in insulators. However, in nanoscale devices extrinsic contributions including magnon/magnon interactions and scattering at surfaces become important. Theoretically, several models have attempted to explain the origin of damping, such as the *s-d* exchange scattering model, the Fermi surface breathing model, the two-magnon scattering model, and the spin pumping model [4–7]. For interfacial Gilbert damping that arises between ferromagnetic/nonmagnetic bilayers, the spin pumping effect is widely considered to be responsible for the dependence of the damping on the thickness of the nonmagnetic layer [8]. However, most studies focused on the variation of Gilbert damping values at room temperature or lower, with few research groups investigating changes in Gilbert damping over a wider temperature range including close to the Curie temperature. In thin films of permalloy the temperature dependence

of the damping was found to be nonmonotonic for small film thicknesses, with a peak around $T = 50$ K [4] but weakly temperature dependent with a positive temperature coefficient for thicker films. The nonmonotonic behavior was attributed to a potential spin-reorientation transition at low temperature due to enhanced interfacial anisotropy [4]. The microstructure is also known to have an important effect on Gilbert damping and also its temperature dependence with recent studies on FePt [9] finding a decrease in the ferromagnetic resonance linewidth at higher temperatures, which is often interpreted as a decrease in Gilbert damping. However, recent theoretical calculations attributed the effect to inhomogeneous line broadening at low temperatures (increasing the effective damping) that reduces at high temperatures [10].

As well as its fundamental importance, the Gilbert damping parameter is especially important for devices including magnetic random access memory (MRAM) [11,12], heat assisted magnetic recording (HAMR), and magnetic hyperthermia. In MRAM devices low damping can aid switching speeds and is a desirable property for fast switching. However, Joule heating can cause an increase in device temperature during switching that usually increases the effective damping constant. Similarly for HAMR devices, dynamic heating can cause changes in damping that are important for the switching probability and thermal stability of magnetic grains [10]. The Gilbert damping and its temperature dependence is therefore an important engineering parameter for devices.

In this letter, we report the temperature dependence of the Gilbert damping of a Py/Nd bilayer using an atomistic spin dynamics model including spin pumping and complementary time-resolved magneto-optical Kerr effect (TR-MOKE)

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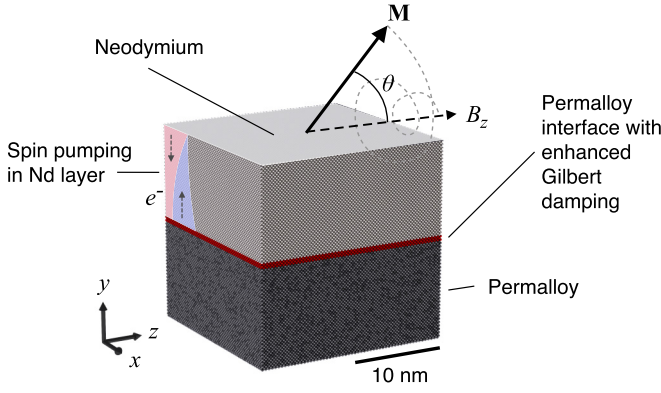


FIG. 1. Schematic view of the simulated Py (6 nm)/Nd (t nm) bilayer. The structural dimensions are $20 \times 20 \times (6 + t)$ nm³, where the dark-gray underlayer and the red inserted layer represent the bulk and interface Py layer, respectively. The light-gray top layer represents the Nd layer.

measurements of the pump fluence dependence of Gilbert damping. To study the temperature effect on Gilbert damping we first consider atomistic spin dynamics simulations of the magnetic relaxation process at a range of temperatures $T = 20$ – 800 K. Based on a spin accumulation model [13], we introduce bulk and interface magnetic damping to investigate the thickness dependence of the damping in Py (6 nm)/Nd (t nm) bilayer films [4,8,13,14], as shown in Fig. 1. The simulations utilize a classical spin model with a three-dimensional Heisenberg spin Hamiltonian [15] describing the energy contribution of the magnetic systems given by

$$\mathcal{H} = - \sum_{i < j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - k_u \sum_i (\mathbf{S}_i \cdot \mathbf{e})^2 - |\mu_s| \sum_i \mathbf{S}_i \cdot \mathbf{B}_{\text{app}}, \quad (1)$$

where J_{ij} is the nearest-neighbor exchange integral between spin sites i and j . Nearest-neighbor exchange is used following Hinzke *et al.* [16] who showed using first-principles calculations that direct exchange was dominant. $\mathbf{S}_i$ is the local normalized spin moment, $\mathbf{S}_j$ is the normalized spin moment of the neighboring atom at site j , k_u is the uniaxial anisotropy constant, and $\mathbf{e}$ is the unit vector of the easy axis. Each magnetic moment is normalized so that $\mathbf{S} = \frac{\boldsymbol{\mu}}{|\mu_s|}$, where $|\mu_s|$ is the magnitude of the spin moment. $\mathbf{B}_{\text{app}}$ is the applied external field. The magnetic parameters of Py and Nd are listed in Supplemental Material (SM) Table I [17] with parameters derived from previous publications [18–22].

We simulate magnetic dynamic behavior using the stochastic Landau-Lifshitz-Gilbert equation applied at the atomistic level [15,23] and given by

$$\frac{\partial \mathbf{S}_i}{\partial t} = - \frac{\gamma}{1 + \alpha^2} [\mathbf{S}_i \times \mathbf{B}_{\text{eff}}^i + \alpha \mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{B}_{\text{eff}}^i)], \quad (2)$$

where $\mathbf{S}$ is the normalized spin moment, γ is the absolute value of the gyromagnetic ratio, and α is the intrinsic damping constant, which represents the coupling of the spin to the heat bath. $\mathbf{B}_{\text{eff}}^i = \mathbf{B}_{\text{th}}^i - \frac{1}{|\mu_s|} \frac{\partial \mathcal{H}}{\partial \mathbf{S}_i}$ is the effective field that acts on the local spin moment determined from the Heisenberg spin Hamiltonian. The effective field is enhanced by a thermal

fluctuating field $\mathbf{B}_{\text{th}}^i(t) = \boldsymbol{\Gamma}(t) \sqrt{\frac{2\alpha k_B T}{\gamma \mu_s \Delta t}}$ within a Langevin Dynamics formalism [24] where $\boldsymbol{\Gamma}(t)$ is the Gaussian distribution and T is the temperature. The intrinsic Gilbert damping α is treated as a zero-temperature constant for the simulations, and is different for the bulk and interface layer. For increasing Nd layer thickness we model the effect of the spin pumping by increasing the intrinsic damping at the Nd/Py interface $\alpha_{\text{int}} = \alpha_{\text{max}}(1 - e^{-2d_{\text{NM}}/\lambda_{\text{sdl}}})$, with d_{NM} the thickness of the Nd layer and λ_{sdl} the spin diffusion length [13,25]. More details of the spin pumping implementation and spin pumping-induced interface damping values in the simulation are presented in the supplemental Material [17].

To calculate the effective Gilbert damping α_{eff} , we applied a $B_z = 10$ T external magnetic field along the z direction [18]. The magnetization is initially oriented in the yz plane at an angle of 30° to the applied field from where it undergoes damped precession into the field direction, as shown in Fig. 1. Different angles between the initial magnetization direction and the applied field are simulated for Py (6 nm)/Nd (32 nm) and are shown in the Supplemental Material [17]. Figure S1 shows similar results independent of the initial magnetization angle. The time evolution of the x component of magnetization for the Py (6 nm)/Nd (32 nm) bilayer in the atomistic model at different temperatures is shown in Fig. 2. For the Py/Nd bilayer, as the net magnetization of the system evolves back to the equilibrium state, the precession frequency and the relaxation time are extracted using the equation

$$M(t) = A \exp\left(-\frac{t}{\tau}\right) \sin(2\pi f t), \quad (3)$$

where A is the amplitude of the magnetization, f is the precession frequency, and τ is the relaxation time. From Eq. (3), the relaxation time of the magnetization at different temperatures is extracted and the effective Gilbert damping (α_{eff}) can be determined using equation $\alpha_{\text{eff}} = 1/2\pi f \tau$. To obtain statistically accurate results, we performed simulations with 28 different numerical random seeds for each parameter set, and then averaged the values and calculated the variance to obtain the mean-effective Gilbert damping, as shown in Supplemental Material Fig. S2 [17]. The mean-effective Gilbert damping for Py/Nd bilayers with different Nd thickness was calculated as a function of temperature and is shown in Fig. 3. In agreement with previous work [4] we can see that for a pure Py thin film the Gilbert damping value increases with temperature as expected from increased thermal fluctuations. In contrast, the Py/Nd (32 nm) bilayers show a remarkable *decrease* in the effective damping with increasing temperature, with an effective Gilbert damping value of $\alpha_{\text{eff}} = 0.0822$ at $T = 20$ K, which decreases to $\alpha_{\text{eff}} = 0.0636$ at $T = 800$ K. This is an unexpected result as higher temperatures induce larger thermal fluctuations in the magnetization that leads to a slightly slower relaxation towards the equilibrium direction. Here, the damping has an additional surface contribution from the spin pumping, which clearly plays a role on the effective damping, especially at higher temperatures.

In order to verify the predictions of the atomistic simulations, we performed experimental measurements of the temperature dependence of the Gilbert damping for the magnetic bilayer using TR-MOKE with variable pump fluence

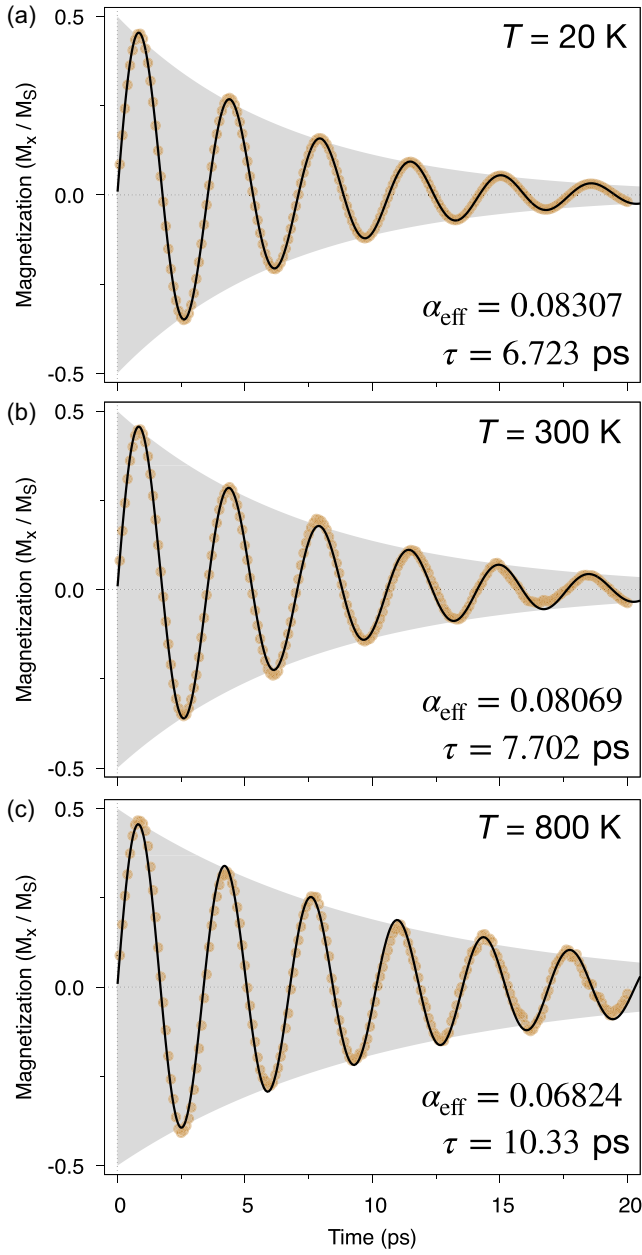


FIG. 2. Simulated time evolution of the magnetization trace (points) for the Py (6 nm)/Nd (32 nm) bilayer at different temperatures (a) $T = 0$ K, (b) $T = 300$ K, and (c) $T = 800$ K showing a systematic *decrease* in the effective Gilbert damping α_{eff} with increasing temperature as seen from the amplitude of the envelope function depicted by the shaded region. The lines are fitted to the data using Eq. (3).

to change the effective temperature during relaxation. The structure and magnetic properties of the bilayer films have been presented in previous work [26]. To study the effect of temperature on the effective Gilbert damping, ultrafast laser pulses are used to excite magnetization dynamics and elevate the temperature of the system. The ultrafast demagnetization behavior modifies the anisotropy field and magnetization within a few picoseconds; then the anisotropy field recovers and the magnetization returns to the initial equilibrium position via a precession process that will last for hundreds of

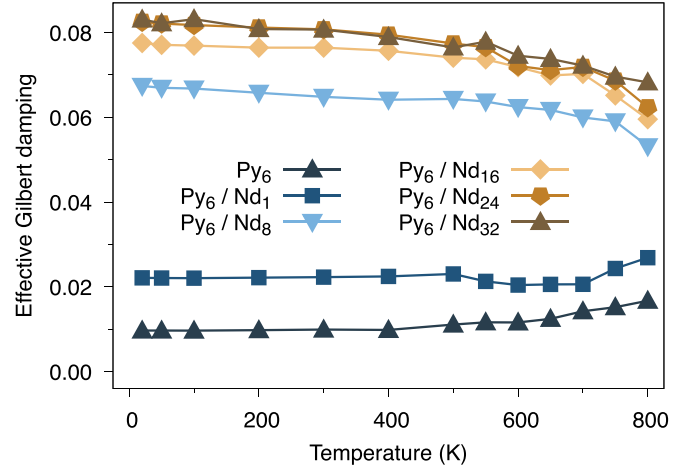


FIG. 3. Simulated mean-effective Gilbert damping as a function of temperature for Py (6 nm)/Nd (0, 1, 8, 16, 24, 32 nm) bilayers. The data show a consistent decrease of the effective damping with temperature for Py (6 nm)/Nd (8, 16, 24, 32 nm) bilayers, and the expected increase for the case for pure Py.

picoseconds. The schematic structure of the sample and the geometry of the TR-MOKE measurement are shown in detail in Fig. S3(a). To quantitatively analyze the data, we use a phenomenological equation to fit the Kerr rotation trace:

$$\Delta\theta = A \exp\left(\frac{-t}{\tau}\right) \cos(2\pi ft + \phi) + B(t), \quad (4)$$

where A , τ , f , and ϕ are the magnetization precession amplitude, the relaxation time, the precession frequency, and the initial phase, respectively. $B(t)$ is the background term associated with the demagnetization recovery process. Fitting the transient Kerr traces under different pump fluences with Eq. (4), we obtain the precession frequency f and the relaxation time τ . In the experiment, we observed only one mode of precession frequency, which excludes the contribution of multimode spin waves to the effective Gilbert damping. Additionally, it is known that the effective Gilbert damping is stable at a high magnetic field [26]. Therefore, we can approximately take into account the effective Gilbert damping under a high magnetic field from the intrinsic damping of the material and the enhancement damping from the spin pumping effect. The effective electron temperature T_e immediately after the pulse can be deduced from the absorbed laser energy per unit volume E_a using [27–29]

$$E_a = \gamma(T_e^2 - T_0^2)/2, \quad (5)$$

where T_0 is the initial electronic temperature (room temperature). This arises from the fact that the electronic specific heat is linearly dependent on temperature such that $C_e = \gamma T_e$, with $\gamma_{\text{Fe}} = 0.7 \text{ mJ/cm}^3 \text{ K}^2$, $\gamma_{\text{Ni}} = 6 \text{ mJ/cm}^3 \text{ K}^2$. Meanwhile, E_a can also be estimated according to the optical parameters:

$$E_a = \left[1 - \exp\left(-\frac{d}{\lambda}\right)\right] \frac{I_F(1-R)}{d}, \quad (6)$$

where I_F is the incident laser fluence, R is the reflectivity of the material at the wavelength, λ is the absorption length, and d

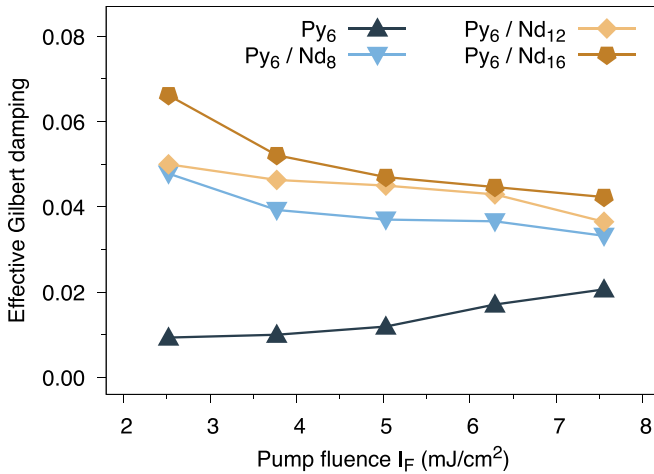


FIG. 4. Experimentally measured effective Gilbert damping as a function of the pump fluence of the laser I_F for Nd(t = 0, 8, 12, 16 nm)/Py (6 nm) bilayers.

is the film thickness. Combining Eqs. (5) and (6), we obtained the electronic temperature of Ni in the range between 540 and 850 K, and the electronic temperature of Fe between 1350 and 2350 K, with a weighted average electron temperature of NiFe between 580 and 920 K. The effective Gilbert damping and the dependence of the electron temperature with pump fluence are shown in Figs. S3(b)–S3(d) [17]. Using this, we can therefore characterize the temperature dependence of the effective Gilbert damping.

The effective Gilbert damping values for the Nd (t nm)/Py (6 nm) bilayer under different pump fluences, are shown in Fig. 4. For a single-layer Py thin film, the effective Gilbert damping value continuously increases with increasing pump fluence due to the increasing presence of magnons with increasing temperature [30,31]. However, for the Nd (8,12,16 nm)/Py bilayer, the effective Gilbert damping values decrease continuously with increasing pump fluence, as seen in the earlier atomistic simulations. Further details of the Py/Nd measurements are presented in Supplemental Material Fig. S4 [17].

To investigate the general effect of spin pumping on temperature-dependent damping, we also performed comparative measurements of the effective Gilbert damping values for Pt (2,4 nm)/Py (6 nm) and $\text{Pt}_{0.68}\text{Nd}_{0.32}$ /Py bilayers under different pump fluence, shown in Fig. S6. The TR-MOKE measurements show an increasing effective Gilbert damping with temperature as with the pure Py thin film. In Pt/Py ($\text{Pt}_{0.68}\text{Nd}_{0.32}$ /Py) bilayers the interface spin accumulation is very small [8,25,32] due to a short spin diffusion length of 0.609 ± 0.092 nm (3.509 ± 0.732 nm), as measured in Fig. S5 [17]. Thus the spin pumping effects in Pt and $\text{Pt}_{0.68}\text{Nd}_{0.32}$ are already saturated for this thickness. This limits the enhancement of interfacial Gilbert damping due to spin pumping, meaning that the bulk and interface damping are close in value. This suggests that the origin of the effect is due to a small separation of the interfacial and bulk magnetization dynamics, as shown in Fig. S7 [17], where the surface experiences much larger thermal fluctuations due to the loss of exchange bonds through reduced coordination at the surface [33].

To evaluate this possibility, we simulate an idealized system where the spin pumping–enhanced damping is distributed much deeper into the Py film. In this computational experiment, we assume a bulk layer and interface layer each 3 nm thick with different Gilbert damping constants. The magnetic parameters and the simulation results are shown in detail in Fig S8 [17]. The mean-effective Gilbert damping value is 0.01 at 300 K, which is consistent with previous results [8,13,26]. In this simulation, the mean-effective Gilbert damping value increases with increasing temperature, as for the pure Py film. Thus, the origin of the negative temperature-dependent coefficient of Gilbert damping can be ascribed to the strong localization of the damping enhancement to the interface Py region. Here the spin fluctuations are largest due to the reduction in coordination [33]. This leads to a rapid loss of enhancement resulting in a decrease in the damping. This behavior originates from the large enhancement of the spin pumping–induced interface damping, combined with enhanced surface spin fluctuations leading to increased surface thermal fluctuations of the magnetization, as shown in Figs. S9 and S10 [17]. This partially decouples the surface and bulk magnetization, thus reducing the effective damping enhancement due to the spin pumping. Finally, considering the case of small spin pumping–enhanced interface damping in the case of monolayer Nd and Pt layers, we see a small increase in effective damping as a function of temperature in Fig. 3 for the case of 6 Py/1 Nd. In this case the bulk and interfacial damping are close in value, leading to a reduced surface effect.

In summary, the effective Gilbert damping dependence on temperature for Nd/Py bilayers is systematically studied using atomistic spin dynamics simulations and TR-MOKE measurements. When the interface spin accumulation is large, as in the case of the Nd/Py bilayer, the effective Gilbert damping shows that the coefficient of the temperature dependence is negative due to surface spin fluctuations separating the dynamics of the surface and bulk layers. Conversely, when interface spin accumulation is small, as seen in Pt/Py and $\text{Pt}_{0.68}\text{Nd}_{0.32}$ /Py bilayers, the effective Gilbert damping shows a positive correlation with temperature due to the usual thermal fluctuations. The ability to engineer different temperature coefficients of Gilbert damping is particularly useful for deep-nanoscale spintronic devices, especially when operating at elevated temperatures. Other rare-earth metals may also exhibit similar effects, providing a rich optimization space for different materials and devices.

This work is supported by the National Natural Science Foundation of China (Grants No. 52071079, No. 12274071, and No. 12241403); the National Natural Science Foundation of China (Grant No. 12104216); and the National Key R&D Program of China (Grant No. 2021YFB3601600). We gratefully acknowledge computational resources from the VIKING computer system used in this project, which is a high-performance computing facility provided by the University of York.

L.C. performed sample preparation, measurements, atomistic simulations, analyzed the data, and wrote the manuscript; R.F.L.E. and R.W.C. performed atomistic and spin accumulation calculations, analyzed the data, and modified the manuscript; Y.G., X.L., Y.X., and J.W. performed the sam-

ple measurement. Y.Z., J.W., R.F.L.E. and R.W.C. performed project administration and supervision.

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this Letter.

Data availability. The data that support the findings of this study were generated by numerical simulations. The source code used to generate the simulations is publicly available at the VAMPIRE website [34] and the input files are available online at [35]. Final processed data and fits to the data are available in MS Excel format at [35].

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- [35] The data supporting this research is openly available from the research data repository of the University of York at <https://doi.org/10.15124/254bf8a1-42c7-4ea0-ba57-aa35ff9e5928>.