Letter

Significant modulation of Gilbert damping in ultrathin ferromagnetic films by altering the surface magnetic anisotropy

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The ability to control the Gilbert damping which determines the lifetime of spin information is crucial for designing spintronic and magnonic devices. Thus, controlling the Gilbert damping parameter α has been a significant research target for several decades. Although numerous approaches have been explored to control α, few reports of large changes of this parameter have been presented. Herein, we demonstrate significant change of α in 2-nm-thick $Co_{25}Fe_{75}$ films originating from uniaxial surface magnetic anisotropy, which affects the two-magnon scattering. We report a change in α by approximately 0.02, or 300%. The value for α and its change are comparable to those observed in a previous study using a film that is one order of magnitude thicker. Our results achieved with $Co_{25}Fe_{75}$ can be directly transferred to other ultrathin ferromagnetic materials, which are a promising platform for spin information processing, and thus represents a versatile approach to modulate the Gilbert damping.

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Magnetization damping is one of the crucial parameters for the design of spintronic and magnonic devices as well as for quantum hybrid systems based on magnons [1–5]. In addition, the damping also characterizes the usability of the materials for these research directions. Whereas strong magnetization damping is desirable for faster magnetization switching, a material with weak magnetization damping is beneficial for long-lived magnon excitations and their transport [6–9]. This directly signifies the importance to control and engineer magnetization damping in spintronics, magnonics, and quantum technologies. The control of magnon damping over a broad range helps to create magnonic devices with intriguing functionalities that apply to novel spin devices such as the magnon transistor [8,9] and tunable magnon-photon coupling systems [10]. Therefore, the engineering of magnetization damping (i.e., tunable magnetization damping) is a significant research interest. Indeed, altering the composition of ferromagnetic (FM) alloys [11–14] and the optimization of buffer layers beneath the FM layer [15,16] yielded controllability of magnetization damping. In addition, recent studies on magnetization damping in bulklike FM films [17–19] have addressed possibilities of efficient tuning mechanisms without changing the film shape or thickness. Considering the significance of magnetization damping control, in particular for classical and quantum applications, warrants the development of an *in situ* control of the magnetization damping rate, preferably by gate tuning. In particular, gate tuning is considered for ultrathin magnetic films (with a thickness of less than 2 nm) [20].

To construct a material platform suitable for gate-tunable magnetization damping, we focus on the manipulation of magnetization damping in ultrathin conducting FM films. Here, the Gilbert damping constant α is [21]

$$\alpha = \alpha_{\mathrm{int}} + \alpha_{\mathrm{SP}} t_{\mathrm{FM}}^{-1} + \beta_{\mathrm{TMS}} t_{\mathrm{FM}}^{-2}, \tag{1}$$

where t_{FM} is the thickness of the FM layer and α_{int}, α_{SP}, and β_{TMS} parametrize the intrinsic, spin-pumping, and two-magnon scattering (TMS) contribution to the Gilbert damping, respectively. Notably, TMS control has not been elucidated, whereas modulation of α_{SP} in an FM film has been well studied [22–29]. However, there are some advantages to controlling β_{TMS} in ultrathin FM films: (1) Large modulation of α can be realized because the TMS contribution $\beta_{\mathrm{TMS}} t_{\mathrm{FM}}^{-2}$ can dominate in ultrathin FM films for certain magnetic-field orientations. (2) Controlling magnetic anisotropy by conventional methods, such as the application of electrical voltages [30–34] and surface engineering at heavy-metal/FM and MgO/FM interfaces [35–38], is expected to allow modulation of β_{TMS} as β_{TMS} is proportional to the square of the uniaxial surface magnetic anisotropy (USMA) field [21,39–41]. Furthermore, despite dedicated efforts to modulate the Gilbert damping by controlling surface magnetic anisotropy (SMA), the physical origin of modulation remains unclear, and the modulation amplitude of the Gilbert damping constant $\Delta\alpha$ has been limited to within 0.002 [42,43]. Hence, clarifying the origin of an altered Gilbert damping due to the change of SMA is essential to determine a guiding principle

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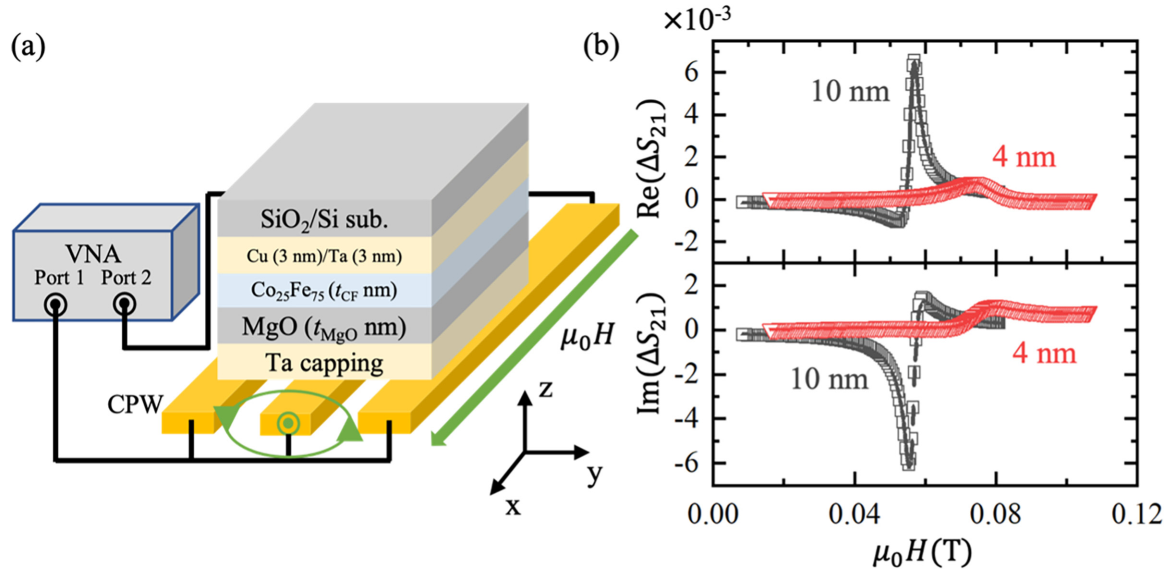

FIG. 1. (a) Schematic image of measurement setup for bbFMR. The S_{21} of the CPW on which the sample is attached was measured by VNA. (b) Real and imaginary parts of ΔS_{21} for the samples with 10-nm (black)- and 4-nm (red)-thick $Co_{25}Fe_{75}$ and a 2-nm-thick MgO capping layer, using a microwave frequency of 10 GHz.

for achieving larger modulation of the Gilbert damping in ultrathin FM films that are applicable to gate-tunable magnon devices. Herein, we report the substantially large modification of α originating from TMS contribution $\beta_{\mathrm{TMS}}t_{\mathrm{FM}}^{-2}$ (0.02) in ultrathin $Co_{25}Fe_{75}$ films (2-nm thickness) with an intrinsically low Gilbert damping (4.0×10^{-3}) by tuning the USMA field of the FM layer. Furthermore, the origin of large modulation is confirmed to stem from the change in USMA. The method of modulating α by controlling TMS is widely applicable to all FM films and can be utilized to facilitate further progress in spintronic/magnonic devices.

We fabricated Ta capping/MgO (t_{MgO} nm)/$Co_{25}Fe_{75}$ (t_{CF} nm)/Cu (3 nm)/Ta (3 nm) multilayers, which were deposited *in situ* on a thermally oxidized Si substrate by dc and rf magnetron sputtering at room temperature (for more details of the deposition process, see Supplemental Material [44]). The thicknesses of the MgO and $Co_{25}Fe_{75}$ were varied from 0.4 to 4.0 nm and from 2 to 20 nm, respectively. We performed broadband FM resonance (bbFMR) measurements at room temperature, where an in-plane external magnetic field was applied. Figure 1(a) shows a schematic image of the measurement setup for bbFMR. The prepared samples were mounted face down onto a coplanar waveguide (CPW), and the complex microwave transmission S_{21} was measured using a vector network analyzer (VNA). The microwave frequency f ranged from 5 to 20 GHz, and the in-plane external magnetic field was swept from 0 to 0.3 T. The microwave power was set to 0 dBm for all bbFMR measurements. Details concerning the experiment can be found in Ref. [45].

Figure 1(b) depicts the real and imaginary parts of the background-corrected transmission, denoted as $\Delta S_{21} = S_{21} - S_{21}^0$, obtained from the samples with 4- and 10-nm-thick $Co_{25}Fe_{75}$ layers and a 2-nm-thick MgO capping layer, where S_{21}^0 is the background signal represented by a linear function. In a thin FM film, the Gilbert damping constant and magnetic anisotropy are easily influenced by altering the surface roughness and SMA, and thus manifest as changes to linewidth and resonance field in the bbFMR signal. Indeed, the signals of ΔS_{21} for the 4-nm-thick $Co_{25}Fe_{75}$ have a larger linewidth and resonance field than those of the 10-nm-thick $Co_{25}Fe_{75}$. To investigate magnetization damping and magnetic anisotropy from our field-dependent complex microwave transmission signals, we utilize a fit based on the Polder susceptibility (for details see Eqs. (S13)–S(15) in Supplemental Material [44,46,47]) to extract the resonance field H_{res} and half-width at half-maximum magnetic-field linewidth ΔH of the FM resonance for different fixed microwave frequencies. Following this procedure, we obtain for $\mu_0 H_{\mathrm{res}}$ and $\mu_0 \Delta H$ for our 10-nm (4-nm) $Co_{25}Fe_{75}$ films 56.3 mT (75.1 mT) and 3.2 mT (12.6 mT), respectively. This observation is rationalized due to the enhancement of the magnetization damping and effective magnetic anisotropy with the decreasing thickness of the $Co_{25}Fe_{75}$ layer, as we discuss in the following.

The Gilbert damping constant can be determined from the frequency dependence of ΔH using the following equation [48]: $\mu_0 \Delta H = \mu_0 \Delta H_0 + \frac{\alpha}{\gamma}\omega$, where $\mu_0 \Delta H_0$ and γ are the inhomogeneous linewidth and gyromagnetic ratio, respectively and $\omega = 2\pi f$. The microwave frequency dependence of the extracted ΔH obtained from the bbFMR measurement is shown in Fig. 2(a) for our samples with varying $Co_{25}Fe_{75}$ and MgO capping-layer thicknesses. We observe that the FMR linewidth increases for decreasing thicknesses of the $Co_{25}Fe_{75}$ layer for all MgO layer thicknesses. In addition, we find that α also increases with the thickness of the MgO capping layer.

This enhancement is achieved when decreasing the FM thickness t_{FM} can be attributed to surface magnetization damping originating from spin pumping from the FM into the adjacent nonmagnetic layer [49] and/or TMS [39], as captured by Eq. (1). Here, the magnetic proximity effect (MPE) term on Gilbert damping from Ta buffer layers as discussed

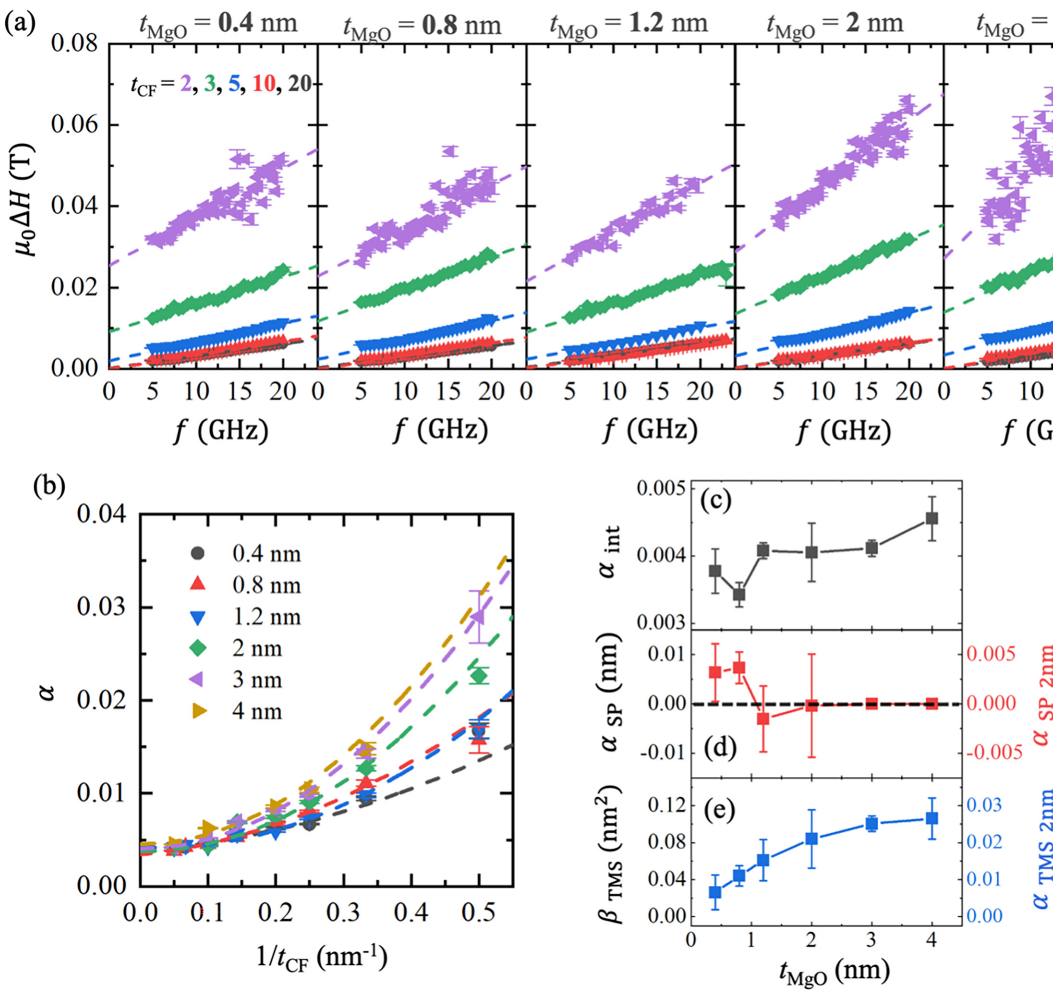

FIG. 2. (a) Microwave frequency dependences of the half-width at half maximum obtained by fitting the FMR signals of the samples with different MgO and $Co_{25}Fe_{75}$ thicknesses. The broken line shows the fitting obtained using the Polder susceptibility (see details in Supplemental Material [44]). (b) $Co_{25}Fe_{75}$ thickness dependence of the estimated Gilbert damping constant. The broken line shows the fitting obtained using Eq. (1). MgO thickness dependences of (c) α_{int}, (d) α_{SP}, and (e) β_{TMS}. The right axes of Figs. 2(d) and 2(e) show the Gilbert damping constants corresponding to that of the 2-nm-thick $Co_{25}Fe_{75}$ film.

in Ref. [50] is negligible because 3-nm-thick Cu interlayer is sufficient to eliminate MPE between Ta layer and CoFe alloys [51]. To distinguish between the two effects (α_{SP} and β_{TMS}) and assess the impact of the MgO capping-layer thickness, we plot the Gilbert damping constant as function of the $Co_{25}Fe_{75}$ thickness t_{CF} [see Fig. 2(b)] and fit these data using Eq. (1) to determine α_{int}, α_{SP}, and β_{TMS} [see Figs. 2(c)–2(e)]. In detail, the obtained α_{int} of the $Co_{25}Fe_{75}$ for different MgO capping-layer thicknesses is approximately 4.0×10^{-3}, as shown in Fig. 2(c), which is consistent with the values reported in previous studies [11,13,14]. The α_{SP} of the 2-nm-thick $Co_{25}Fe_{75}$ is negligible compared to the other damping contributions when the MgO thickness exceeds 1 nm since the α_{SP} of the $Co_{25}Fe_{75}$ with MgO capping layer over 1 nm thick are zero within the margin of error [see Fig. 2(d)]. Meanwhile, the α_{SP} is sizable when the MgO layer is 0.4 and 0.8 nm in thickness, which might be due to a finite spin pumping from the $Co_{25}Fe_{75}$ into the Ta capping layer, as reported in Ref. [52]. One interpretation could be that those MgO thicknesses still include pinholes enabling spin pumping into the Ta capping layer [49]. The evolution of β_{TMS} with MgO thickness is shown in Fig. 2(e). The TMS coefficient β_{TMS} monotonically increases with the increasing thickness of the MgO capping layer up to 2 nm [see Fig. 2(e)], which is attributed to the fact that TMS is modulated by altering the USMA as reported in a previous study [38]. Significantly, the TMS contribution is tunable by changing the MgO capping-layer thickness and the modulation ratio of β_{TMS} is approximately 300%. The Gilbert damping α stemming from TMS was altered from 0.0065 to 0.0265. Moreover, the modulation range $\Delta\alpha$ is comparable to that observed in a recent study using 20-nm-thick FM films [17].

Two-magnon scattering essentially depends on USMA and surface roughness [39]. Since the magnitude of β_{TMS} is proportional to the square of the USMA field [41], the giant modulation of β_{TMS} can be interpreted as an increase of the USMA field at the MgO/$Co_{25}Fe_{75}$ interface as a function of the MgO capping-layer thickness. An independent quantification of the magnetic anisotropies in thin FM films can be obtained from the dispersion of the FMR which are shown in Fig. 3(a). We find that thinner $Co_{25}Fe_{75}$ films show a modification from the thick ($t_{CF} = 20$nm) behavior, hinting an

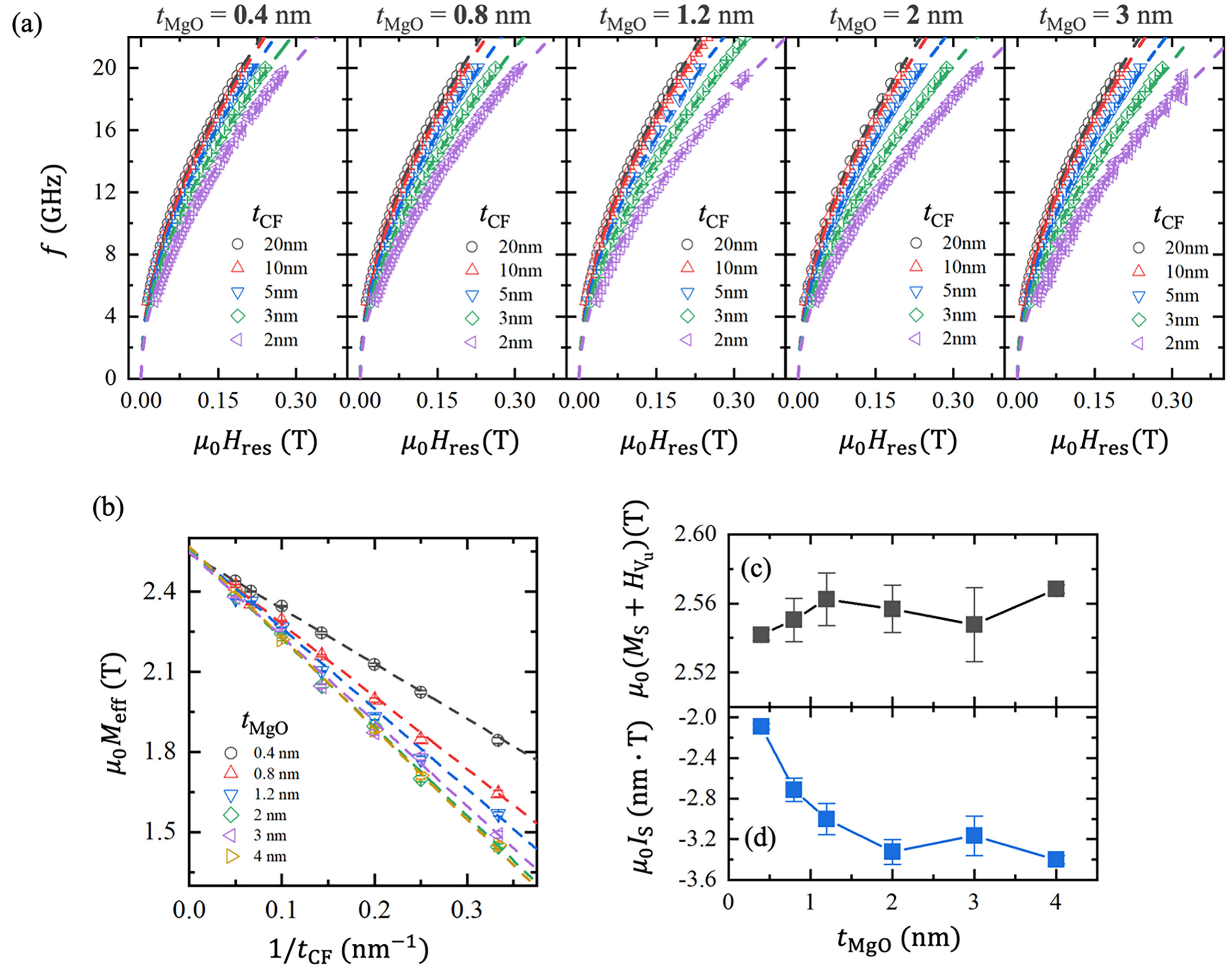

FIG. 3. (a) Microwave frequency dependences of the resonance field obtained by fitting the FMR signals of the samples with different MgO and $Co_{25}Fe_{75}$ thicknesses. The broken lines depict fitting obtained using Eq. (2). (b) $Co_{25}Fe_{75}$ thickness dependence of the effective magnetization. MgO thickness dependences of (c) the effective magnetic anisotropy field $\mu_0 H_{eff}$, and (d) the SMA field $\mu_0 H_S$, where the black dashed line shows zero value.

interface contribution to the total magnetic anisotropy. When comparing different MgO layer thicknesses, we find that this interface-like contribution increases [see Fig. 3(a) subpanels for $t_{MgO} = 3$ nm]. Thus, the MgO layer thickness seems to impact the USMA field, showing an increasing trend for increasing t_{MgO}.

We quantify an effective thickness-dependent anisotropy parameter by fitting the Kittel mode for the in-plane configuration to the data presented in Fig. 3(a). In detail, we use [53]

$$f = \frac{\gamma}{2\pi}\sqrt{\mu_0 H(\mu_0 H + \mu_0 M_{eff})}, \tag{2}$$

where f and M_{eff} are the microwave frequency and effective magnetization, respectively. Figure 3(b) shows M_{eff} as function of the inverse $Co_{25}Fe_{75}$ layer thickness. Note that Eq. (2) already accounts for the thin-film limit with the static magnetic field applied in plane. The effective magnetization M_{eff} summarizes contributions like the saturation magnetization M_S, bulk- (H_{V_u}) and interface- (I_S) related anisotropies. This can be parametrized as follows [41]:

$$M_{eff} = (M_S + H_{V_u}) + I_S t_{CF}^{-1}. \tag{3}$$

Figure 3(c) shows the MgO capping-layer thickness dependence for $\mu_0(M_S + H_{V_u})$, which is determined from the value of M_{eff} at $1/t_{CF} = 0$, which we obtain by fitting Eq. (3) to the data. The MgO thickness only weakly affects $M_S + H_{V_u}$, as evident from Fig. 3(c). Thus, we imply that the insertion of the MgO capping layer does not alter the bulk magnetic properties ($M_S + H_{V_u}, \alpha_{int}$) in $Co_{25}Fe_{75}$. The absolute value of the estimated I_S, which contains the USMA, increases with the increasing MgO thickness and saturates when the thickness exceeds 2 nm [see Fig. 3(d)], suggesting that the modulation of the USMA field at the MgO/$Co_{25}Fe_{75}$ interface is controlled by the MgO capping-layer thickness. Since β_{TMS} exhibits a similar dependence for the MgO thickness, controlling USMA likely enables tunability of α in the ultrathin $Co_{25}Fe_{75}$ film. To corroborate this, the correlation between β_{TMS} and the square of the USMA field, represented via I_S^2, is

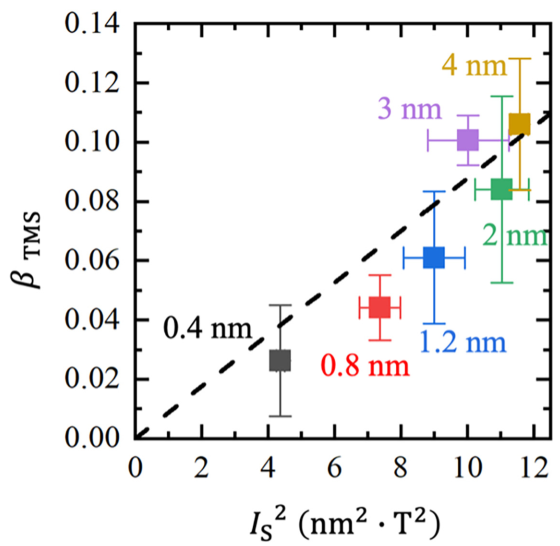

FIG. 4. Correlation between the TMS coefficient and the square of the USMA field represented by I_S^2.

displayed in Fig. 4, supporting the connecting between β_{TMS} and the USMA field. By subtracting α_{SP} from α, the modulation of α is estimated to be ranging from $\Delta\alpha = 0.0105 - 0.0305$, which is close to that observed in previous work using 20-nm-thick FM film, from $\Delta\alpha = 0.0054 - 0.0240$ [17]; however, here we studied an ultrathin FM film. Thus, our investigations indicate that the findings obtained for a 20-nm-thick FM film can be extended to the regime of ultrathin magnetic layers, with the advantage that this thickness regime is suited for gate-tunable Gilbert damping.

In conclusion, we demonstrated significant modulation of the Gilbert damping constant in ultrathin $Co_{25}Fe_{75}$ films by changing the thickness of the MgO layer. Moreover, our work hints at a connection between the increase in Gilbert damping and the change in the USMA field at the $Co_{25}Fe_{75}$/MgO interface, which both depend on the MgO thickness. The modulation amplitude of the Gilbert damping constant $\Delta\alpha$ in the 2-nm-thick $Co_{25}Fe_{75}$ film was approximately 0.02, without altering the bulk magnetic properties. The modulation principle of the Gilbert damping constant found in this study is versatile and applicable to gate-tunable magnon systems consisting of ultrathin conducting FMs and promises high tunability.

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