Two-magnon frequency-pulling effect in ferromagnetic resonance

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We report the experimental observation in thin films of the hybridization of the uniform ferromagnetic resonance mode with nonuniform magnons as a result of the two-magnon scattering mechanism, leading to a frequency-pulling effect on the ferromagnetic resonance. This effect, when not properly accounted for, leads to a discrepancy in the dependence of the ferromagnetic resonance field on frequency for different field orientations. The frequency-pulling effect is the complement of the broadening of the ferromagnetic resonance lineshape by two-magnon scattering and can be calculated using the same parameters. By accounting for the two-magnon frequency shifts through these means, consistency is achieved in fitting data from in-plane and perpendicular-to-plane resonance conditions.

Magnetization dynamics of ferromagnets are influenced by many factors, such as magnetocrystalline anisotropy, interfacial anisotropy, and Gilbert damping. It is of technological interest to study these phenomena in order to understand, for example, the physics governing magnetization switching by spin torque. Two-magnon scattering (TMS), an extrinsic relaxation process of uniform magnetization precession in ferromagnets, is an important phenomenon that influences magnetization dynamics. TMS is commonly observed as a broadening of ferromagnetic resonance (FMR) lineshapes, but it may also lead to a shift in the FMR frequency. Although the broadening of the FMR lineshape caused by TMS is often impossible to ignore, the latter effect is more subtle and almost universally neglected when FMR data are used to extract static magnetic properties of materials.

In this letter, we demonstrate the existence of a frequency-pulling effect in ferromagnetic resonance induced by two-magnon scattering in a polycrystalline Fe$_{0.7}$Ga$_{0.3}$ thin film for in-plane applied magnetic fields. We showed that the line shifts can be accounted for in a manner that is self-consistent with the observed linewidth-broadening induced by two-magnon scattering and the field-dependent resonance dispersion when the applied magnetic field is perpendicular to the plane.

The samples used in this report are 33 nm Fe$_{0.7}$Ga$_{0.3}$ films deposited on Si/SiO$_2$ substrates at room temperature by sputtering. The working pressure was adjusted by Ar (99.999%). The films were verified to be polycrystalline and without texture, i.e. preferred crystalline orientation, using x-ray diffraction (XRD). The lack of magnetic anisotropy in the plane of the film was verified with vibrating sample magnetometry (VSM) and FMR. Grain boundaries were observed with atomic force microscopy (AFM), yielding an average grain diameter of ~15 nm [see Fig. 1(a)]. This is in good agreement with the structural coherence length, which was estimated to be 13 nm with XRD. Figure 1(b) shows a symmetric $\theta/2\theta$ XRD scan of the Fe$_{0.7}$Ga$_{0.3}$ (110) peak with the scattering vector normal to the plane. It was verified with a two-dimensional detector that the intensity of this Bragg peak is constant for fixed values of the scattering angle $2\theta$ as the scattering vector is canted into the plane, indicating the absence of texture.

![Image](https://example.com/fig1.png)

**FIG. 1.** (a) Atomic force microscopy image of the Fe$_{0.7}$Ga$_{0.3}$ film. Root-mean-square roughness is 0.7 nm. (b) X-ray diffraction symmetric scan of the Fe$_{0.7}$Ga$_{0.3}$ film showing the (110) Bragg peak. The structural coherence length determined from the full-width-at-half-maximum of the Bragg peak is 13 nm. (c) Thin-film magnon dispersion for in-plane magnetization and wavevectors $\mathbf{q} \parallel \mathbf{M}$, with an arrow indicating the two-magnon scattering process. (d) Field-swept FMR linewidths of the Fe$_{0.7}$Ga$_{0.3}$ film with in-plane applied magnetic field overlaid with a fit to a combined two-magnon scattering and Gilbert damping model. The Gilbert damping $\alpha$ (a fit parameter) and defect correlation length $\xi$ (fixed) are shown on the figure.

Ferromagnetic resonance lineshapes were measured at room temperature using a coplanar waveguide with modulation of the applied magnetic field for lock-in detection as described in Ref. 5. The lineshapes were measured for both in-plane (IP) and perpendicular-to-plane (PP) applied fields. When the field (and hence, magnetization) is IP, there are...
magnons degenerate with the $q = 0$ FMR magnon [see Fig. 1(c)]. This leads to a possible scattering mechanism of the FMR mode, observable through its nonlinear effect on the field-swept linewidth as a function of frequency, shown in Fig. 1(d) for the Fe$_7$Ga$_{0.3}$ film. This is the TMS process, and it is allowed as long as some assisting process enables conservation of momentum.

The resonance frequency was fit as a function of the applied magnetic field $H_0$ to the Kittel equation for a thin film with no in-plane magnetic anisotropy, which reads as

$$\omega_{FMR} = \gamma \sqrt{H_0(H_0 + 4\pi M_{eff})} \tag{1}$$

for $H_0$ in the plane and

$$\omega_{FMR} = \gamma (H_0 - 4\pi M_{eff}) \tag{2}$$

where $H'$ is the root-mean-square inhomogeneity field, $\xi$ is the defect correlation length, $\Lambda_{0q}$ is the magnon-magnon coupling strength, and $\delta \omega = (d\omega / dH)_{H_{FMR}}$ is the Gilbert frequency half-width-at-half-maximum linewidth. The imaginary part of Eq. 3 corresponds to the well-known TMS contribution to the FMR scattering rate, i.e. linewidth.

$$\Delta \omega_{TMS} = \frac{\gamma^2 \xi^2 H'^2}{\Lambda_{0q}(1 + (q\xi)^2)^{3/2}} \frac{1}{\pi(\omega - \omega_{FMR}) - i \delta \omega} \tag{3}$$
A small amount of surface anisotropy may lead to an anisotropy factor, which together can in principle be used to predict the IP FMR frequencies to Eq. (1), fixing $g$ using the red curve shown in Fig. 3, representing the FMR field-dependent dispersion for IP magnetization. A direct measurement of the dispersion for IP magnetization is not possible due to the line shifts caused by TMS. To address this problem, the IP FMR frequencies are blue-shifted using the red curve shown in Fig. 3, representing the FMR frequencies in the absence of TMS. We then fit the corrected IP FMR frequencies to Eq. (1), fixing $4\pi M_{eff}$ to the PP value of 13.821 kOe and leaving the $g$-factor as a free parameter. A small amount of surface anisotropy may lead to an anisotropy factor. The absence of TMS for PP magnetization is of particular convenience in our case because it allows determination of the effective demagnetizing field $4\pi M_{eff}$ and the Landé $g$-factor, which together can in principle be used to predict the FMR field-dependent dispersion for IP magnetization. A direct measurement of the dispersion for IP magnetization is not possible due to the line shifts caused by TMS. To address this problem, the IP FMR frequencies are blue-shifted using the red curve shown in Fig. 3, representing the FMR frequencies in the absence of TMS. We then fit the corrected IP FMR frequencies to Eq. (1), fixing $4\pi M_{eff}$ to the PP value of 13.821 kOe and leaving the $g$-factor as a free parameter. A small amount of surface anisotropy may lead to an anisotropy factor. 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the ferromagnetic resonance response of ultrathin films,”  
Physical Review B 60, 7395–7409 (1999)
3 R. McMichael and P. Krivosik, “Classical Model of Extrinsic Ferromagnetic Resonance Linewidth in Ultrathin Films,”  
IEEE Transactions on Magnetics 40, 2–11 (2004)
4 P. Krivosik, N. Mo, S. Kalarickal, and C. E. Patton, “Hamiltonian formalism for two magnon scattering microwave relaxation: Theory and applications,”  
Journal of Applied Physics 101, 083901 (2007)
5 W. K. Peria, T. A. Peterson, A. P. McFadden, T. Qu, C. Liu, C. J. Palmstrøm, and P. A. Crowell, “Interplay of large two-magnon ferromagnetic resonance linewidths and low Gilbert damping in Heusler thin films,”  
Physical Review B 101, 134430 (2020)
6 M. Farle, “Ferromagnetic resonance of ultrathin metallic layers,”  
Reports on Progress in Physics 61, 755–826 (1998)
7 A. E. Clark, M. Wun-Fogle, J. B. Restorff, K. W. Dennis, T. A. Lograsso, and R. W. McCallum, “Temperature dependence of the magnetic anisotropy and magnetostriction of Fe$_{100-x}$Ga$_x$ (x = 8.6, 16.6, 28.5),”  
Journal of Applied Physics 97, 10M316 (2005)
8 D. B. Gopman, V. Sampath, H. Ahmad, S. Bandyopadhyay, and J. Atulasimha, “Static and Dynamic Magnetic Properties of Sputtered Fe–Ga Thin Films,” IEEE Transactions on Magnetics 53, 1–4 (2017)
9 M. J. Hurben and C. E. Patton, “Theory of two magnon scattering microwave relaxation and ferromagnetic resonance linewidth in magnetic thin films,”  
Journal of Applied Physics 103, 4344–4365 (1998)
10 A. Y. Dobin and R. H. Victoria, “Surface Roughness Induced Extrinsic Damping in Thin Magnetic Films,”  
Physical Review Letters 92, 257204 (2004)
11 G. Woltersdorf and B. Heinrich, “Two-magnon scattering in a self-assembled nanoscale network of misfit dislocations,”  
Physical Review B 69, 184417 (2004)
12 S. S. Kalarickal, P. Krivosik, J. Das, K. S. Kim, and C. E. Patton, “Microwave damping in polycrystalline Fe–Ti–N films: Physical mechanisms and correlations with composition and structure,”  
Physical Review B 77, 054427 (2008)
13 A. Azevedo, A. B. Oliveira, F. M. de Aguiar, and S. M. Rezende, “Extrinsic contributions to spin-wave damping and renormalization in thin Ni$_{50}$Fe$_{50}$ films,”  
Physical Review B 62, 5331–5333 (2000)