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Probe of spin dynamics in superconducting NbN thin films via spin pumpingYunyan Yao,^{1,2} Qi Song,^{1,2} Yota Takamura,^{3,4} Juan Pedro Cascales,³ Wei Yuan,^{1,2} Yang Ma,^{1,2} Yu Yun,^{1,2} X. C. Xie,^{1,2} Jagadeesh S. Moodera,^{3,5} and Wei Han^{1,2,*}¹*International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, China*²*Collaborative Innovation Center of Quantum Matter, Beijing 100871, China*³*Plasma Science and Fusion Center and Francis Bitter Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*⁴*School of Engineering, Tokyo Institute of Technology, Tokyo 152–8550, Japan*⁵*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*

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The emerging field of superconductor (SC) spintronics has attracted intensive attentions recently. Many fantastic spin-dependent properties in SCs have been discovered, including large magnetoresistance, long spin lifetimes, and the giant spin Hall effect, etc. Regarding the spin dynamics in superconducting thin films, few studies have been reported yet. Here, we report the investigation of the spin dynamics in an *s*-wave superconducting NbN film via spin pumping from an adjacent insulating ferromagnet GdN film. A profound coherence peak of the Gilbert damping of GdN is observed slightly below the superconducting critical temperature of the NbN, which agrees well with recent theoretical prediction for *s*-wave SCs in the presence of impurity spin-orbit scattering. This observation is also a manifestation of the dynamic spin injection into superconducting NbN thin film. Our results demonstrate that spin pumping could be used to probe the dynamic spin susceptibility of superconducting thin films, thus pave the way for future investigation of spin dynamics of interfacial and two-dimensional crystalline SCs.

DOI: [10.1103/PhysRevB.97.224414](https://doi.org/10.1103/PhysRevB.97.224414)**I. INTRODUCTION**

The interplay between superconductivity and spintronics has been intensively investigated in the last decade [1–3]. In the content of superconductivity, the ferromagnetic magnetization has been found to play an important role in the superconducting critical temperature (T_C) in the ferromagnet (FM)/superconductor (SC) junctions [4–9]. Besides, unusual spin-polarized supercurrent has been observed in ferromagnetic Josephson junctions [2,3,10–13]. Furthermore, non-Abelian Majorana fermions have been proposed for $p_x + ip_y$ superconductor surfaces driven by magnetic proximity effect, which has the potential for topological quantum computation [14]. In the content of spintronics [15,16], the Cooper pairs in SCs have exhibited many fantastic spin-dependent properties that are promising for future information technologies. For instance, large magnetoresistance has been achieved in the spin valve with SCs between two ferromagnetic films [7,17], extremely long spin lifetimes and large spin Hall effect have been discovered for the quasiparticles of SCs, which exceeds several orders compared to the normal states above T_C [18,19]. Despite these intensive studies of the spin-dependent properties in the FM/SC junctions, the spin dynamics of the superconducting thin films has not been reported yet.

Very intriguingly, it has been proposed recently to investigate the spin dynamics in superconducting films via spin pumping [20], a well-established technique to perform dynamic spin injection and to probe the dynamic spin susceptibility

in various materials, including metal, semiconductors, Rashba 2DEGs, and topological insulators, etc. [21–30].

In this paper, we report the experimental investigation of the spin dynamics in *s*-wave superconducting NbN thin films via spin pumping. A profound coherence peak of the Gilbert damping of GdN is observed slightly below the T_C of the NbN in the NbN/GdN/NbN trilayer samples, which indicates dynamic spin injection into the NbN thin films. Besides, the interface-enhanced Gilbert damping probes dynamic spin susceptibility in the NbN thin films in the presence of impurity spin-orbit scattering, which is consistent with the recent theoretical study [20]. Our experimental results further demonstrate that spin pumping could be a powerful tool to study the spin dynamics in the emerging two dimensional SCs [31–33].

II. EXPERIMENTAL DETAILS

The NbN (t)/GdN (d)/NbN (t) trilayer samples are grown on Al_2O_3 (~5 nm)-buffered thermally oxidized Si substrates by dc reactive magnetron sputtering at 300 °C in an ultrahigh vacuum chamber. The NbN layers are deposited from a pure Nb target (99.95%) in Ar and N_2 gas mixture at a pressure of 2.3 mTorr (20% N_2), and GdN films are deposited from a pure Gd target (99.9%) in Ar and N_2 gas mixture at a pressure of 2.8 mTorr (6% N_2). The NbN and GdN layers are of textured crystalline quality with a preferred direction along (111) orientation, as evidenced by x-ray diffraction results (see Fig. S1 in Ref. [34]). After the growth, a thin Al_2O_3 layer (~10 nm) was deposited *in situ* as a capping layer to avoid sample degradation with air exposure. The Curie temperature (T_{Curie}) of the GdN film was determined via the offset magnetization measurement as

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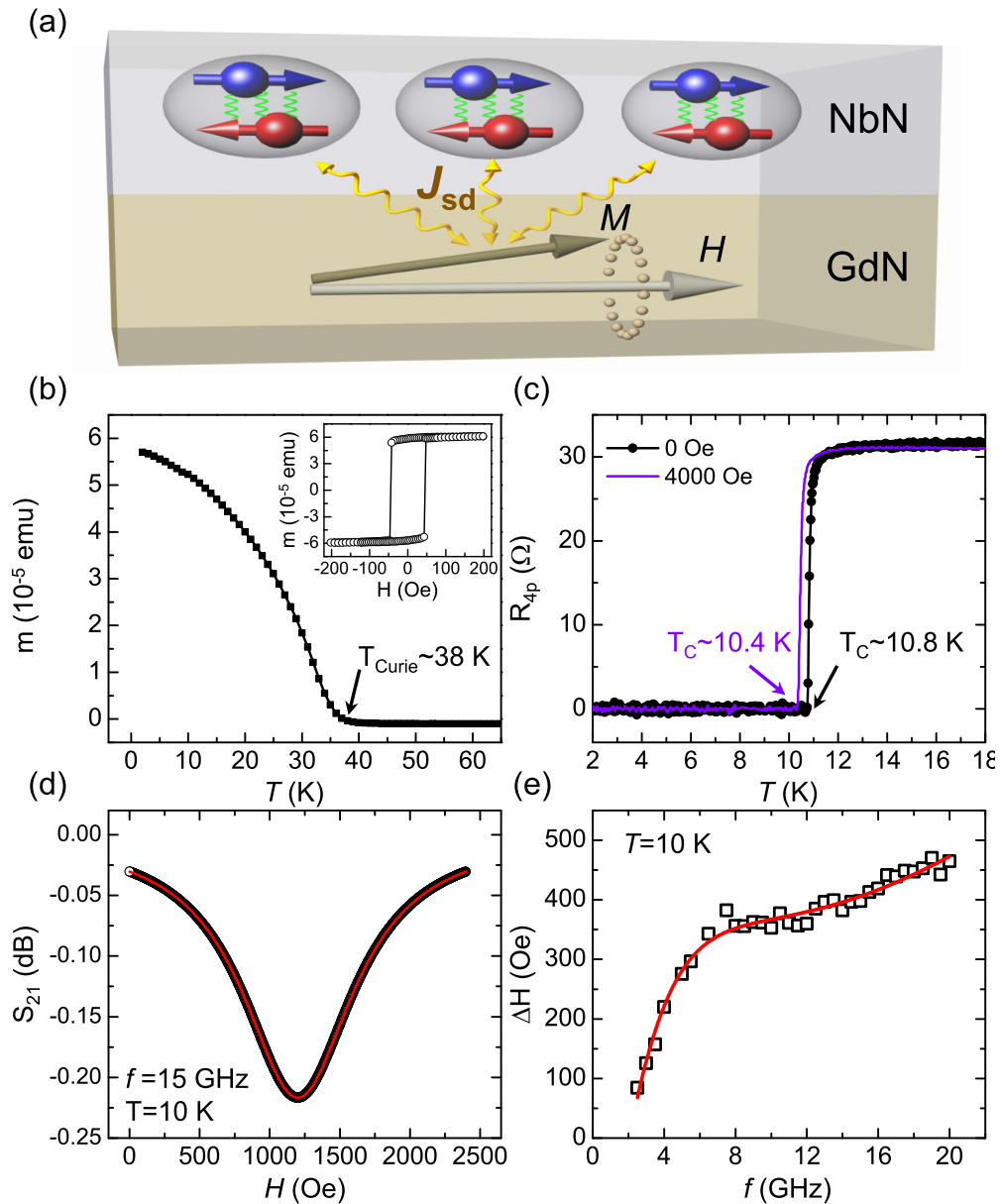


FIG. 1. Spin pumping into the superconducting NbN thin films. (a) Schematic of the interfacial s - d exchange interaction (J_{sd}) between the spins in the NbN layer and the rotating magnetization of the GdN layer under the ferromagnetic resonance (FMR) conditions. (b) The magnetic moment as a function of the temperature. Inset: the magnetic hysteresis loop at $T = 5$ K. (c) The four-probe resistance as a function of the temperature with in-plane magnetic field at 0 and 4000 Oe. (d) The typical FMR signal measured at $T = 10$ K and $f = 15$ GHz. The red line indicates the Lorentz fitting curve to obtain the half linewidth based on Eq. (1). (e) The half linewidth as a function of the microwave frequency at $T = 10$ K. The red solid line is the fitting curve based on spin-relaxation model. The results in (b)–(d) are obtained on the NbN (10)/GdN (5)/NbN (10) sample.

a function of the temperature using a Magnetic Properties Measurement System (MPMS; Quantum Design). The T_C of the SC NbN thin films was measured by four-probe resistance technique as a function of the temperature in a Physical Properties Measurement System (PPMS; Quantum Design) using standard ac lock-in technique at low frequency of 7 Hz. The FMR spectra of the multilayer samples were measured using the coplanar wave guide technique with a vector network analyzer (VNA, Agilent E5071C) in the variable temperature insert of PPMS. The samples were attached to the coplanar wave guide using insulating silicon paste. During the mea-

surement, the amplitudes of forward complex transmission coefficients (S_{21}) were recorded as a function of the in-plane magnetic field from ~ 4000 to 0 Oe at various temperatures under different microwave frequencies and microwave power of 5 dBm.

III. RESULTS AND DISCUSSION

Figure 1(a) illustrates spin pumping and the interfacial s - d exchange coupling between spins in the NbN layer and magnetic moments in the GdN layer. Due to the interfacial s -

exchange interaction (J_{sd}), the time-dependent magnetization in the GdN layer pumps a quasiparticles-mediated spin current into the NbN layer, and the spin-flip scattering of quasiparticles accompanies a quantum process of magnon annihilation in the GdN layer, giving rise to enhanced Gilbert damping [20]. Hence, the interfacial s - d exchange coupling provides the route to detect the spin dynamics of the NbN layer by measuring the magnetization dynamics of GdN. The spin pumping is performed by measuring Gilbert damping of the GdN film in the NbN (t)/GdN (d)/NbN (t) trilayer heterostructures via ferromagnetic resonance (FMR) technique [35]. NbN is an s -wave SC with short coherence length of ~ 5 nm and spin diffusion length of ~ 7 nm [19,36]. GdN is an insulating FM. A 10-nm NbN film is used for the NbN (t)/GdN (d)/NbN (t) samples to justify the assumption that the spin backflow from SC is small in the theoretical study [20]. In a typical sample of NbN (10)/GdN (5)/NbN (10) (with thickness in nanometers), T_{Curie} of GdN is determined to be ~ 38 K from the temperature dependence of magnetic moments [Fig. 1(b)], and T_C of NbN is obtained to be ~ 10.8 K via resistivity versus temperature measurement [Fig. 1(c)]. The T_C is slightly affected by the external in-plane magnetic field of 4000 Oe due to the large critical field.

Figure 1(d) shows a typical FMR signal (S_{21}) versus the magnetic field measured at $T = 10$ K, with a microwave excitation frequency (f) of 15 GHz. The half linewidth (ΔH) could be obtained by the Lorentz fitting of the FMR signal following the relationship [37]

$$S_{21} \propto S_0 \frac{(\Delta H)^2}{(\Delta H)^2 + (H - H_{\text{res}})^2}, \quad (1)$$

where S_0 is the coefficient for the transmitted microwave power, H is the external magnetic field, and H_{res} is the resonance magnetic field. Gilbert damping (α) is determined using numerical fitting of ΔH versus f [Fig. 1(e)] based on the spin-relaxation mechanism [34,38]:

$$\Delta H = \Delta H_0 + \frac{4\pi\alpha f}{\gamma} + A \frac{2\pi f \tau}{1 + (2\pi f \tau)^2}, \quad (2)$$

where ΔH_0 is related to the inhomogeneous properties, γ is the gyromagnetic ratio, A is the spin-relaxation coefficient, and τ is the spin-relaxation time constant (Fig. S2 in Ref. [34]).

The Gilbert damping of GdN is studied as a function of the temperature for two trilayer samples with the same interface: NbN (2)/GdN (5)/NbN (2) and NbN (10)/GdN (5)/NbN (10).

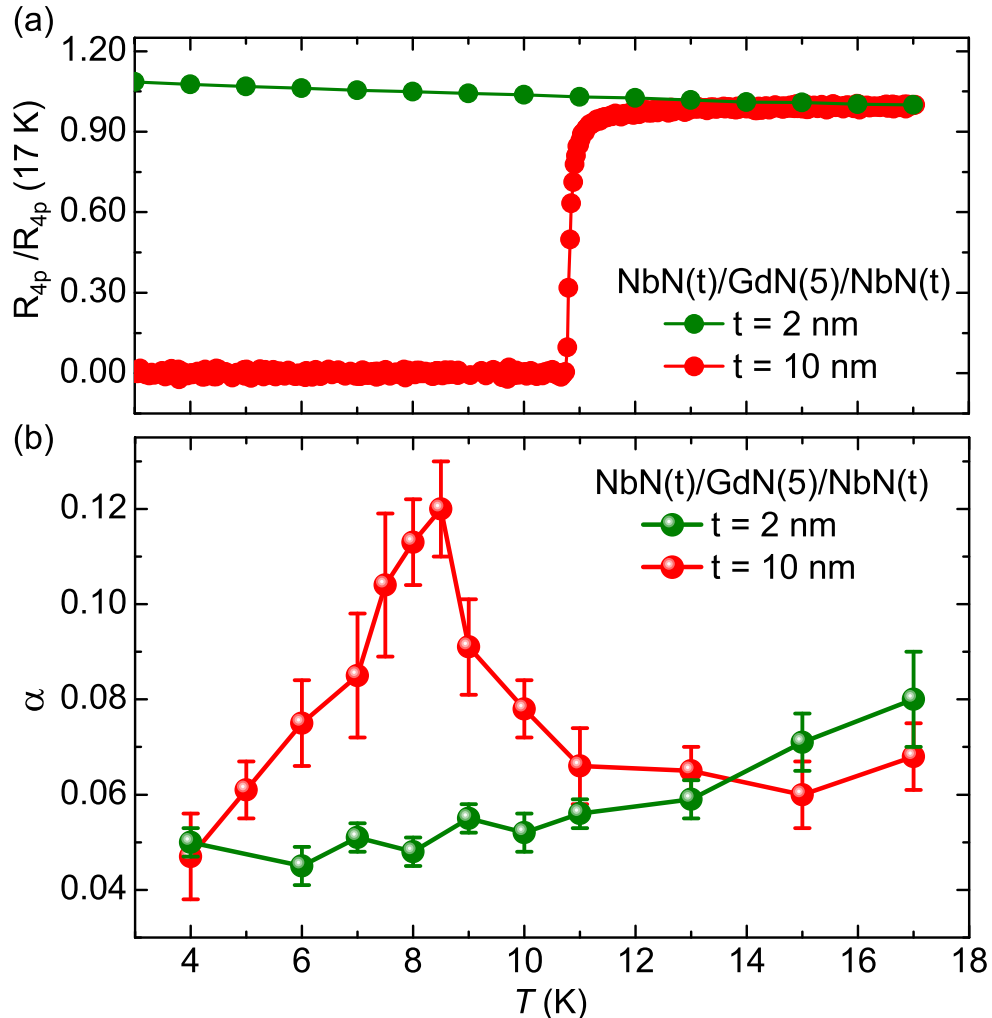


FIG. 2. Spin dynamics of the superconducting NbN thin films probed via spin pumping. The normalized four-probe resistance (a) and Gilbert damping (b) as a function of the temperature for the samples of NbN (2)/GdN (5)/NbN (2) and NbN (10)/GdN (5)/NbN (10), respectively.

The 2-nm NbN layer is not superconducting down to 2 K, while the NbN (10)/GdN (5)/NbN (10) exhibits a T_C of ~ 10.8 K [Fig. 2(a)]. The nonsuperconducting feature of 2-nm NbN sample and lower T_C of the 10-nm NbN sample compared to over 16 K for a single-crystalline NbN could be attributed to reduced thickness, polycrystalline property, and magnetic proximity effect [1,36,39]. Interestingly, a profound coherence peak of the Gilbert damping is observed in the NbN (10)/GdN (5)/NbN (10), but not on NbN (2)/GdN (5)/NbN (2), as shown in Fig. 2(b). This feature is also evident from the temperature dependence of the half linewidth (Fig. S3 in Ref. [34]). The peak of the Gilbert damping in the NbN (10)/GdN (5)/NbN (10) is observed at ~ 8.5 K, which is slightly below the T_C (~ 10.8 K) of the 10-nm NbN layers. These results indicate the successful dynamic spin injection into the 10-nm NbN superconducting layer, which authenticates a charge-free method to inject spin-polarized carriers into SCs beyond previous reports of electrical spin injection [18,40–42]. Furthermore, the observation of the profound coherence peak at $T \approx 0.8 T_C$ is expected based on recent theoretical studies of the spin dynamics for s -wave superconducting thin films via spin pumping [20]. Since the thickness of NbN layer ($d = 10$ nm) is longer than its spin diffusion length of ~ 7 nm, the spin backflow effect, which is expected to reduce the damping peak, is negligible here [20,21]. According to the theory, Gilbert damping is related to the interfacial s - d exchange interaction and the imaginary part of the dynamic spin susceptibility of the SC [20]:

$$\delta\alpha \propto J_{sd}^2 \sum_q \text{Im}x_q^R(\omega). \quad (3)$$

For the s -wave superconducting NbN thin films, the superconducting gap Δ forms below T_C . At the temperature slightly below T_C , two coherence peaks of the density states exist around the edge of the superconducting gap following the BCS theory [43], and these peaks in turn give rise to the enhancement of the dynamic spin susceptibility in the presence of impurity spin-orbit scattering. Quantitatively, the ratio of the peak value of Gilbert damping over the value at T_C is ~ 1.8 , which also agrees well with the theoretical calculation using previous experimental values of spin diffusion length (~ 7 nm), phase coherence length (~ 5 nm), and mean free path (~ 0.3 nm) for NbN thin films [19,20,36]. As the temperature further decreases, the number of quasiparticles decreases rapidly as the Δ grows, giving rise to the fast decrease of Gilbert damping below $T \approx 7$ K.

Next, the thickness of the GdN layer is varied to further study spin pumping and the spin dynamics in NbN layer. For both samples of NbN (10)/GdN (d)/NbN (10) with $d = 10$ and 30 nm, a profound coherence peak of Gilbert damping is observed slightly below the superconducting temperature of NbN, as shown in Figs. 3(a) and 3(b) (red circles). While for all the samples of NbN (2)/GdN (d)/NbN (2), no such coherence peak of the Gilbert damping is noticeable [green circles in Figs. 3(a) and 3(b)]. The role of the effective magnetization in the observed coherence peak has been ruled out since it exhibits similar temperature dependence for NbN (2)/GdN (d)/NbN (2) and NbN (10)/GdN (d)/NbN (10) samples (Fig. S4 in Ref. [34]). Clearly, a more profound damping peak for the $d = 10$ nm sample is observed compared to $d = 5$ and

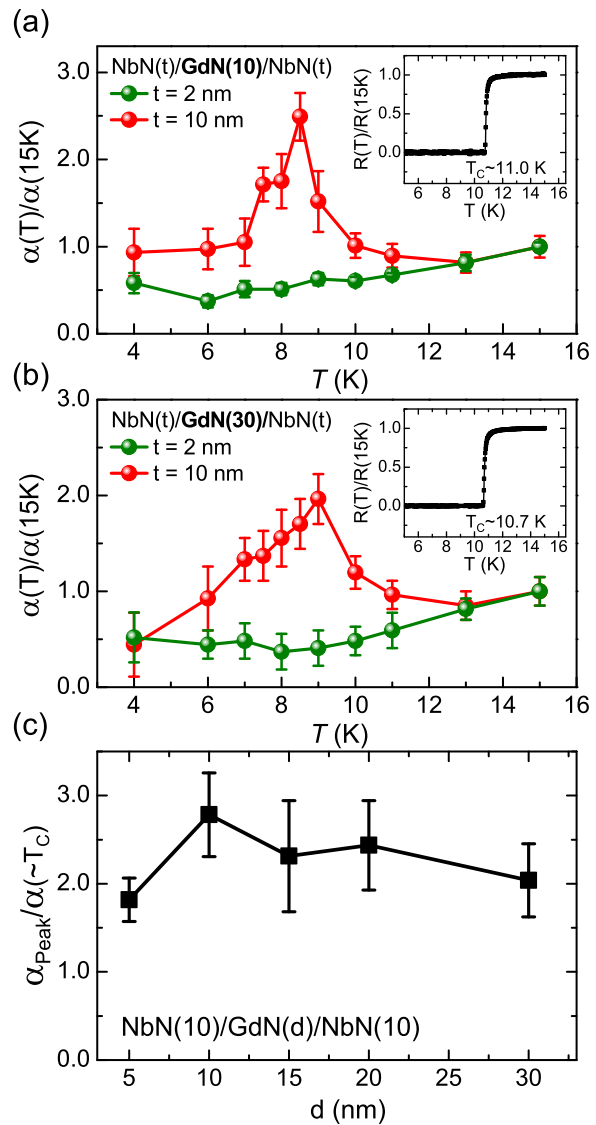


FIG. 3. The GdN thickness effect on spin dynamics of the superconducting NbN thin films. [(a) and (b)]. The Gilbert damping as a function of the temperature for the samples of NbN (t)/GdN (10)/NbN (t), and NbN (t)/GdN (30)/NbN (t). (Insets) The normalized four-probe resistance as a function of the temperature for the samples of NbN (10)/GdN (10)/NbN (10) and NbN (10)/GdN (30)/NbN (10). (c) The ratio of the peak Gilbert damping over the value at $T \approx T_C$ [$\alpha_{\text{peak}}/\alpha(\sim T_C)$] as a function of the GdN layer thickness.

30 nm samples. Figure 3(c) shows the ratio of the peak Gilbert damping over the value at $T \approx T_C$ as a function of the GdN thickness, which is in the range from 1.8 to 2.8. The T_C exhibit little variation as a function of the GdN thickness (Fig. S5 in Ref. [34]). For a deeper understanding of the underlying mechanism to account for the thickness dependence of the ratio, further theoretical and experimental studies would be essential. One possible cause might be related to the interface proximity exchange effect and/or the presence of magnetic loose spins leading to scattering at the interface, which could affect the spin diffusion length, coherence length, and mean free path of the NbN layer.

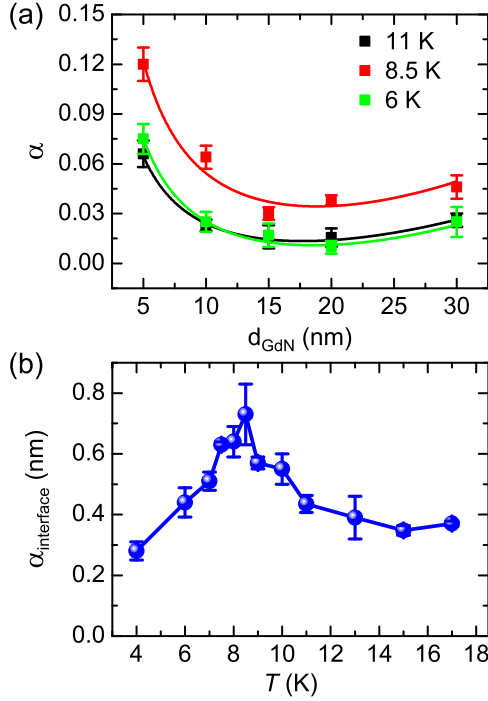


FIG. 4. The interface-induced Gilbert damping at the NbN/GdN interface. (a) The Gilbert damping as a function of the GdN thickness for the samples of NbN (10)/GdN (d)/NbN (10) at $T = 11, 8.5,$ and 6 K, respectively. (b) The interface-induced Gilbert damping as a function of temperature for NbN (10)/GdN (d)/NbN (10).

To further confirm that the observed coherence peak of the Gilbert damping arises from the s - d exchange interaction at the SC/FM interface, the interface-induced Gilbert damping (α_S) is studied as a function of temperature. α_S is obtained from the thickness dependence of total Gilbert damping at each temperature [34], as shown in Fig. 4(a). The contribution from GdN itself is significantly small compared to α_S . The unperturbed peak at ~ 8.5 K [Fig. 4(b)] unambiguously demonstrates that the origin of the coherence peak in the Gilbert damping is indeed due to the interfacial s - d exchange interaction between the magnetization of GdN and the spins of the quasiparticles in superconducting NbN thin films. The slightly upturn of the interface-induced Gilbert damping starting at $T = 11$ K might be associated with the fluctuation superconductivity and/or the higher superconducting transition temperature for some regions of the NbN films than the zero-resistance temperature. For comparison, the interface-induced Gilbert damping in the samples of NbN (2)/GdN (d)/NbN (2) does not exhibit any signature of coherence peak between 4 and 15 K (Fig. S6 in Ref. [34]).

Noteworthy is that our results are essentially different from previous reports of spin pumping into superconducting Nb films using the ferromagnetic metal permalloy (Py), where a monotonic decrease of the Gilbert damping is reported when the temperature decreases [44,45]. This feature is also confirmed in our studies by measuring the temperature dependence of Gilbert damping of Py in Nb(100)/Py(20)/Nb(100) (Fig. S7 in Ref. [34]). Since Py is a FM metal, the interface exchange

interaction would be significant to strongly or even completely suppress the superconducting gap of Nb at the Nb/Py interface [17,20,43]. Hence, no coherence peak of Gilbert damping is expected based on the theoretical study that assumes the completely suppression of the gap at the interface [45]. Besides, the strong suppression of the superconducting gap at the Nb/Py interface is in good agreement of previous tunnel spectroscopy measurements of vanishing superconducting gap at the interface between Nb and Ni [46]. However, for FM insulating GdN, charge carriers from NbN do not penetrate into GdN, thus not weakening the superconductivity at the interface, resulting in the survival of the superconducting gap [47,48].

The experimental demonstration of the dynamic spin susceptibility in superconducting thin films via spin pumping could be essential for the field of superconductors. For bulk SCs, dynamic spin susceptibility has been studied from the temperature dependent spin relaxation rate via the nuclear magnetic resonance [43,49,50]. It also provides an avenue for identifying unconventional superconducting pairing mechanisms [51], while limited to mostly bulk SCs due to low signal-to-noise ratio [43,49–51]. The spin pumping method has much better signal-to-noise ratio so it could be very sensitive to probe the dynamic spin susceptibility of superconducting thin films. Furthermore, the spin pumping offers a special technique to probe the pair breaking strength, the impurity spin-orbit scattering, and the magnetic proximity effect in the SC/FM junctions, which is also of considerable interest for the field of SC spintronics [1–3].

IV. CONCLUSION

In conclusion, the spin dynamics of superconducting NbN films are investigated via spin pumping from an adjacent FM insulating layer. A profound coherence peak of the Gilbert damping is observed below T_C , which indicates the dynamic spin injection into the superconducting NbN films. Our results pave the way for future investigation of interface impurity spin-orbit scattering, and pair breaking strength in FM/SC junctions, as well as the spin dynamics in the interfacial and two-dimensional crystalline SCs. It may also be useful for the search for Majorana fermions in the SC/ferromagnetic insulator heterostructures.

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