



CHORUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Room-temperature intrinsic and extrinsic damping in polycrystalline Fe thin films

Shuang Wu, David A. Smith, Prabandha Nakarmi, Anish Rai, Michael Clavel, Mantu K. Hudait, Jing Zhao, F. Marc Michel, Claudia Mewes, Tim Mewes, and Satoru Emori

Phys. Rev. B **105**, 174408 — Published 11 May 2022

DOI: [10.1103/PhysRevB.105.174408](https://doi.org/10.1103/PhysRevB.105.174408)

Room-Temperature Intrinsic and Extrinsic Damping in Polycrystalline Fe Thin Films

Shuang Wu,¹ David A. Smith,¹ Prabandha Nakarmi,² Anish Rai,² Michael Clavel,³ Mantu
K. Hudait,³ Jing Zhao,⁴ F. Marc Michel,⁴ Claudia Mewes,² Tim Mewes,² and Satoru Emori¹

¹*Department of Physics, Virginia Polytechnic Institute
and State University, Blacksburg, VA 24061, USA*

²*Department of Physics and Astronomy,
The University of Alabama, Tuscaloosa, AL 35487 USA*

³*Department of Electrical and Computer Engineering,
Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA*

⁴*Department of Geosciences, Virginia Polytechnic Institute
and State University, Blacksburg, VA 24061, USA*

Abstract

We examine room-temperature magnetic relaxation in polycrystalline Fe films. Out-of-plane ferromagnetic resonance (FMR) measurements reveal Gilbert damping parameters of ≈ 0.0024 for Fe films with thicknesses of 4-25 nm, regardless of their microstructural properties. [This observation runs counter to the intuition that various film defects heavily influence Gilbert damping.](#) The remarkable invariance with film microstructure suggests that room-temperature intrinsic Gilbert damping in [the Fe films is mostly fixed by the bcc crystal structure within the bulk of nanoscale grains](#), with limited impact from grain boundaries and film roughness. By contrast, the in-plane FMR linewidths of the Fe films exhibit distinct nonlinear frequency dependences, indicating the presence of strong extrinsic damping. To fit our in-plane FMR data, we have used a grain-to-grain two-magnon scattering model with two types of correlation functions aimed at describing the spatial distribution of inhomogeneities in the film. However, neither of the two correlation functions is able to reproduce the experimental data quantitatively with physically reasonable parameters. Our findings advance the fundamental understanding of intrinsic Gilbert damping in structurally disordered films, while demonstrating the need for a deeper examination of how microstructural disorder governs extrinsic damping.

29 I. INTRODUCTION

30 In all magnetic materials, magnetization has the tendency to relax toward an effective
31 magnetic field. How fast the magnetization relaxes governs the performance of a variety
32 of magnetic devices. For example, magnetization relaxation hinders efficient precessional
33 dynamics and should be minimized in devices such as precessional magnetic random access
34 memories, spin-torque oscillators, and magnonic circuits¹⁻⁴. From the technological perspec-
35 tive, it is important to understand the mechanisms behind magnetic relaxation in thin-film
36 materials that comprise various nanomagnetic devices. Among these materials, bcc Fe is
37 a prototypical elemental ferromagnet with attractive properties, including high saturation
38 magnetization, soft magnetism⁵, and large tunnel magnetoresistance^{6,7}. Our present study
39 is therefore motivated by the need to uncover magnetic relaxation mechanisms in Fe thin
40 films – particularly polycrystalline films that can be easily grown on arbitrary substrates for
41 diverse applications.

42 To gain insights into the contributions to magnetic relaxation, a common approach is to
43 examine the frequency dependence of the ferromagnetic resonance (FMR) linewidth. The
44 most often studied contribution is viscous Gilbert damping⁸⁻¹³, which yields a linear increase
45 in FMR linewidth with increasing precessional frequency. In ferromagnetic metals, Gilbert
46 damping arises predominately from “intrinsic” mechanisms¹⁴⁻¹⁶ governed by the electronic
47 band structure¹⁷. Indeed, a recent experimental study by Khodadadi *et al.*¹⁸ has shown
48 that intrinsic, band-structure-based Gilbert damping dominates magnetic relaxation in high-
49 quality crystalline thin films of Fe, epitaxially grown on lattice-matched substrates. However,
50 it is yet unclear how intrinsic damping is impacted by the microstructure of polycrystalline
51 Fe films.

52 Microstructural disorder in polycrystalline Fe films can also introduce *extrinsic* magnetic
53 relaxation. A well-known extrinsic relaxation mechanism is two-magnon scattering, where
54 the uniform precession mode with zero wave vector scatters into a degenerate magnon mode
55 with a finite wave vector¹⁹⁻²². Two-magnon scattering generally leads to a nonlinear fre-
56 quency dependence of the FMR linewidth, governed by the nature of magnon scattering
57 centers at the surfaces^{23,24} or in the bulk of the film²⁵⁻²⁸. While some prior experiments
58 point to the prominent roles of extrinsic magnetic relaxation in polycrystalline ferromag-
59 netic films²⁹⁻³¹, systematic studies of extrinsic relaxation (e.g., two-magnon scattering) on

60 polycrystalline Fe thin films are still lacking.

61 Here, we investigate both the intrinsic and extrinsic contributions to magnetic relax-
62 ation at room temperature in polycrystalline Fe films. We have measured the frequency
63 dependence of the FMR linewidth with (1) the film magnetized out-of-plane (OOP), where
64 two-magnon scattering is suppressed²⁵ such that intrinsic Gilbert damping is quantified re-
65 liably, and (2) the film magnetized in-plane (IP), where two-magnon scattering is generally
66 expected to coexist with intrinsic Gilbert damping.

67 From OOP FMR results, we find that the intrinsic Gilbert damping of polycrystalline Fe
68 films at room temperature is independent of their structural properties and almost identical
69 to that of epitaxial films. Such insensitivity to microstructure is in contrast to disorder-
70 sensitive Gilbert damping recently shown in epitaxial Fe at *cryogenic* temperature¹⁸. Our
71 present work implies that, [in Fe thin films](#), Gilbert damping at a sufficiently high temperature
72 [is](#) primarily governed by the structure *within* nanoscale crystal grains, rather than grain
73 boundaries or interfacial disorder. This implication refutes the intuitive expectation that
74 intrinsic Gilbert damping should depend on structural disorder in polycrystalline films.

75 In IP FMR results, the frequency dependence of the FMR linewidth exhibits strong
76 nonlinear trends that vary significantly with film microstructure. To analyze the nonlin-
77 ear trends, we have employed the grain-to-grain two-magnon scattering model developed
78 by McMichael and Krivosik²⁵ with two types of correlation functions for capturing inho-
79 mogeneities in the film. However, neither of the correlation functions yields quantitative
80 agreement with the experimental results or physically consistent, reasonable parameters.
81 This finding implies that a physical, quantitative understanding of extrinsic magnetic relax-
82 ation requires further corrections of the existing two-magnon scattering model, along with
83 much more detailed characterization of the nanoscale inhomogeneities of the [Fe](#) film. Our
84 study stimulates opportunities for a deeper examination of fundamental magnetic relaxation
85 mechanisms in structurally disordered ferromagnetic metal films.

86 II. FILM DEPOSITION AND STRUCTURAL PROPERTIES

87 Polycrystalline Fe thin films were deposited using DC magnetron sputtering at room
88 temperature on Si substrates with a native oxide layer of SiO₂. The base pressure of the
89 chamber was below 1×10^{-7} Torr and all films were deposited with 3 mTorr Ar pressure. Two

90 sample series with different seed layers were prepared in our study: subs./Ti(3 nm)/Cu(3
91 nm)/Fe(2-25 nm)/Ti(3 nm) and subs./Ti(3 nm)/Ag(3 nm)/Fe(2-25 nm)/Ti(3 nm). In this
92 paper we refer to these two sample series as Cu/Fe and Ag/Fe, respectively. The layer
93 thicknesses are based on deposition rates derived from x-ray reflectivity (XRR) of thick
94 calibration films. The Ti layer grown directly on the substrate ensures good adhesion of
95 the film, whereas the Cu and Ag layers yield distinct microstructural properties for Fe
96 as described below. We note that Cu is often used as a seed layer for growing textured
97 polycrystalline ferromagnetic metal films^{32,33}. Our initial motivation for selecting Ag as an
98 alternative seed layer was that it might promote qualitatively different Fe film growth³⁴,
99 owing to a better match in bulk lattice parameter a between Fe ($a \approx 2.86 \text{ \AA}$) and Ag
100 ($a/\sqrt{2} \approx 2.88 \text{ \AA}$) compared to Fe and Cu ($a/\sqrt{2} \approx 2.55 \text{ \AA}$).

101 We performed x-ray diffraction (XRD) measurements to compare the structural properties
102 of the Cu/Fe and Ag/Fe films. Figure 1(a,b) shows symmetric θ - 2θ XRD scan curves
103 for several films from both the Cu/Fe and Ag/Fe sample series. For all Cu/Fe films, the
104 (110) body-center-cubic (bcc) peak can be observed around $2\theta = 44^\circ - 45^\circ$ (Fig. 1(a)). This
105 observation confirms that the Fe films grown on Cu are polycrystalline and textured, where
106 the crystal grains predominantly possess (110)-oriented planes that are parallel to the sample
107 surface. For Ag/Fe (Fig. 1(b)), the (110) bcc peak is absent or extremely weak, from
108 which one might surmise that the Fe films grown on Ag are amorphous or only possess
109 weak crystallographic texture. However, we find that the Ag/Fe films are, in fact, also
110 polycrystalline with evidence of (110) texturing. In the following, we elaborate on our XRD
111 results, first for Cu/Fe and then Ag/Fe.

112 We observe evidence for a peculiar, non-monotonic trend in the microstructural properties
113 of the Cu/Fe films. Specifically, the height of the θ - 2θ diffraction peak (Fig. 1(a)) increases
114 with Fe film thickness up to ≈ 10 nm but then decreases at higher Fe film thicknesses. While
115 we do not have a complete explanation for this peculiar nonmonotonic trend with film
116 thickness, a closer inspection of the XRD results (Fig. 1) provides useful insights. First, the
117 Fe film diffraction peak shifts toward a higher 2θ value with increasing film thickness. This
118 signifies that thinner Fe films on Cu are strained (with the Fe crystal lattice tetragonally
119 distorted), whereas thicker Fe films undergo structural relaxation such that the out-of-plane
120 lattice parameter converges toward the bulk value of $\approx 2.86 \text{ \AA}$, as summarized in Fig. 1(e).
121 Second, as the Fe film thickness approaches ≈ 10 nm, additional diffraction peaks appear to

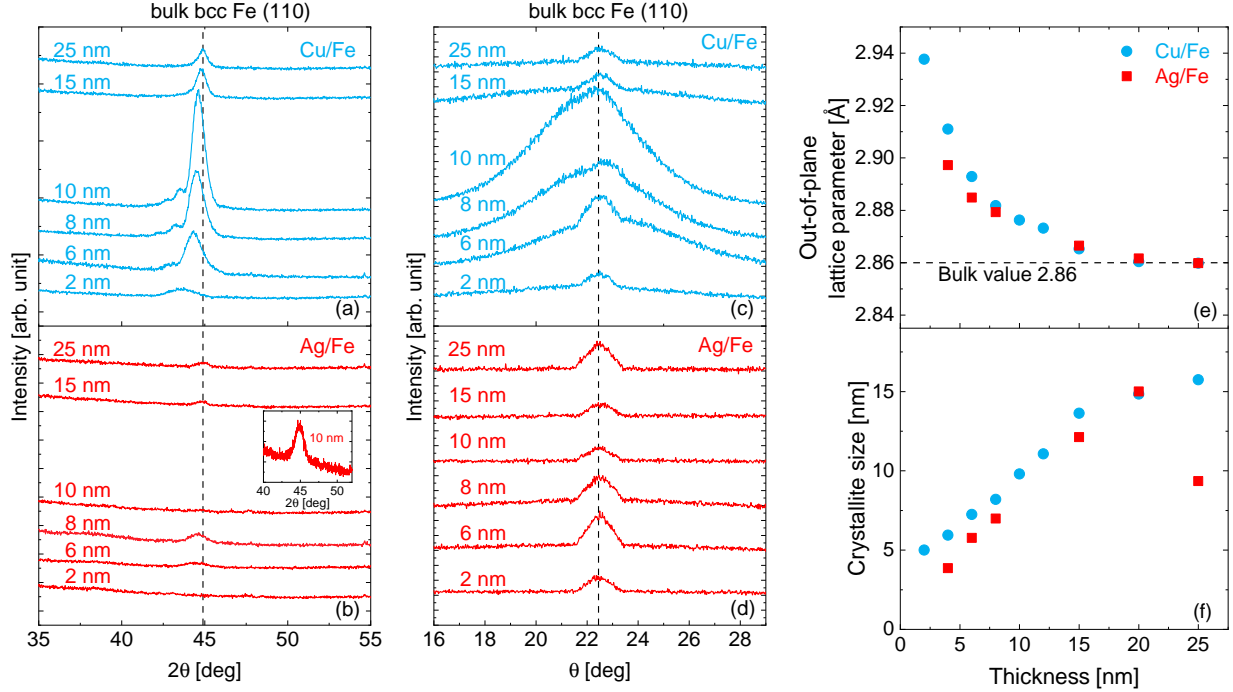


FIG. 1. (Color online) θ - 2θ X-ray diffraction scan curves for (a) Cu/Fe (blue lines) and (b) Ag/Fe (red lines) sample series. The inset in (b) is the grazing-incidence XRD scan curve for 10 nm thick Ag/Fe film. Rocking curves for (c) Cu/Fe (blue lines) and (d) Ag/Fe (red lines) sample series. (e) Out-of-plane lattice parameter estimated via Bragg's law using the 2θ value at the maximum of the tallest film diffraction peak. (f) Crystallite size estimated via the Scherrer equation using the full-width-at-half-maximum of the tallest film diffraction peak. In (e) and (f), the data for the Ag/Fe film series at a few thickness values are missing because of the absence of the bcc (110) peak in θ - 2θ XRD scans.

122 the left of the tall primary peak. We speculate that these additional peaks may originate
 123 from Fe crystals that remain relatively strained (i.e., with an out-of-plane lattice parameter
 124 larger than the bulk value), while the primary peak arises from more relaxed Fe crystals
 125 (i.e., with a lattice parameter closer to the bulk value). The coexistence of such different
 126 Fe crystals appears to be consistent with the rocking curve measurements (Fig. 1(c)), which
 127 exhibit a large broad background peak in addition to a small sharp peak for Cu/Fe films
 128 with thicknesses near ≈ 10 nm. As we describe in Sec. IV, these ≈ 10 nm thick Cu/Fe samples
 129 also show distinct behaviors in extrinsic damping (highly nonlinear frequency dependence of
 130 the FMR linewidth) and static magnetization reversal (enhanced coercivity), which appear

131 to be correlated with the peculiar microstructural properties evidenced by our XRD results.
132 On the other hand, it is worth noting that the estimated crystal grain size (Fig. 1(f)) –
133 derived from the width of the θ - 2θ diffraction peak – does not exhibit any anomaly near the
134 film thickness of ≈ 10 nm, but rather increases monotonically with film thickness.

135 Unlike the Cu/Fe films discussed above, the Ag/Fe films do not show a strong (110) bcc
136 peak in the θ - 2θ XRD results. However, the lack of pronounced peaks in the symmetric θ - 2θ
137 scans does not necessarily signify that Ag/Fe is amorphous. This is because symmetric θ - 2θ
138 XRD is sensitive to crystal planes that are nearly parallel to the sample surface, such that the
139 diffraction peaks capture only the crystal planes with out-of-plane orientation with a rather
140 small range of misalignment (within $\sim 1^\circ$, dictated by incident X-ray beam divergence). In
141 fact, from *asymmetric* grazing-incidence XRD scans that are sensitive to other planes, we
142 are able to observe a clear bcc Fe (110) diffraction peak even for Ag/Fe samples that lack
143 an obvious diffraction peak in θ - 2θ scans (see e.g. inset of Fig. 1(b)). Furthermore, rocking
144 curve scans (conducted with 2θ fixed to the expected position of the (110) Fe film diffraction
145 peak) provide orientation information over an angular range much wider than $\sim 1^\circ$. As shown
146 in Fig. 1(d), a clear rocking curve peak is observed for each Ag/Fe sample, suggesting that
147 Fe films grown on Ag are polycrystalline and (110)-textured – albeit with the (110) crystal
148 planes more misaligned from the sample surface compared to the Cu/Fe samples. The out-
149 of-plane lattice parameters of Ag/Fe films (with discernible θ - 2θ diffraction film peaks) show
150 the trend of relaxation towards the bulk value with increasing Fe thickness, similar to the
151 Cu/Fe series. Yet, the lattice parameters for Ag/Fe at small thicknesses are systematically
152 closer to the bulk value, possibly because Fe is less strained (i.e., better lattice matched)
153 on Ag than on Cu. We also find that the estimation of the crystal grain size for Ag/Fe –
154 although made difficult by the smallness of the diffraction peak – yields a trend comparable
155 to Cu/Fe, as shown in Fig. 1(f).

156 We also observe a notable difference between Cu/Fe and Ag/Fe in the properties of film
157 interfaces, as revealed by XRR scans in Fig. 2. The oscillation period depends inversely
158 on the film thickness. The faster decay of the oscillatory reflectivity signal at high angles
159 for the Ag/Fe films suggests that the Ag/Fe films may have rougher interfaces compared to
160 the Cu/Fe films. Another interpretation of the XRR results is that the Ag/Fe interface is
161 more diffuse than the Cu/Fe interface – i.e., due to interfacial intermixing of Ag and Fe. By
162 fitting the XRR results³⁵, we estimate an average roughness (or the thickness of the diffuse

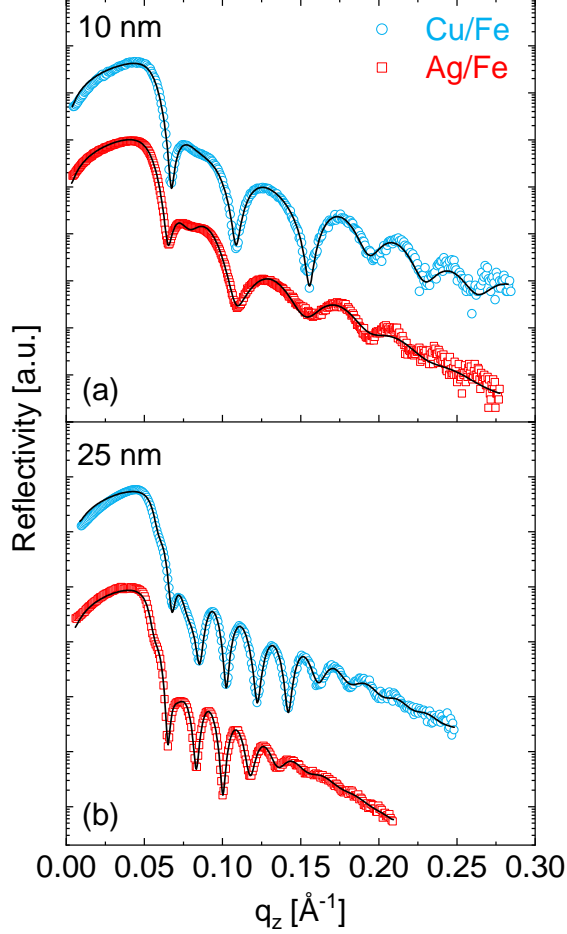


FIG. 2. (Color online) X-ray reflectivity scans of 10 nm and 25 nm thick films from (a) Cu/Fe (blue circles) and (b) Ag/Fe (red squares) sample series. Black solid curves are fits to the data.

163 interfacial layer) of $\lesssim 1$ nm for the Fe layer in Cu/Fe, while it is much greater at ≈ 2 -3 nm
 164 for Ag/Fe³⁶.

165 Our structural characterization described above thus reveals key attributes of the Cu/Fe
 166 and Ag/Fe sample series. Both film series are polycrystalline, exhibit (110) texture, and
 167 have grain sizes of order film thickness. Nevertheless, there are also crucial differences
 168 between Cu/Fe and Ag/Fe. The Cu/Fe series overall exhibits stronger θ - 2θ diffraction
 169 peaks than the Ag/Fe series, suggesting that the (110) bcc crystal planes of Fe grown on
 170 Cu are aligned within a tighter angular range than those grown on Ag. Moreover, Fe grown
 171 on Cu has relatively smooth or sharp interfaces compared to Fe grown on Ag. Although
 172 identifying the origin of such structural differences is beyond the scope of this work, Cu/Fe
 173 and Ag/Fe constitute two qualitatively distinct series of polycrystalline Fe films for exploring

174 the influence of microstructure on magnetic relaxation.

175 III. INTRINSIC GILBERT DAMPING PROBED BY OUT-OF-PLANE FMR

176 Having established the difference in structural properties between Cu/Fe and Ag/Fe, we
177 characterize room-temperature intrinsic damping for these samples with OOP FMR mea-
178 surements. The OOP geometry suppresses two-magnon scattering²⁵ such that the Gilbert
179 damping parameter can be quantified in a straightforward manner. We use a W-band
180 shorted waveguide in a superconducting magnet, which permits FMR measurements at high
181 fields ($\gtrsim 4$ T) that completely magnetize the Fe films out of plane. The details of the mea-
182 surement method are found in Refs.^{18,37}. Figure 3(a) shows the frequency dependence of
183 half-width-at-half-maximum (HWHM) linewidth ΔH_{OOP} for selected thicknesses from both
184 sample series. The linewidth data of 25 nm thick epitaxial Fe film from a previous study¹⁸
185 is plotted in Fig. 3(a) as well. The intrinsic damping parameter can be extracted from the
186 linewidth plot using

$$\Delta H_{\text{OOP}} = \Delta H_0 + \frac{2\pi}{\gamma} \alpha_{\text{OOP}} f, \quad (1)$$

187 where ΔH_0 is the inhomogeneous broadening³⁸, $\gamma = \frac{g\mu_B}{\hbar}$ is the gyromagnetic ratio ($\gamma/2\pi \approx$
188 2.9 MHz/Oe [Ref.³⁹], obtained from the frequency dependence of the resonance field³⁷), and
189 α_{OOP} is the measured viscous damping parameter. In general, α_{OOP} can include not only
190 intrinsic Gilbert damping, parameterized by α_{int} , but also eddy-current, radiative damping,
191 and spin pumping contributions⁴⁰, which all yield a linear frequency dependence of the
192 linewidth. Damping due to eddy current is estimated to make up less than 10% of the total
193 measured damping parameter³⁷ and is ignored here. Since we used a shorted waveguide in
194 our setup, the radiative damping does not apply here. Spin pumping is also negligible for
195 most of the samples here because the materials in the seed and capping layers (i.e., Ti, Cu,
196 and Ag) possess weak spin-orbit coupling and are hence poor spin sinks^{31,41,42}. We therefore
197 proceed by assuming that the measured OOP damping parameter α_{OOP} is equivalent to the
198 *intrinsic* Gilbert damping parameter.

199 The extracted damping parameter is plotted as a function of Fe film thickness in Fig.
200 3(b). The room-temperature damping parameters of all Fe films with thicknesses of 4-25
201 nm fall in the range of 0.0024 ± 0.0004 , which is shaded in red in Fig. 3(b). This damping

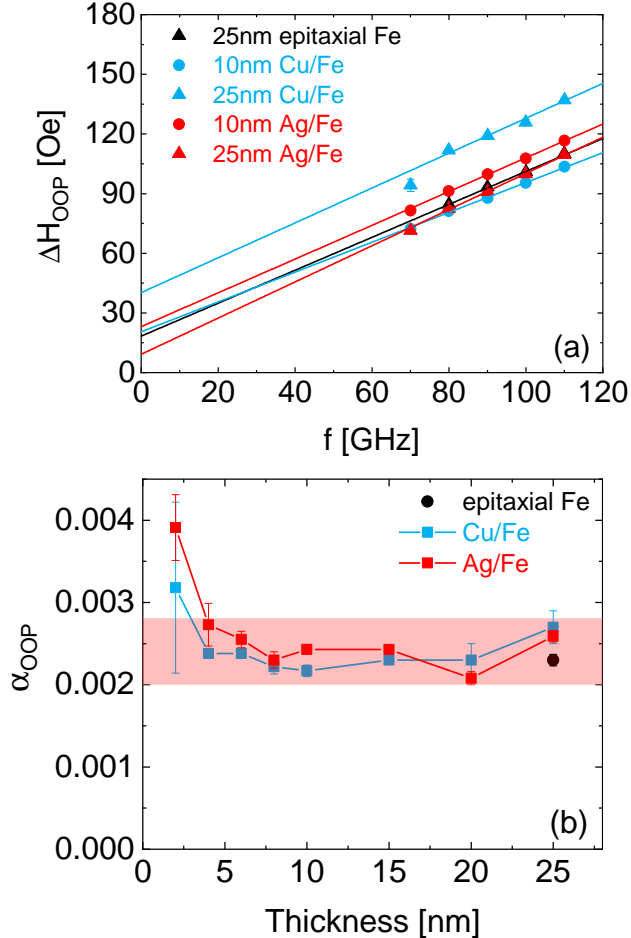


FIG. 3. (Color online) (a) OOP FMR half-width-at-half-maximum linewidth ΔH_{OOP} as a function of resonance frequency f . Lines correspond to fits to the data. (b) Gilbert damping parameter α_{OOP} extracted from OOP FMR as a function of film thickness. The red shaded area highlights the damping value range that contains data points of all films thicker than 4 nm. The data for the epitaxial Fe sample (25 nm thick Fe grown on MgAl_2O_4) are adapted from Ref.¹⁸.

parameter range is quantitatively in line with the value reported for epitaxial Fe (black
 202 symbol in Fig. 3(b))¹⁸. For 2 nm thick samples, the damping parameter is larger likely
 203 due to an additional interfacial contribution^{43–45} – e.g., spin relaxation through interfacial
 204 Rashba spin-orbit coupling⁴⁶ that becomes evident only for ultrathin Fe. The results in
 205 Fig. 3(b) therefore indicate that the structural properties of the $\gtrsim 4$ nm thick polycrystalline
 206 bcc Fe films have little influence on their intrinsic damping.
 207

It is remarkable that these polycrystalline Cu/Fe and Ag/Fe films – with different thick-
 208 nesses and microstructural properties (as revealed in Sec. II) – exhibit essentially the same
 209

210 *room-temperature* intrinsic Gilbert damping parameter as single-crystalline bcc Fe. This
211 finding is qualitatively distinct from a prior report¹⁸ on intrinsic Gilbert damping in single-
212 crystalline Fe films at *cryogenic* temperature, which is sensitive to microstructural disorder.
213 In the following, we discuss the possible differences in the mechanisms of intrinsic damping
214 between these temperature regimes.

215 Intrinsic Gilbert damping in ferromagnetic metals is predominantly governed by transi-
216 tions of spin-polarized electrons between electronic states, within a given electronic band
217 (intraband scattering) or in different electronic bands (interband scattering) near the Fermi
218 level¹⁵. For Fe, previous studies^{15,18,47} indicate that intraband scattering tends to dominate
219 at low temperature where the electronic scattering rate is low (e.g., $\sim 10^{13} \text{ s}^{-1}$); by contrast,
220 interband scattering likely dominates at room temperature where the electronic scattering
221 rate is higher (e.g., $\sim 10^{14} \text{ s}^{-1}$). According to our results (Fig. 3(b)), intrinsic damping at
222 room temperature is evidently unaffected by the variation in the structural properties of the
223 Fe films. Hence, the observed intrinsic damping is mostly governed by the electronic band
224 structure *within the Fe grains*, such that disorder in grain boundaries or film interfaces has
225 minimal impact.

226 The question remains as to why interband scattering at room temperature leads to Gilbert
227 damping that is insensitive to microstructural disorder, in contrast to intraband scattering
228 at low temperature yielding damping that is quite sensitive to microstructure¹⁸. This dis-
229 tinction may be governed by what predominantly drives electronic scattering – specifically,
230 defects (e.g., grain boundaries, rough or diffuse interfaces) at low temperature, as opposed
231 to phonons at high temperature. That is, the dominance of phonon-driven scattering at
232 room temperature may effectively diminish the roles of microstructural defects in Gilbert
233 damping. Future experimental studies of temperature-dependent damping in polycrystalline
234 Fe films may provide deeper insights. Regardless of the underlying mechanisms, the robust
235 consistency of α_{OOP} (Fig. 3(b)) could be an indication that the intrinsic Gilbert damping
236 parameter at a sufficiently high temperature is a **nanoscale** property of the **Fe thin film**,
237 possibly averaged over the ferromagnetic exchange length of just a few nm (Ref.⁴⁸) that is
238 comparable or smaller than the grain size. In this scenario, the impact on damping from
239 grain boundaries would be limited in comparison to the contributions to damping within
240 the grains.

241 Moreover, the misalignment of Fe grains evidently does not have much influence on the

242 intrinsic damping. This is reasonable considering that intrinsic Gilbert damping is predicted
 243 to be nearly isotropic in Fe at sufficiently high electronic scattering rates⁴⁹ – e.g., $\sim 10^{14}$ s⁻¹
 244 at room temperature where interband scattering is expected to be dominant^{15,18,47}. It is
 245 also worth emphasizing that α_{OOP} remains unchanged for Fe films of various thicknesses
 246 with different magnitudes of strain (tetragonal distortion, as evidenced by the variation in
 247 the out-of-plane lattice parameter in Fig. 1(e)). Strain in Fe grains is not expected to impact
 248 the intrinsic damping, as Ref.¹⁸ suggests that strain in bcc Fe does not significantly alter
 249 the band structure near the Fermi level. Thus, polycrystalline Fe films exhibit essentially
 250 the same magnitude of room-temperature intrinsic Gilbert damping as epitaxial Fe, as long
 251 as the grains retain the bcc crystal structure.

252 The observed invariance of intrinsic damping here is quite different from the recent study
 253 of polycrystalline Co₂₅Fe₇₅ alloy films³¹, reporting a decrease in intrinsic damping with
 254 increasing structural disorder. This inverse correlation between intrinsic damping and dis-
 255 order in Ref.³¹ is attributed to the dominance of intraband scattering, which is inversely
 256 proportional to the electronic scattering rate. It remains an open challenge to understand
 257 why the room-temperature intrinsic Gilbert damping of some ferromagnetic metals might
 258 be more sensitive to structural disorder than others. [Different electronic band structures of](#)
 259 [diverse ferromagnetic metals could strongly influence whether defects or phonons dominate](#)
 260 [electronic scattering, which underpins Gilbert damping, at a given temperature. Further](#)
 261 [experiments on additional ferromagnetic metals beyond elemental Fe could reveal a more](#)
 262 [general relationship between microstructural properties and intrinsic Gilbert damping.](#)

263 **IV. EXTRINSIC MAGNETIC RELAXATION PROBED BY IN-PLANE FMR**

264 Although we have shown via OOP FMR in Sec. III that intrinsic Gilbert damping is
 265 essentially independent of the structural properties of the Fe films, it might be expected
 266 that microstructure has a pronounced impact on *extrinsic* magnetic relaxation driven by
 267 two-magnon scattering, which is generally present in IP FMR. IP magnetized films are more
 268 common in device applications than OOP magnetized films, since the shape anisotropy of
 269 thin films tends to keep the magnetization in the film plane. What governs the performance
 270 of such magnetic devices (e.g., quality factor^{50,51}) may not be the intrinsic Gilbert damping
 271 parameter but the total FMR linewidth. Thus, for many magnetic device applications, it is

272 essential to understand the contributions to the IP FMR linewidth.

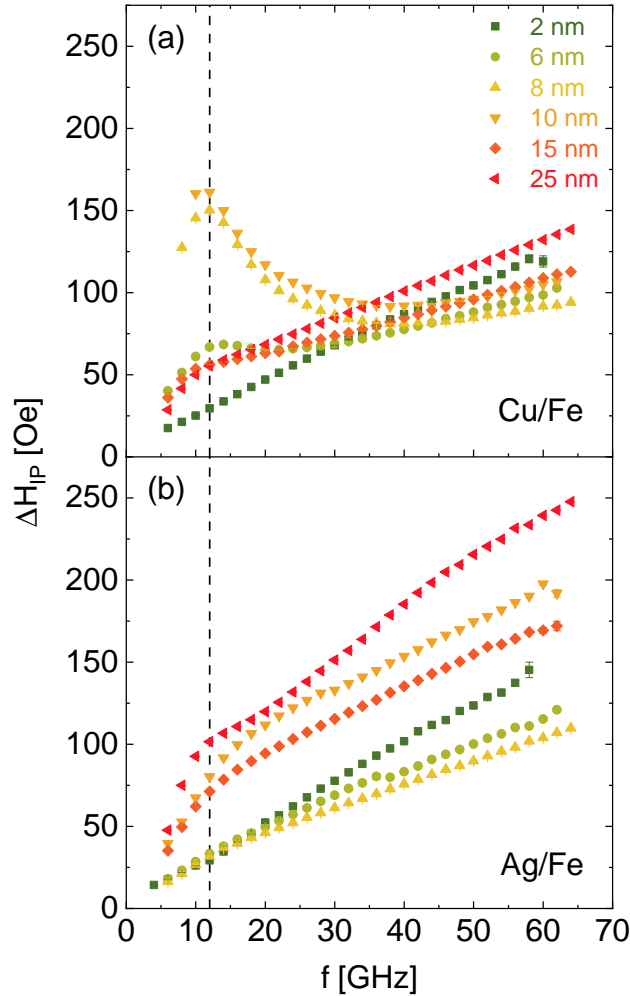


FIG. 4. (Color online) IP FMR half-width-at-half-maximum linewidth ΔH_{IP} as a function of resonance frequency f for (a) Cu/Fe and (b) Ag/Fe. The vertical dashed line at 12 GHz highlights the hump in linewidth vs frequency seen for many of the samples.

273 IP FMR measurements have been performed using a coplanar-waveguide-based spectrom-
 274 eter, as detailed in Refs.^{18,37}. Examples of the frequency dependence of IP FMR linewidth
 275 are shown in Fig. 4. In contrast to the linear frequency dependence that arises from in-
 276 trinsic Gilbert damping in Fig. 3(a), a nonlinear hump is observed for most of the films
 277 in the vicinity of ≈ 12 GHz. In some films, e.g., 10 nm thick Cu/Fe film, the hump is so
 278 large that its peak even exceeds the linewidth at the highest measured frequency. Similar
 279 nonlinear IP FMR linewidth behavior has been observed in Fe alloy films⁵² and epitaxial
 280 Heusler films⁵³ in previous studies, where two-magnon scattering has been identified as a

281 significant contributor to the FMR linewidth. Therefore, in the following, we attribute the
 282 nonlinear behavior to two-magnon scattering.

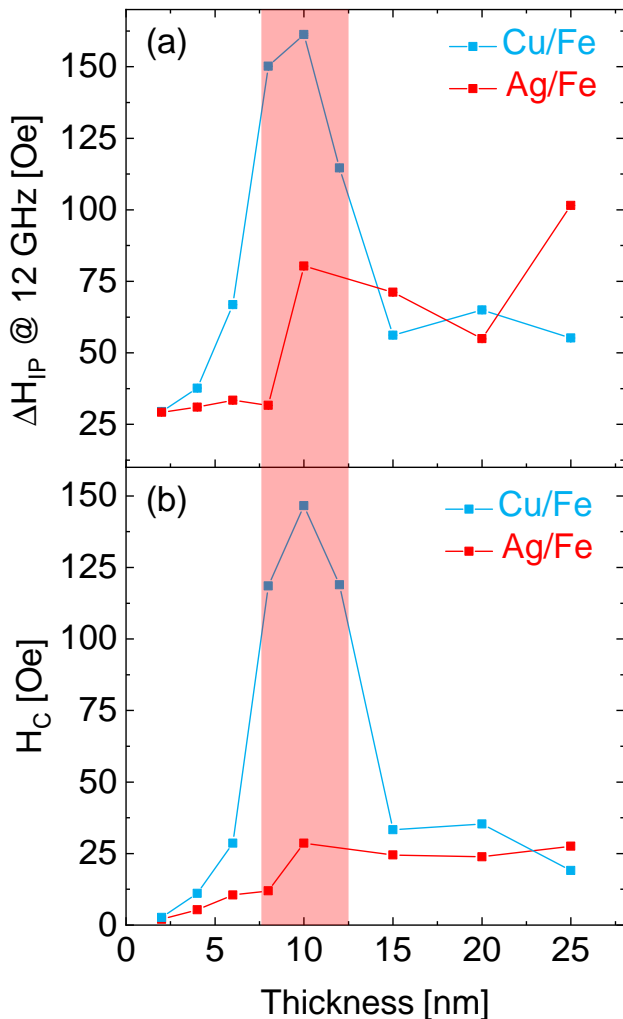


FIG. 5. (Color online) (a) IP FMR half-width-at-half-maximum linewidth at 12 GHz – approximately where the maximum (“hump”) in linewidth vs frequency is seen (see Fig. 4) – as a function of film thickness for both Cu/Fe and Ag/Fe. (b) Coercivity H_c as a function of film thickness for both Cu/Fe and Ag/Fe. The red shaded area highlights thickness region where the Cu/Fe sample series show a peak behavior in both plots.

283 To gain insight into the origin of two-magnon scattering, we plot the linewidth at 12
 284 GHz – approximately where the hump is seen in Fig. 4 – against the Fe film thickness in
 285 Fig. 5(a). We do not observe a monotonic decay in the linewidth with increasing thickness
 286 that would result from two-magnon scattering of interfacial origin⁵⁴. Rather, we observe
 287 a non-monotonic thickness dependence in Fig. 5(a), which indicates that the observed

288 two-magnon scattering originates within the bulk of the films. We note that Ag/Fe with
 289 greater interfacial disorder (see Sec. II) exhibits weaker two-magnon scattering than Cu/Fe,
 290 particularly in the lower thickness regime ($\lesssim 10$ nm). This observation further corroborates
 291 that the two-magnon scattering here is not governed by the interfacial roughness of Fe
 292 films. The contrast between Cu/Fe and Ag/Fe also might appear counterintuitive, since
 293 two-magnon scattering is induced by defects and hence might be expected to be stronger
 294 for more “defective” films (i.e., Ag/Fe in this case). The counterintuitive nature of the
 295 two-magnon scattering here points to more subtle mechanisms at work.

296 To search for a possible correlation between static magnetic properties and two-magnon
 297 scattering, we have performed vibrating sample magnetometry (VSM) measurements with a
 298 Microsense EZ9 VSM. Coercivity extracted from VSM measurements is plotted as a function
 299 of film thickness in Fig. 5(b), which shows a remarkably close correspondence with linewidth
 300 vs thickness (Fig. 5(a)). In particular, a pronounced peak in coercivity is observed for Cu/Fe
 301 around 10 nm, corresponding to the same thickness regime where the 12 GHz FMR linewidth
 302 for Cu/Fe is maximized. Moreover, the 10 nm Cu/Fe sample (see Sec. II) exhibits a tall,
 303 narrow bcc (110) diffraction peak, which suggests that its peculiar microstructure plays a
 304 possible role in the large two-magnon scattering and coercivity (e.g., via stronger domain
 305 wall pinning).

306 While the trends shown in Fig. 5 provide some qualitative insights, we now attempt to
 307 quantitatively analyze the frequency dependence of FMR linewidth for the Cu/Fe and Ag/Fe
 308 films. We assume that the Gilbert damping parameter for IP FMR is equal to that for OOP
 309 FMR, i.e., $\alpha_{\text{IP}} = \alpha_{\text{OOP}}$. This assumption is physically reasonable, considering that Gilbert
 310 damping is theoretically expected to be isotropic in Fe films near room temperature⁴⁹. While
 311 a recent study has reported anisotropic Gilbert damping that scales quadratically with
 312 magnetostriction⁵⁵, this effect is likely negligible in elemental Fe whose magnetostriction is
 313 several times smaller^{56,57} than that of the $\text{Fe}_{0.7}\text{Ga}_{0.3}$ alloy in Ref.⁵⁵.

314 Thus, from the measured IP linewidth ΔH_{IP} , the extrinsic two-magnon scattering
 315 linewidth ΔH_{TMS} can be obtained by

$$\Delta H_{\text{TMS}} = \Delta H_{\text{IP}} - \frac{2\pi}{\gamma} \alpha_{\text{IP}}, \quad (2)$$

316 where $\frac{2\pi}{\gamma} \alpha_{\text{IP}}$ is the Gilbert damping contribution. Figure 6 shows the obtained ΔH_{TMS} and fit
 317 attempts using the “grain-to-grain” two-magnon scattering model developed by McMichael

318 and Krivosik²⁵. This model captures the inhomogeneity of the effective internal magnetic
 319 field in a film consisting of many magnetic grains. The magnetic inhomogeneity can arise
 320 from the distribution of magnetocrystalline anisotropy field directions associated with the
 321 randomly oriented crystal grains⁵². In this model the two-magnon scattering linewidth
 322 ΔH_{TMS} is a function of the Gilbert damping parameter α_{IP} , the effective anisotropy field
 323 H_a of the randomly oriented grain, and the correlation length ξ within which the effective
 324 internal magnetic field is correlated. Further details for computing ΔH_{TMS} are provided in
 325 the Appendix and Refs.^{25,52,53}. As we have specified above, α_{IP} is set to the value derived
 326 from OOP FMR results (i.e., α_{OOP} in Fig. 3(b)). This leaves ξ and H_a as the only free
 327 parameters in the fitting process.

328 The modeling results are dependent on the choice of the correlation function $C(\mathbf{R})$, which
 329 captures how the effective internal magnetic field is correlated as a function of lateral distance
 330 \mathbf{R} in the film plane. We first show results obtained with a simple exponentially decaying
 331 correlation function, as done in prior studies of two-magnon scattering^{25,52,53}, i.e.,

$$C(\mathbf{R}) = \exp\left(-\frac{|\mathbf{R}|}{\xi}\right). \quad (3)$$

332 Equation 3 has the same form as the simplest correlation function used to model rough
 333 topographical surfaces (when they are assumed to be “self-affine”)⁵⁸. Fit results with Eq. (3)
 334 are shown in dashed blue curves in Fig. 6. For most samples, the fitted curve does not
 335 reproduce the experimental data quantitatively. Moreover, the fitted values of ξ and H_a
 336 often reach physically unrealistic values, e.g., with $H_a > 10^4$ Oe and $\xi < 1$ nm (see Table I).
 337 These results suggest that the model does not properly capture the underlying physics of
 338 two-magnon scattering in our samples.

339 A possible cause for the failure to fit the data is that the simple correlation function
 340 (Eq. 3) is inadequate. We therefore consider an alternative correlation function by again
 341 invoking an analogy between the spatially varying height of a rough surface⁵⁸ and the spa-
 342 tially varying effective internal magnetic field in a film. Specifically, we apply a correlation
 343 function (i.e., a special case of Eq. (4.3) in Ref.⁵⁸ where short-range roughness $\alpha = 1$) for
 344 the so-called “mounded surface,” which incorporates the average distance λ between peaks
 345 in topographical height (or, analogously, effective internal magnetic field):

$$C(\mathbf{R}) = \frac{\sqrt{2}|\mathbf{R}|}{\xi} K_1\left(\frac{\sqrt{2}|\mathbf{R}|}{\xi}\right) J_0\left(\frac{2\pi|\mathbf{R}|}{\lambda}\right), \quad (4)$$

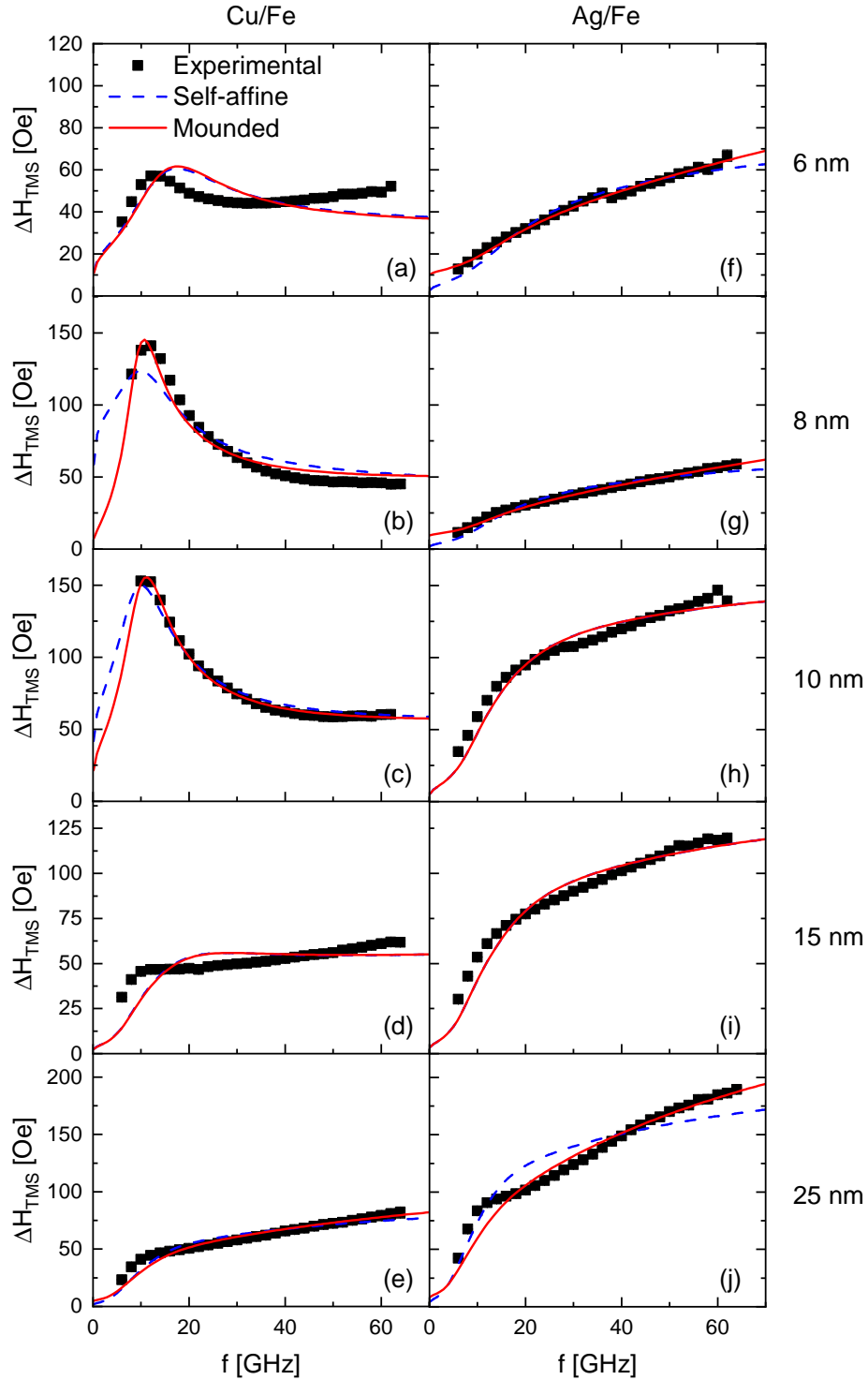


FIG. 6. (Color online) Extrinsic two-magnon scattering linewidth ΔH_{TMS} vs frequency f and fitted curves for 6, 8, 10, 15, and 25 nm Cu/Fe and Ag/Fe films. Black squares represent experimental FMR linewidth data. Dashed blue and solid red curves represent the fitted curves using correlation functions proposed for modeling self-affine and mounded surfaces, respectively. In (d), (e), (h), (i), dashed blue curves overlap with solid red curves.

346 where J_0 and K_1 are the Bessel function of the first kind of order zero and the modified Bessel
347 function of the second kind of order one, respectively. This oscillatory decaying function is
348 chosen because its Fourier transform (see Appendix) does not contain any transcendental
349 functions, which simplifies the numerical calculations. We also stress that while Eq. (4) in
350 the original context (Ref.⁵⁸) was used to model topographical roughness, we are applying
351 Eq. (4) in an attempt to model the spatial fluctuations (“roughness”) of the effective internal
352 magnetic field – rather than the roughness of the film topography.

353 The fitted curves using the model with Eq. (4) are shown in solid red curves in Fig. 6. Fit
354 results for some samples show visible improvement, although this is perhaps not surprising
355 with the introduction of λ as an additional free parameter. Nevertheless, the fitted values
356 of H_a or λ still diverge to unrealistic values of $> 10^4$ Oe or $> 10^4$ nm in some cases (see
357 Table I), which means that the new correlation function (Eq. (4)) does not fully reflect
358 the meaningful underlying physics of our samples either. More detailed characterization of
359 the microstructure and inhomogeneities, e.g., via synchrotron x-ray and neutron scattering,
360 could help determine the appropriate correlation function. It is also worth pointing out that
361 for some samples (e.g. 15 nm Cu/Fe and Ag/Fe films), essentially identical fit curves are
362 obtained regardless of the correlation function. This is because when $\lambda \gg \xi$, the Fourier
363 transform of Eq. (4) has a very similar form as the Fourier transform of Eq. (3), as shown in
364 the Appendix. In such cases, the choice of the correlation function has almost no influence
365 on the behavior of the two-magnon scattering model in the fitting process.

366 V. SUMMARY

367 We have examined room-temperature intrinsic and extrinsic damping in two series of
368 polycrystalline Fe thin films with distinct structural properties. Out-of-plane FMR mea-
369 surements confirm constant intrinsic Gilbert damping of ≈ 0.0024 , essentially independent
370 of film thickness and structural properties. **We deduce** that intrinsic damping in Fe at room
371 temperature is predominantly governed by the crystalline and electronic band structures
372 within the grains, rather than scattering at grain boundaries or film surfaces. **This presents**
373 **a distinct counterexample to the intuition that scattering by defects should impact Gilbert**
374 **damping.**

375 The results from in-plane FMR, where extrinsic damping (i.e., two-magnon scattering)

TABLE I. Summary of IP FMR linewidth fit results. Note the divergence to physically unreasonable values in many of the results. Standard error is calculated using equation $\sqrt{\text{SSR}/\text{DOF} \times \text{diag}(\mathbf{COV})}$, where SSR stands for the sum of squared residuals, DOF stands for degrees of freedom, and \mathbf{COV} stands for the covariance matrix.

Sample Series	Thickness (nm)	Self-affine		Mounded		
		ξ (nm)	H_a (Oe)	ξ (nm)	H_a (Oe)	λ (nm)
Cu/Fe	6	70 ± 10	170 ± 10	80 ± 90	24 ± 3	$>1 \times 10^4$
	8	200 ± 100	150 ± 20	700 ± 1000	25 ± 2	900 ± 100
	10	140 ± 40	200 ± 20	160 ± 50	33 ± 1	800 ± 200
	15	9 ± 2	800 ± 100	10 ± 20	100 ± 80	$>1 \times 10^4$
	25	0 ± 5	$>1 \times 10^4$	60 ± 30	$>1 \times 10^4$	10.41 ± 0.01
Ag/Fe	6	0 ± 40	$>1 \times 10^4$	150 ± 40	$>1 \times 10^4$	11.7 ± 0.7
	8	0 ± 30	$>1 \times 10^4$	170 ± 50	$>1 \times 10^4$	12 ± 4
	10	6 ± 1	1500 ± 300	8 ± 40	200 ± 500	$>1 \times 10^4$
	15	2 ± 2	4000 ± 3000	3 ± 9	500 ± 900	$>6 \times 10^3$
	25	0 ± 6	$>1 \times 10^4$	140 ± 50	$>1 \times 10^4$	15 ± 6

376 plays a significant role, are far more nuanced. The conventional grain-to-grain two-magnon
377 scattering model fails to reproduce the in-plane FMR linewidth data with physically rea-
378 sonable parameters – pointing to the need to modify the model, along with more detailed
379 characterization of the film microstructure. Our experimental findings advance the under-
380 standing of intrinsic Gilbert damping in polycrystalline Fe, while motivating further studies
381 to uncover the mechanisms of extrinsic damping in structurally disordered thin films.

382 ACKNOWLEDGMENTS

383 S.W. acknowledges support by the ICTAS Junior Faculty Program. D.A.S. and S.E.
384 acknowledge support by the National Science Foundation, Grant No. DMR-2003914. P.
385 N. would like to acknowledge support through NASA Grant NASA CAN80NSSC18M0023.
386 A. R. would like to acknowledge support through the Defense Advanced Research Project
387 Agency (DARPA) program on Topological Excitations in Electronics (TEE) under Grant

388 No. D18AP00011. This work was supported by NanoEarth, a member of National Nan-
 389 otechnology Coordinated Infrastructure (NNCI), supported by NSF (ECCS 1542100).

390 **Appendix A: Details of the Two-Magnon Scattering Model**

391 In the model developed by McMichael and Krivosik, the two-magnon scattering contri-
 392 bution ΔH_{TMS} to the FMR linewidth is given by^{25,52,53}

$$\Delta H_{\text{TMS}} = \frac{\gamma^2 H_a^2}{2\pi P_A(\omega)} \int \Lambda_{0k} C_k(\xi) \delta_\alpha(\omega - \omega_k) d^2k \quad (\text{A1})$$

393 where ξ is correlation length, H_a is the effective anisotropy field of the randomly oriented
 394 grain. $P_A(\omega) = \frac{\partial \omega}{\partial H} \Big|_{H=H_{\text{FMR}}} = \sqrt{1 + \left(\frac{4\pi M_s}{2\omega/\gamma}\right)^2}$ accounts for the conversion between the frequency
 395 and field swept linewidth. Λ_{0k} represents the averaging of the anisotropy axis fluctuations
 396 over the sample. It also takes into account the ellipticity of the precession for both the
 397 uniform FMR mode and the spin wave mode⁵². The detailed expression of Λ_{0k} can be found
 398 in the Appendix of Ref.⁵². The coefficients in the expression of Λ_{0k} depend on the type of
 399 anisotropy of the system. Here, we used first-order cubic anisotropy for bcc Fe. $\delta_\alpha(\omega - \omega_k)$
 400 selects all the degenerate modes, where ω represents the FMR mode frequency and ω_k
 401 represents the spin wave mode frequency. The detailed expression of ω_k can be found in
 402 Ref.²⁵. In the ideal case where Gilbert damping is 0, δ_α is the Dirac delta function. For a
 403 finite damping, $\delta_\alpha(\omega_0 - \omega_k)$ is replaced by a Lorentzian function $\frac{1}{\pi} \frac{(\alpha_{\text{IP}} \omega_k / \gamma) \partial \omega / \partial H}{(\omega_k - \omega)^2 + [(\alpha_{\text{IP}} \omega_k / \gamma) \partial \omega / \partial H]^2}$,
 404 which is centered at ω and has the width of $(2\alpha_{\text{IP}} \omega_k / \gamma) \partial \omega / \partial H$.

405 Finally, $C_k(\xi)$ (or $C_k(\xi, \lambda)$) is the Fourier transform of the grain-to-grain internal field
 406 correlation function, Eq. (3) (or Eq. (4)). For the description of magnetic inhomogeneity
 407 analogous to the simple self-affine topographical surface⁵⁸, the Fourier transform of the
 408 correlation function, Eq. (3), is

$$C_k(\xi) = \frac{2\pi\xi^2}{[1 + (k\xi)^2]^{\frac{3}{2}}}, \quad (\text{A2})$$

409 as also used in Refs.^{25,52,53}. For the description analogous to the mounded surface, the
 410 Fourier transform of the correlation function, Eq. (4), is⁵⁸

$$C_k(\xi, \lambda) = \frac{8\pi^3 \xi^2 \left(1 + \frac{2\pi^2 \xi^2}{\lambda^2} + \frac{\xi^2}{2} k^2\right)}{\left[\left(1 + \frac{2\pi^2 \xi^2}{\lambda^2} + \frac{\xi^2}{2} k^2\right)^2 - \left(\frac{2\pi \xi^2}{\lambda} k\right)^2\right]^{3/2}}. \quad (\text{A3})$$

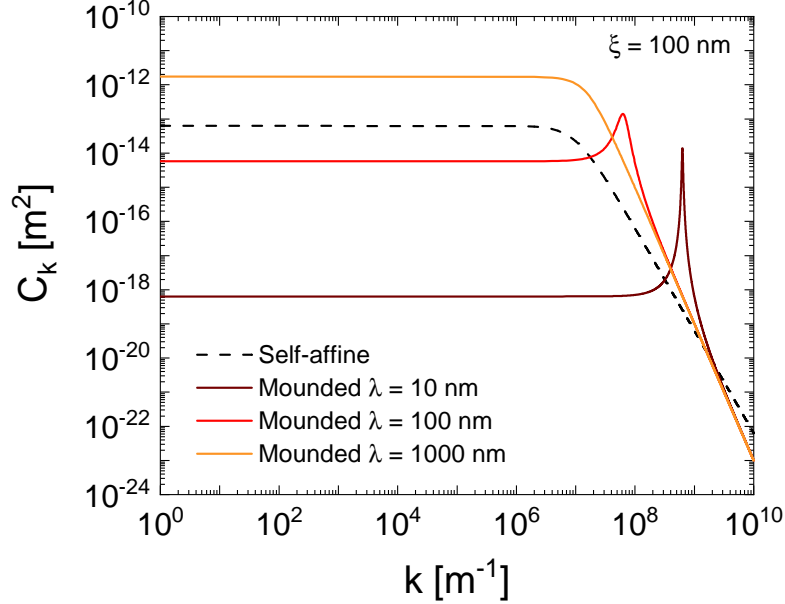


FIG. 7. Fourier transform of correlation function for mounded surfaces as a function of wavenumber k for three different λ values. Fourier transform of correlation function for self-affine surfaces as a function of k is also included for comparison purpose. ξ is set as 100 nm for all curves.

411 When $\lambda \gg \xi$, Eq. (A3) becomes

$$C_k(\xi) \approx \frac{8\pi^3 \xi^2}{\left(1 + \frac{\xi^2}{2} k^2\right)^2}, \quad (\text{A4})$$

412 which has a similar form as Eq. (A2). This similarity can also be demonstrated graphically.
 413 Figure 7 plots a self-affine C_k curve (Eq. (A2)) at $\xi = 100$ nm and three mounded C_k curves
 414 (Eq. (A3)) at $\lambda = 10, 100, 1000$ nm. ξ in mounded C_k curves is set as 100 nm as well. It
 415 is clearly shown in Fig. 7 that when $\lambda = 1000$ nm, the peak appearing in $\lambda = 10$ and 100
 416 nm mounded C_k curves disappears and the curve shape of mounded C_k resembles that of
 417 self-affine C_k .

418 The hump feature in Fig. 4 is governed by both δ_α and C_k (see Eq. A1). δ_α has the shape
 419 of ∞ in reciprocal space (k space), as shown in our videos in the Supplemental Material⁵⁹ as
 420 well as Fig. 5(b) of Ref.⁵³ and Fig 2 (b) of Ref.²⁵. The size of the contour of the degenerated
 421 spin wave modes in k space increases as the microwave frequency f increases, which means
 422 the number of available degenerate spin wave modes increases as f increases. As shown
 423 in Fig. 7, self-affine C_k is nearly constant with the wavenumber k until k reaches $\sim 1/\xi$.

424 This suggests that the system becomes effectively more uniform (i.e. weaker inhomogeneous
 425 perturbation) when the length scale falls below the characteristic correlation length ξ (i.e.,
 426 $k > 1/\xi$). Because inhomogeneities serve as the scattering centers of two-magnon scattering
 427 process, degenerate spin wave modes with $k > 1/\xi$ are less likely to be scattered into.

428 Now we consider the f dependence of the two-magnon scattering rate. When f is small,
 429 the two-magnon scattering rate increases as f increases because more degenerate spin wave
 430 modes become available as f increases. When f further increases, the wavenumber k of
 431 some degenerate spin wave modes exceeds $1/\xi$. This will decrease the overall two-magnon
 432 scattering rate because the degenerate spin wave modes with $k > 1/\xi$ are less likely to be
 433 scattered into, as discussed above. Furthermore, the portion of degenerate spin wave modes
 434 with $k > 1/\xi$ increases as f continues to increase. When the impact of decreasing two-
 435 magnon scattering rate for degenerate spin wave modes with high k surpasses the impact
 436 of increasing available degenerate spin wave modes, the overall two-magnon scattering rate
 437 will start to decrease as f increases. Consequently, the nonlinear trend – i.e., a “hump” –
 438 in FMR linewidth ΔH_{TMS} vs f appears in Fig. 4.

439 However, the scenario discussed above can only happen when ξ is large enough, because
 440 the wavenumber k of degenerate spin wave modes saturates (i.e., reaches a limit) as f
 441 approaches infinity. If the limit value of k is smaller than $1/\xi$, the two-magnon scattering
 442 rate will increase monotonically as f increases. In that case the hump feature will not
 443 appear. See our videos in the Supplemental Material⁵⁹ that display the f dependence of
 444 Λ_{0k} , $\delta_\alpha(\omega - \omega_k)$, $\frac{C_k(\xi)}{2\pi\xi^2}$, $\frac{\Lambda_{0k}C_k(\xi)\delta_\alpha(\omega - \omega_k)}{2\pi\xi^2}$, and ΔH_{TMS} for various ξ values.

445 Previous discussions of the hump feature are all based on the self-affine correlation func-
 446 tion (Eq. 3). The main difference between the mounded correlation function (Eq. 4) and the
 447 self-affine correlation function (Eq. 3) is that the mounded correlation function has a peak
 448 when λ is not much larger than ξ as shown in Fig. 7. This means when the wavenumber
 449 k of degenerate spin wave modes enters (leaves) the peak region, two-magnon scattering
 450 rate will increase (decrease) much faster compared to the self-affine correlation function. In
 451 other words, the mounded correlation function can generate a narrower hump compared to
 452 the self-affine correlation function in the two-magnon linewidth ΔH_{TMS} vs f plot, which is

453 shown in Fig. 6 (b, c).

-
- 454 ¹ Z. Diao, Z. Li, S. Wang, Y. Ding, A. Panchula, E. Chen, L.-C. Wang, and Y. Huai, *J. Phys.*
455 *Condens. Matter* **19**, 165209 (2007).
- 456 ² X. Zhu and J.-G. Zhu, *IEEE Transactions on Magnetics* **43**, 2349 (2007).
- 457 ³ H. Yu, O. d'Allivy Kelly, V. Cros, R. Bernard, P. Bortolotti, A. Anane, F. Brandl, R. Huber,
458 I. Stasinopoulos, and D. Grundler, *Sci. Rep.* **4** (2014), 10.1038/srep06848.
- 459 ⁴ G. E. Rowlands, C. A. Ryan, L. Ye, L. Rehm, D. Pinna, A. D. Kent, and T. A. Ohki, *Sci. Rep.*
460 **9** (2019), 10.1038/s41598-018-37204-3.
- 461 ⁵ I. Kanada, A. Cruce, T. Mewes, S. Wu, C. Mewes, G. Mankey, and T. Suzuki, *AIP Adv.* **7**,
462 056105 (2017).
- 463 ⁶ S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S.-H. Yang,
464 *Nat. Mater.* **3**, 862 (2004).
- 465 ⁷ Y. Ando, T. Miyakoshi, M. Oogane, T. Miyazaki, H. Kubota, K. Ando, and S. Yuasa, *Appl.*
466 *Phys. Lett.* **87**, 142502 (2005).
- 467 ⁸ T. L. Gilbert, *Phys. Rev.*, **100**, 1243 (1955).
- 468 ⁹ T. Gilbert, *IEEE Transactions on Magnetics* **40**, 3443 (2004).
- 469 ¹⁰ B. Heinrich, D. Fraitová, and V. Kamberský, *Phys. Status Solidi B* **23**, 501 (1967).
- 470 ¹¹ V. Kamberský, *Czech. J. Phys.* **26**, 1366 (1976).
- 471 ¹² Y. Tserkovnyak, G. A. Fiete, and B. I. Halperin, *Appl. Phys. Lett.* **84**, 5234 (2004).
- 472 ¹³ E. Rossi, O. G. Heinonen, and A. H. MacDonald, *Phys. Rev. B* **72**, 174412 (2005).
- 473 ¹⁴ M. A. W. Schoen, D. Thonig, M. L. Schneider, T. J. Silva, H. T. Nembach, O. Eriksson,
474 O. Karis, and J. M. Shaw, *Nat. Phys.* **12**, 839 (2016).
- 475 ¹⁵ K. Gilmore, Y. U. Idzerda, and M. D. Stiles, *Phys. Rev. Lett.* **99**, 027204 (2007).
- 476 ¹⁶ S. Mankovsky, D. Ködderitzsch, G. Woltersdorf, and H. Ebert, *Phys. Rev. B* **87**, 014430 (2013).
- 477 ¹⁷ Eddy-current damping[?] and radiative damping⁴⁰ can also contribute to viscous damping, but
478 they typically constitute a small correction that is $\lesssim 10\%$ of intrinsic Gilbert damping in $\lesssim 20$
479 nm thick ferromagnetic thin films^{18,37}, which is thought to be rooted in the electronic band
480 structure of the ferromagnetic metal^{14–16}.
- 481 ¹⁸ B. Khodadadi, A. Rai, A. Sapkota, A. Srivastava, B. Nepal, Y. Lim, D. A. Smith, C. Mewes,

482 S. Budhathoki, A. Hauser, M. Gao, J.-F. Li, D. Viehland, Z. Jiang, J. Heremans, P. Balachan-
483 dran, T. Mewes, and S. Emori, Phys. Rev. Lett. **124**, 157201 (2020).

484 ¹⁹ S. Geschwind and A. M. Clogston, Phys. Rev. **108**, 49 (1957).

485 ²⁰ R. C. LeCraw, E. G. Spencer, and C. S. Porter, Phys. Rev. **110**, 1311 (1958).

486 ²¹ E. Schlömann, J. Phys. Chem. Solids **6**, 257 (1958).

487 ²² C. E. Patton, C. H. Wilts, and F. B. Humphrey, J. Appl. Phys. **38**, 1358 (1967).

488 ²³ R. Arias and D. L. Mills, Phys. Rev. B **60**, 7395 (1999).

489 ²⁴ R. Arias and D. L. Mills, J. Appl. Phys. **87**, 5455 (2000).

490 ²⁵ R. McMichael and P. Krivosik, IEEE Transactions on Magnetics **40**, 2 (2004).

491 ²⁶ G. Woltersdorf and B. Heinrich, Phys. Rev. B **69**, 184417 (2004).

492 ²⁷ N. Mo, Y.-Y. Song, and C. E. Patton, J. Appl. Phys. **97**, 093901 (2005).

493 ²⁸ S. S. Kalarickal, N. Mo, P. Krivosik, and C. E. Patton, Phys. Rev. B **79**, 094427 (2009).

494 ²⁹ J. Lindner, I. Barsukov, C. Raeder, C. Hassel, O. Posth, R. Meckenstock, P. Landeros, and
495 D. L. Mills, Phys. Rev. B **80**, 224421 (2009).

496 ³⁰ S. Jiang, L. Sun, Y. Yin, Y. Fu, C. Luo, Y. Zhai, and H. Zhai, AIP Adv. **7**, 056029 (2017).

497 ³¹ E. R. Edwards, H. T. Nembach, and J. M. Shaw, Phys. Rev. Appl **11**, 054036 (2019).

498 ³² A. Ghosh, S. Auffret, U. Ebels, and W. E. Bailey, Phys. Rev. Lett. **109**, 127202 (2012).

499 ³³ M. A. W. Schoen, J. Lucassen, H. T. Nembach, T. J. Silva, B. Koopmans, C. H. Back, and
500 J. M. Shaw, Phys. Rev. B **95**, 134410 (2017).

501 ³⁴ D. E. Bürgler, C. M. Schmidt, D. M. Schaller, F. Meisinger, R. Hofer, and H.-J. Güntherodt,
502 Phys. Rev. B **56**, 4149 (1997).

503 ³⁵ G. Vignaud and A. Gibaud, J. Appl. Crystallogr. **52**, 201 (2019).

504 ³⁶ Here, the “average roughness” is the average of the roughness of the top and bottom interfaces
505 of the Fe layer.

506 ³⁷ D. A. Smith, A. Rai, Y. Lim, T. Q. Hartnett, A. Sapkota, A. Srivastava, C. Mewes, Z. Jiang,
507 M. Clavel, M. K. Hudait, D. D. Viehland, J. J. Heremans, P. V. Balachandran, T. Mewes, and
508 S. Emori, Phys. Rev. Appl **14**, 034042 (2020).

509 ³⁸ The magnitude of the inhomogenous broadening ΔH_0 seen in OP FMR ranges from ≈ 10 to 50
510 Oe with no clear systematic dependence on Fe film thickness or seed layer material.

511 ³⁹ $\gamma/2\pi \approx 2.9$ MHz/Oe corresponds to a spectroscopic g -factor of $g \approx 2.08$, in line with Ref.³³.

512 ⁴⁰ M. A. W. Schoen, J. M. Shaw, H. T. Nembach, M. Weiler, and T. J. Silva, Phys. Rev. B **92**,

513 184417 (2015).

514 ⁴¹ H. Wang, C. Du, Y. Pu, R. Adur, P. Hammel, and F. Yang, *Physical Review Letters* **112**,
515 197201 (2014).

516 ⁴² C. Du, H. Wang, F. Yang, and P. C. Hammel, *Physical Review B* **90**, 140407 (2014).

517 ⁴³ B. Heinrich, K. B. Urquhart, A. S. Arrott, J. F. Cochran, K. Myrtle, and S. T. Purcell, *Phys.*
518 *Rev. Lett.* **59**, 1756 (1987).

519 ⁴⁴ Z. Celinski and B. Heinrich, *J. Appl. Phys.* **70**, 5935 (1991).

520 ⁴⁵ L. Chen, S. Mankovsky, S. Wimmer, M. A. W. Schoen, H. S. Korner, M. Kronseder, D. Schuh,
521 D. Bougeard, H. Ebert, D. Weiss, and C. H. Back, *Nature Phys.* **14**, 490–494 (2018).

522 ⁴⁶ K. Chen and Z. Shufeng, *Phys. Rev. Lett*IEEE *Trans. Magn.* **11453**, 126602 (20157).

523 ⁴⁷ K. Gilmore, *Precession damping in itinerant ferromagnets*, Ph.D. thesis, Montana State
524 University-Bozeman, College of Letters & Science (2007).

525 ⁴⁸ G. S. Abo, Y.-K. Hong, J. Park, J. Lee, W. Lee, and B.-C. Choi, *IEEE Transactions on*
526 *Magnetics* **49**, 4937 (2013).

527 ⁴⁹ K. Gilmore, M. D. Stiles, J. Seib, D. Steiauf, and M. Fähnle, *Phys. Rev. B* **81**, 174414 (2010).

528 ⁵⁰ J. T. Hou and L. Liu, *Phys. Rev. Lett.* **123**, 107702 (2019).

529 ⁵¹ Y. Li, T. Polakovic, Y.-L. Wang, J. Xu, S. Lendinez, Z. Zhang, J. Ding, T. Khaire, H. Saglam,
530 R. Divan, J. Pearson, W.-K. Kwok, Z. Xiao, V. Novosad, A. Hoffmann, and W. Zhang, *Phys.*
531 *Rev. Lett.* **123**, 107701 (2019).

532 ⁵² S. S. Kalarickal, P. Krivosik, J. Das, K. S. Kim, and C. E. Patton, *Phys. Rev. B* **77**, 054427
533 (2008).

534 ⁵³ W. K. Peria, T. A. Peterson, A. P. McFadden, T. Qu, C. Liu, C. J. Palmstrøm, and P. A.
535 Crowell, *Phys. Rev. B* **101**, 134430 (2020).

536 ⁵⁴ A. Azevedo, A. B. Oliveira, F. M. de Aguiar, and S. M. Rezende, *Phys. Rev. B* **62**, 5331 (2000).

537 ⁵⁵ W. K. Peria, X. Wang, H. Yu, S. Lee, I. Takeuchi, and P. A. Crowell, *Phys. Rev. B* **103**,
538 L220403 (2021).

539 ⁵⁶ A. E. Clark, K. B. Hathaway, M. Wun-Fogle, J. B. Restorff, T. A. Lograsso, V. M. Keppens,
540 G. Petculescu, and R. A. Taylor, *J. Appl. Phys.* **93**, 8621 (2003).

541 ⁵⁷ E. M. Summers, T. A. Lograsso, and M. Wun-Fogle, *J Mater Sci* **42**, 9582 (2007).

542 ⁵⁸ M. Pelliccione and T.-M. Lu, in *Springer Series in Materials Science*, Vol. 108 (Springer, 2008).

543 ⁵⁹ See Supplemental Material at <http://link.aps.org/supplemental/xx.xxxx> for additional infor-

544 mation on the frequency dependence of two-magnon scattering, including videos of simulation
545 results with several values of ξ .