Nonlinear control of magnon polaritons

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We experimentally and theoretically demonstrate that nonlinear spin-wave interactions suppress the hybrid magnon-photon quasiparticle or “magnon polariton” in microwave spectra of an yttrium iron garnet film detected by an on-chip split-ring resonator. We observe a strong coupling between the Kittel magnon and microwave cavity mode in terms of an anti-crossing as a function of magnetic fields at low microwave input powers, but a complete closing of the anti-crossing gap at high powers. The experimental results are well explained by a theoretical model including the three-magnon decay of the Kittel magnon into spin waves. The gap closure originates from the saturation of the ferromagnetic resonance above the Suhl instability threshold by a coherent back reaction from the spin waves.

The spectral properties of many-body systems can often be captured in terms of weakly interacting quasiparticles. In magnets, the low-energy excitations of the ordered local moments are Bosonic quasiparticles called magnons [1]. Their unique and complex dispersion relations are governed by short-range exchange and long-range magnetic dipolar interactions, of which the latter depends strongly on the shape of the ferromagnet [2]. In thin films, the dispersion becomes strongly anisotropic, with negative (positive) group velocity for an in-plane wave vector $k$ parallel (perpendicular) to the magnetization direction $M$, as illustrated in Fig. 1(a). The competition between exchange and dipolar energies leads to two valleys in the dispersion relation around a minimum energy for $k \parallel M$ that may host magnon Bose-Einstein condensates [3].

The spectrum of non-interacting quasiparticles is fully characterized by the dispersion relation alone. Theoretically, non-interacting magnons emerge as the leading term in the Holstein-Primakoff expansion of the Heisenberg spin Hamiltonian that holds in the limit of a small number of magnons, i.e. when the magnet is weakly excited and at temperatures sufficiently below the Curie temperature. When this is not the case, nonlinear corrections become increasingly important. To leading order these are three- and four-magnon interaction terms that conserve energy and linear momentum of the quasiparticles. The dispersion relation governs which many-body processes obey the conservation laws, and the three-magnon splitting does in moderately thin films (Fig. 1(a)), which therefore generically dominates over the higher-order processes [4]. The nonlinear interactions can significantly alter the spectral properties from what is predicted from the dispersion relation. For example, the ferromagnetic resonance by microwave magnetic fields encounters a Suhl instability at a threshold power [5–7] that can be explained by three-magnon splitting [8–14]. In this process, the spatially uniform Kittel mode decays into a counter-propagating pair of magnons at half the FMR frequency [Fig. 1(b)]. At the Suhl excitation threshold, the energy flux from the spatially uniform Kittel mode to the magnon pair at nonzero wave numbers exceeds the relaxation rate of the latter. This critical power is therefore small for low-damping magnets such as yttrium iron garnet (YIG). The formation of a coherently coupled hybrid of three magnon modes suppresses microwave absorption by Kittel mode [6, 15, 16] and the spectral line shape deviates from Lorentzian [4], which is a clear signature of the nonlinearity.

Recent research in magnon-photon interactions focuses on the magnon polariton hybrids of the Kittel magnon and a microwave cavity mode [17–20]. Purely magnetic systems strongly coupled to cavity photons can operate as gradient memories [21], probabilistic-bits [22], or to distill the entanglement of interacting magnons [23]. It also realizes (quantum) information transfer between a localized spin system and a distant superconducting qubit [24]. The emphasis of this field has been on studies of the strong coupling limit in the weakly excited regime [25–41], i.e. that described by the coupling of two harmonic oscillators. Nonlinearities of magnon-polaritons are essential for considering them in novel computing paradigms [42–44], but they have been hardly studied so far [45, 46].

In this Letter, we report experimental and theoretical results on a first-order Suhl instability [6] of magnon polaritons, i.e. the quasi-particles formed by the strong coupling of the Kittel magnon with a cavity photon [25–30] as illustrated in Fig. 1(b). We reveal suppression of the anti-crossing gaps in the microwave reflection spectra.

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The Suhl instability alters magnetic susceptibility [6, 15, 16] and lineshape [4] due to a nonlinear back reaction of the unstable pairs of nonzero-$\mathbf{k}$ magnons on the Kittel mode. We therefore measured the $P$ dependence of $\Delta \omega_0$ for $H = 14.4$ mT ($<16.8$ mT) as shown in Fig. 2(d) and observe that the peaks broaden when $P \gtrsim -10$ dBm. This indicates a power-dependent distortion of lineshape, and thereby the onset of Suhl instability. For $H = 26.1$ mT (>16.8 mT) the broadening begins at a higher power $P \gtrsim 0$ dBm. This difference may be caused by field-dependent nonlinear coupling and relaxation processes or simply by the choice of the field value that is farther away from the low-$P$ anticrossing field of 16.8 mT.

In order to highlight the role of three-magnon scattering, we tune the FMR to a higher cavity mode of our SRR by a larger magnetic field. The three-magnon decay process conserves energy, i.e., $\omega_0 \geq 2 \omega_0^\text{bottom}$ where $\omega_0^\text{bottom}$ is the minimum of the magnon band. When increasing the magnetic field and hence $\omega_0$, $\omega_k$, the Kittel magnon cannot decay when $H > H_{cr}$. This threshold field corresponds to the frequency $\omega_0 (H_{cr}) = 2.3$ GHz for a YIG sample with thickness of 5 $\mu$m, $M_s = 1.15 \times 10^5$ A/m and an stiffness constant of $\lambda_{xx} = 3 \times 10^{-16}$ m$^2$ [51] (see Supplementary Materials II). The magnon polariton of the 3 GHz SRR mode in Fig. 3(a) should therefore be much less dependent on the microwave power. By increasing $P$ up to 8 dBm as before, the reflection spectrum (Fig. 3(b)) and the fixed-field plot in Fig. 3(c) confirm that the anticrossing gap does not vanish now. The FMR linewidth in Fig. 3(d) also does not change, which supports our hypothesis that the Suhl instability is the culprit for the apparent vanishing of the magnon polariton of the low-frequency cavity mode.

We address the nonlinearities of the magnon polariton by extending the kinetic theory of nonlinear spin wave dynamics [4, 52]. We start from the model Hamiltonian $\mathcal{H} = \mathcal{H}_2 + \mathcal{H}_3$, in which $\mathcal{H}_2$ describes non-interacting quasi particles

$$\mathcal{H}_2 = \omega_0 b_k^\dagger b_k + \left[ \frac{\hbar c}{\omega} U_0 b_k^\dagger b_k + \text{h.c.} \right] + \sum_{k \neq 0} \omega_k b_k^\dagger b_k$$

$$+ \omega_1 b_0^\dagger b_r + \left[ g b_0^\dagger b_r + h \right] + \text{h.c.},$$  

where $\omega_k$ is the frequency of a magnon with wave vector $\mathbf{k}$, $\omega_r$ the selected cavity mode frequency, $\hbar$ and $\omega$ are the amplitude and frequency of the applied microwave field, and $U_0$ and $U_r$ are the coupling strengths of the microwave with the Kittel and cavity modes, respectively. The nonlinear term $\mathcal{H}_3$ represents the three-magnon scattering

$$\mathcal{H}_3 = \frac{1}{2} \sum_{k \neq 0} V_k b_0 b_k^\dagger b_{-k} + \text{h.c.},$$

in which the nonlinear coupling $V_k$ is a function of the material parameters [4]. Overlines are used to de-
FIG. 2. (a) Microwave absorption spectra ($S_{11}$ (dB)) as a function of microwave frequency (from 1.3 to 1.7 GHz) and magnetic field strength (from 14 to 20 mT), for different microwave power ranging from -20 to 8 dBm. (b)&(c) Collection of frequency domain scans for a fixed magnetic field of 16.8 mT. (d) Peak positions are extracted from individual fits for different microwave powers (red and blue dots for the upper and lower modes). The gap size represented by the black dots is then calculated by using the two peak positions for each power. (e) Microwave power evolution of extracted Kittel mode linewidth before and after the anti-crossing, respectively at 14.4 (blue crosses) and 26.1 mT (red squares).

FIG. 3. (a)&(b) $|S_{11}|$ as a function of microwave frequency (from 1 to 4 GHz) and magnetic field strength (from 0 to 75 mT), for low and high microwave powers, respectively at -20 and 8 dBm. (c) A collection of microwave absorption spectra for the 3 GHz SRR mode at a fixed magnetic field of 60 mT, where the mode hybridization is at its strongest. (d) Power evolution of Kittel mode linewidth ($\Delta \omega_0$) at a fixed magnetic field above the anti-crossing (63 mT) for the 3 GHz SRR mode.
note complex conjugation throughout. We omitted fourmagnon scattering terms since the critical power for the first order Suhl instability is much smaller than the second order one in the present setup (see Supplementary Materials for details). The $b_k$ and $b_r$ are quantum mechanical annihilation operators for the magnons and photons. However, we may treat them as well as classical random wave amplitudes, noting that thermal fluctuations wipe out the quantum effects at room temperature. In the film geometry with an in-plane static magnetic field, only a narrow band of magnons are involved in the onset of the instabilities [53], which we approximate here by a single pair $\pm k \parallel H$ with smallest $\eta_k/|V_k|$ where $\eta_k = \eta_{-k}$ is the relaxation rate of the magnon at wavevector $k$. We are then looking for the steady-state solutions $\langle b_{0,r} \rangle$ and $\langle b_k b_{-k} \rangle$ where the $\langle \cdots \rangle$ denotes an average over the thermal noise that is governed by the phenomenological relaxation rates $\eta_k$ and the temperature via the fluctuation dissipation theorem. The coherent amplitude of the Kittel mode $\langle b_0 \rangle$ is a root of a (complex) cubic algebraic equation (Eq. (S25) in Supplementary Materials), which in the high-power limit $|h| \rightarrow \infty$ has a finite limit

$$\langle b_0 \rangle \rightarrow -e^{-i\omega t+i\psi_k}c_{cr}, \quad c_{cr} = \frac{\omega_k - \omega/2 + i\eta_k}{V_k},$$

where $\psi_k$ is the phase of the magnon pair amplitude $\langle b_k b_{-k} \rangle$. The absence of $h$ on the r.h.s. implies saturation, i.e. the number of Kittel magnons cannot increase beyond the critical value $|c_{cr}|^2$, which depends only on the magnon Hamiltonian. Furthermore,

$$\langle b_r \rangle = \frac{\bar{g}\langle b_0 \rangle + h U_r e^{-i\omega t}}{\omega - \omega_r + i\eta_r}, \quad (4)$$

$$\langle b_k b_{-k} \rangle = -\frac{c_{cr} \langle b_0 \rangle}{|c_{cr}|^2 - |\langle b_0 \rangle|^2} \frac{k_B T}{\hbar \omega_k}, \quad (5)$$

where $T$ is the temperature and $k_B$ the Boltzmann constant. Photon and magnon pair amplitudes share the phase of the Kittel mode, which is in turn fixed by the phase of the driving field $h$.

Figures 4(a,b) summarize the theoretical results. Here we adopted the standard parameters for YIG in combination with the extracted value of the saturation magnetization $M_s = 1.15 \times 10^5$ A/m, viz. $\gamma/2\pi = 28$ GHz/T, and magnetic relaxation $\eta_0/2\pi = 15$ MHz. We model the microwaves system by $\omega_r/2\pi = 1.53$ GHz, $\eta_c/2\pi = 33.5$ MHz, $U_r = i$, $U_0 = 0.3$, and use the magnon-photon coupling $g = 40.9$ MHz from the observed anticrossing at low $P$. The Kittel formula is $\omega_0 = \mu_0 \gamma/\hbar (H + M_s)$. For the coherently coupled magnon pair at $\omega_k = \omega/2$, we assume $\eta_k = 0.01 \times \omega_0/2$ and $V_k = \mu_0 \gamma M_s \times 10^{-11}/2$. These choices are rationalized in Supplementary Materials.

Figures 4(a,b) compare favorably with the experimental results in Fig. 2 for not only the gap closure but also the lineshapes. The latter seem somewhat similar to the level attraction associated with dissipative coupling [39, 54], but we use here a constant and real photon-magnon coupling. We can now explain the quenching of the anti-crossing in terms of a back reaction of the magnon pair on the Kittel magnon [Fig. 4(c)] that both

FIG. 4. (a) Microwave reflection spectrum $S_{11}$ (defined in Supplementary Materials) calculated from Eqs. (3) and (4) for model parameters in the text and an arbitrary reference input power $h_{ref}$. (b) $S_{11}$ at $\mu_0 H = 16.8$ mT. (c) Schematic of the particle number growth with $h$ for magnon polaritons with the Suhl instability and energy flux diagram for $U_0 = 0$ in the stationary state. The yellow arrows indicate the energy flux, $n_{r,0,k}$ denote the number of photons, Kittel mode magnons, and valley magnons, respectively.
limits the growth of the unstable magnon pair numbers and saturates the Kittel mode Eq. (3). Below the critical power, a photon injected into the cavity hybridizes with the Kittel mode, which opens the anti-crossing gap. When $\langle n_0 \rangle$ is fully saturated, however, an added cavity photon does not have a partner to hybridize with. The cavity thus becomes transparent, with a single peak at the original resonance frequency $\omega_r$ and damping $\eta_r$.

In summary, we discovered that a high microwave power input suppresses the strong magnon-photon coupling in microwave cavities via the first-order Suhl instability. We reproduced the closure of the hybridization gap by a nonlinear spin-wave model coupled to a microwave cavity photon mode and explained the physics in terms of the saturation of Kittel magnon numbers for large microwave drives. This effect is a result of the phase coherence between the photons and the entire spin wave system. The ability to coherently excite or detect magnon pairs in the low energy valleys opens new avenues in magnonics, such as the microwave spectroscopy of magnon Bose Einstein condensates. We remark that the complete saturation of Eq. (3) relies on the simplifying assumptions made in the theoretical modeling. The $k$ dependence of $V_k$ and inclusion of four-magnon contributions can lead to even richer dynamical structures. The present work points to an ample room for unexpected discoveries in nonlinear magnonics and makes it an exciting research frontier.

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I. EXPERIMENTAL GEOMETRY AND MODE PROFILE OF THE RESONATOR

We employed a combination of the time domain and the eigenmode solvers from the CST STUDIO SUITE 2021 package to simulate and calculate the response to a microwave input signal and its corresponding field distributions on the split ring resonator (SRR) used in our experiments. The schematic and dimensions of the SRR are shown in Fig. S1(a). A square-shaped resonator that consists an outer length of \(a = 10\) mm and an inner width of \(b = 0.25\) mm with a gap of distance \(c = 0.125\) mm has been prepared with a 17.5 \(\mu\)m double-sided copper coating, fabricated on a ROGERS RT6010LM substrate of height \(h = 1.5\) mm with a relative permittivity, \(\epsilon_r = 10.7\) and loss \(\tan\delta = 2.3 \times 10^{-3}\). A 50 \(\Omega\) impedance-matched microstrip line of width \(w = 1.5\) mm is inductively coupled to the SRR, separated by a distance \(d = 0.125\) mm. Figure S1(b) shows comparison between experimental results of the microwave absorption parameters \(S_{11}\) (dotted green) in a vector network analyser (VNA) and simulated \(S_{11}\) (blue) and \(S_{21}\) (red) as a function of frequency. Between 1 and 4 GHz, two resonance modes at 1.51 and 2.87 GHz are obtained by simulation, which we respectively attribute the lower and the upper modes as \(\omega_r\) and \(\omega_{\text{ref}}\). From our experimental \(S_{11}\) data (green) with the YIG sample placed on the SRR (see Fig. 1(c) in main text), we find \(\omega_{r(\text{exp})} = 1.53\) and \(\omega_{\text{ref}(\text{exp})} = 3.09\) GHz. Although a trivial shift of \(\omega_{\text{ref}}\) mode is observed (difference \(\approx 7.1\) %), we notice that the resonance position of \(\omega_r\) mode is hardly affected (difference \(\approx 1.3\) %) when the sample is introduced. In Figs. S1(c)-(e), we respectively show the local distributions for different magnetic field components \(H_x\), \(H_y\) and \(H_z\) of the \(S_{11}\) mode at \(\omega/2\pi = \omega_r\) (left) and \(\omega/2\pi = \omega_{\text{ref}}\) (right). At the resonance frequency, as the microwave currents are flown through the microstrip line, the energy transfer of the electromagnetic fields are absorbed by the SRR, which subsequently induces an oscillating magnetic field excitation in its local geometry.
Figure S1. (a) Schematic drawing of the SRR used in the experimental setup. (b) Calculated microwave reflection and transmission spectra ($S_{11}$ & $S_{21}$) as a function of frequency (from 1.0 to 4 GHz) of dimensions depicted in (a). (c)-(e) Surface plots of local magnetic field distribution components for (c) $H_x$, (d) $H_y$ and (e) $H_z$ of the $S_{11}$ mode at the primary ($\omega_r/2\pi = 1.51$ GHz) (left) and the secondary ($\omega_{ref}/2\pi = 2.87$ GHz) (right) mode frequencies, respectively.
II. SPIN WAVE DISPERSION IN THE YIG FILM

The YIG film used in the experiment is $d = 5 \mu m$ thick with magnetic-field ($H$) dependent dispersion relation [2]

$$\omega_{nk}(H) = \mu_0 \gamma \sqrt{\left\{ H + M_s \left( 1 + \lambda_{ex} k_n^2 - P_n \right) \right\} \left\{ H + M_s \left( \lambda_{ex} k_n^2 + P_n \sin^2 \phi \right) \right\}}.$$  \hspace{1cm} (S1)

Here $M_s$, $\gamma$, $\lambda_{ex}$ are the saturation magnetization, gyromagnetic ratio and stiffness constant, respectively. $k = (k_x, k_y)^T = k(\cos \phi \sin \phi)^T$ denotes the wavevector in the plane, $n\pi/d, n \in \mathbb{Z}$ is the quantized out-of-plane component such that $k_n^2 = k^2 + (n\pi/d)^2$, and

$$P_n = \frac{k^2}{k_n^2} - \frac{2}{1 + \delta_n} \frac{k^4 \left( 1 - (-1)^n e^{-kd} \right)}{kd}$$ \hspace{1cm} (S2)

for free surface boundary conditions.

To compute the threshold field value $H_{cr}$ for the occurrence of three-magnon splitting, we study the backward volume wave branch $\phi = 0$ without a node in the thickness direction $n = 0$, which has the lowest resonance frequency. For each given value of $H$, the energy-momentum conservation requires

$$H (H + M_s) = 4 \left( H + M_s \lambda_{ex} k^2 \right) \left\{ H + M_s \left( \lambda_{ex} k^2 + \frac{1 - e^{-kd}}{kd} \right) \right\}. \hspace{1cm} (S3)$$

The condition on $k$ for $\omega_{nk}(H)$ to be the bottom of dispersion is

$$\left( 4\lambda_{ex} k^2 + e^{-kd} - \frac{1 - e^{-kd}}{kd} \right) H + \left( 4\lambda_{ex} k^2 + e^{-kd} + \frac{1 - e^{-kd}}{kd} \right) M_s \lambda_{ex} k^2 = 0. \hspace{1cm} (S4)$$

The simultaneous solution of Eqs. (S3) and (S4) for $(H, k)$ determines $H_{cr}$ and the associated $k_{cr}$ that minimizes $\omega_{nk}(H_{cr})$.

![Figure S2](image)

Figure S2. (a) Magnetic field dependence of microwave excitation frequency $\omega_0/2\pi$. The dots indicate the experimental data in combination with the Kittel fit shown by dashed yellow lines. The solid green curve is the calculated spin wave dispersion obtained by Eq. (S1) for the lowest energy mode, $n = 0$. The dashed red line represents the onset frequency of the three-magnons. (b) $\omega_0/4\pi$ and $\omega_{k,\text{bottom}}/2\pi$ as a function of magnetic field. The intersection point corresponds to the critical field $\mu_0 H_{cr}$ for the onset of three-magnon splitting and half the onset frequency. (c) Magnon dispersion relationships in YIG of thickness 5 $\mu m$ for different external magnetic fields for $\pm k \parallel \mathbf{H}$ calculated using Eq. (S1).

In order to estimate the value of $M_s$ for our sample, we performed broadband spin wave spectroscopy measurements with our YIG sample placed on a co-planar waveguide (CPW). $S_{11}$ spectra were measured by the VNA for various magnetic fields. We extracted the ferromagnetic resonance (FMR) peak position $\omega_0$ from each scan and plot them in Fig. S2(a). We first fitted the resonance data with the Kittel formula $\omega_0 = \mu_0 \gamma \sqrt{H(H + M_s)}$, and extracted $M_s = 1.15 \times 10^5$ A/m. Note that for $n = 0$ and $k \to 0$, Eq. (S1) reduces to the Kittel formula.

We now evaluate $H_{cr}$ and $\omega_{nk}(H_{cr})$. In Fig. S2(b), we plot half the magnon frequency $\omega_0/4\pi$ and the minimum magnon dispersion frequency $\omega_{k,\text{bottom}}$ as a function of magnetic field, calculated using Eq. S1 with $\gamma = 2\pi \times 28$ GHz/T.
and $\lambda_{\text{ex}} = 3 \times 10^{-16} \text{ m}^2$ [1]. The onset condition of three-magnon splitting is satisfied when $\omega_0/2 = \omega_k^{\text{bottom}}$, which corresponds to the crossing point in this plot. The numerical solution of Eqs. (S3) and (S4) yields

$$k_{cr} = 5.38 \text{ [rad/µm]}, \quad \mu_0 H_{cr} = 37.4 \text{ [mT]}, \quad \left. \frac{\omega_0 k (H_{cr})}{2\pi} \right|_{k \to 0} = 2.30 \text{ [GHz]},$$

which gives the coordinate value of the crossing point. The latter value of the critical frequency is quoted in the main text. We identify $\omega_0 k |_{k \to 0}$ to be the Kittel mode frequency and denote it by $\omega_0$. Finally, we plot in Fig. S2(c) the spin wave dispersion for different magnetic fields, i.e. the first anticrossing (16.8 mT), $\mu_0 H_{cr}$, and the second anticrossing (60 mT), to highlight the presence and absence of spin-wave modes with $\omega_0/2$.

### III. DATA PROCESSING METHODS

To extract the magnetization properties including the resonance position ($\omega_0$) and the linewidth ($\Delta \omega_0/2\pi$), the measured microwave absorption spectra $S_{11}$ (dB) was first translated into a linear scale to obtain $|S_{11}| = 10^{S_{11}/10}$. Subsequently, the converted data was fit using a double Lorentzian profile:

$$|S_{11}| \propto \frac{A_1 \Delta \omega_1}{(\omega - \omega_1)^2 + \Delta \omega_1^2} + \frac{A_2 \Delta \omega_2}{(\omega - \omega_2)^2 + \Delta \omega_2^2} + C \quad (S6)$$

where $\omega$, $\omega_n$, $A_n$, $\Delta \omega_n$, and $C$ corresponds to the frequency, resonance position, relative amplitude, linewidth, and the spectra offset, respectively. Here, $n$ denotes the first and the secondary peaks.

In Figs. S3(a) and (b), we respectively show the individual fitting results of the fields used in the main text at -20 dBm, and for different microwave powers at the anticrossing field (16.8 mT).

![Figure S3. Linear microwave absorption spectra ($|S_{11}|$) as a function of frequency (from 1.0 to 2.2 GHz) (a) at various magnetic fields at a fixed microwave power (-20 dBm) and (b) for different microwave powers at a fixed magnetic field at the anticrossing point (16.8 mT). Red curves are the fits by Eq. (S6).](image-url)
IV. DETAILS OF THE THEORETICAL CALCULATIONS

In this section, we describe the theory that leads to Eqs. (3) - (5) and Figs. 4(a) and 4(b) in the main text. For convenience of the reader, we repeat the model Hamiltonian

\[ H = \omega_0 b_0^\dagger b_0 + \sum_{k \neq 0} \omega_k b_k^\dagger b_k + \omega_r b_r^\dagger b_r + \left[ g b_0^\dagger b_r + h e^{-i\omega t} \left( U_0 b_0^\dagger + U_r b_r^\dagger \right) + \text{h.c.} \right] 
+ \frac{1}{2} \sum_{k \neq 0} \left( V_k b_0 b_{-k}^\dagger + \text{h.c.} \right) + \frac{1}{2} \sum_{k, k'} S_{kk'} b_k^\dagger b_{-k'} b_{k'}^\dagger b_{-k} . \]  

(S7)

The first line describes the usual linear theory of Kittel mode in a microwave cavity augmented by the single particle energy of propagating magnons. The first term in the second line represents the three-magnon interaction arising from the magnetic dipole-dipole interactions, where the coupling constant in a sufficiently thick film is given by

\[ V_k = \omega_M \sqrt{\frac{g_{eff} \mu_B}{2v M_s}} \left( 1 + \frac{\omega_k}{A_k + |B_k|} \right) \frac{k_x(k_y + ik_z)}{k^2} , \quad \omega_M = \mu_0 \gamma M_s, \]  

(S8)

with \( g_{eff} \) the g-factor, \( \mu_B \) the Bohr magneton, \( V \) the volume of the magnet, \( k^2 = k_x^2 + k_y^2 + k_z^2 \), and \( A_k, B_k \) the Bogoliubov coefficients for the magnon eigenstates

\[ A_k = \mu_0 \gamma \left( H + M_s \lambda_{xx} k^2 + M_s \lambda_{xy} k_y^2 + M_s \lambda_{xz} k_z^2 \right) , \quad B_k = \mu_0 \gamma M_s \frac{(k_y + ik_z)^2}{2k^2} . \]  

(S9)

with which the eigenfrequency can be written as \( \omega_k = \sqrt{A_k^2 - B_k^2} \). This expression could be significantly modified for film thicknesses of a few micrometers, but the detailed form is uncomputable as well as unnecessary. We only assume \( V_{-k} = V_k \), which is expected unless the sample shape breaks the inversion symmetry.

We set the \( k \)-axis along the external static magnetic field \( H \), the \( z \)-axis along the film normal, and ignore the crystalline anisotropy so that the ground state magnetization is in the \( x \) direction. The Heisenberg Hamiltonian gives rise to many other nonlinear interaction terms between different spin waves, which were all neglected in the main text. Most of them can be removed by a suitable nonlinear transformation of \( b_k \) [3]. The only resonant contributions up to 4th order in \( b_k \), which cannot be dismissed in this way and are relevant under spatially uniform driving, are the two terms in the second line in Eq. (S7). The expression for \( S_{kk'} \) is complicated [4], but its generic order of magnitude estimate is \( S_{kk'} \approx V_k \) in the GHz range. When \( V_k \) and \( S_{kk'} \) are of similar orders of magnitude, the former usually dominates, which is what we assume. We remark, however, that there could be situations where \( S_{kk'} \) cannot be ignored even qualitatively, depending on the sample geometry and quality, and other factors such as frequency and wavelength.

Here we \textit{a posteriori} justify discarding \( S_{kk} \) by the good qualitative agreement wit the experimental data.

In our convention, \( H \) carries the unit of frequency and the Heisenberg equation of motion reads \( dA/dt = i[H, A] \), which yields

\[ i \left( \frac{d}{dt} + \eta_r \right) b_r = \omega_r b_r + g b_0 + h U_r e^{-i\omega t} + \xi_r , \]  

(S10)

\[ i \left( \frac{d}{dt} + \eta_0 \right) b_0 = \omega_0 b_0 + g b_r + h U_0 e^{-i\omega t} + \frac{1}{2} \sum_k V_k b_k b_{-k} + \xi_0 , \]  

(S11)

\[ i \left( \frac{d}{dt} + \eta_k \right) b_k = \omega_k b_k + \left( V_k b_0 + \sum_{k'} S_{kk'} b_{k'} b_{-k'} \right) b_{-k} + \xi_k . \]  

(S12)

In writing them down, we have made a few important alterations to the setup. First of all, the quantum mechanical operators \( b_{r,0,k} \) have all been relegated to stochastic random variables, and accordingly the Hermitian conjugation \( b^\dagger \) has been replaced by the complex conjugation \( \bar{b} \). It is justifiable at room temperature where quantum fluctuations are overwhelmed by the thermal fluctuations. Second, we have introduced phenomenologically the relaxation rates \( \eta_{r,0,k} \) and added them to the time derivatives on the right-hand-sides. Corresponding to the relaxation is the random noise fields \( \xi_{r,0,k} \) that have vanishing average \( \langle \xi_{r,0,k} \rangle = 0 \). We assume their higher-order correlations are Gaussian and dictated by the fluctuation-dissipation theorem for \( g = V_k = S_{kk'} = 0 \) for simplicity:

\[ \langle \xi_k(t) \xi_{k'}(t') \rangle = 2\eta_k N_k \delta_{kk'} \delta(t-t') , \quad N_k = \frac{k_B T}{\hbar \omega_k} . \]  

(S13)
and similarly for \( \langle \xi_0 \xi_0 \rangle \), \( \langle \xi_r \xi_r \rangle \) with all the other correlations being zero. The number of magnons \( N_k \) is assumed to obey Rayleigh-Jeans distribution in the high-temperature approximation.

Experimentally, we measure the microwave reflection which is ideally equal to 1 minus the appropriately normalized work per unit time done by the external driving \( h \). In order to calculate it, we only need the expectation values of \( b_{r,0} \) that oscillate at the driving frequency \( \omega \). Therefore, we follow Suhl [5] and introduce the rotating frame variables

\[
\frac{c_{r,0}}{} = (h_{r,0}) e^{-i\omega t}.
\]

For the traveling magnons, it turns out that the convenient variables are quadratic correlators

\[
s_k = \langle b_k b_{-k} \rangle, \quad n_k = \left\langle |b_k|^2 \right\rangle.
\]

Foreseeing the subsequent development, let us define the critical amplitude for the Kittel mode by

\[
c_{cr} = \frac{1}{V_k} \left( \omega_k - \frac{\omega}{2} + i\eta_k \right).
\]

It turns out that for each given \( \omega \), the modes with \( k \) that minimizes \( |c_{cr}| \) become unstable first as the input power (i.e. \(|h|\)) is increased. Unless \( h \) gets too large beyond a threshold, these remain the only modes that pick up a non-vanishing \( s_k \). This conclusion is reached by studying a zero-temperature formulation which is not a straightforward low-temperature limit of the nonzero-temperature version used here, so that we do not present the details. The reader is referred to Ref. [4]. We just assume that there is only a single pair \( \pm k \) of wavevectors that minimizes \( |c_{cr}| \) and drop the summation over \( k \). This assumption is reasonable when the symmetry of the system is low, but cannot be fully justified due to remaining discrete symmetries, accidental degeneracies, \( h \) possibly being significantly above the threshold etc. We take it as a working hypothesis, supported again by the good agreement with the experimental data. We try and find a stationary state, i.e. a non-trivial time-independent solution for \( c_{r,0}, s_k, n_k \). After a straightforward algebra, ignoring the 4-magnon contribution, one obtains

\[
\left\{ \omega - \omega_0 + i\eta_0 - \frac{|g|^2}{\omega - \omega_r + i\eta_r} + \frac{V_k N_k c_{cr}}{|c_{cr}|^2 - |c_0|^2} \right\} c_0 = \left( U_0 + \frac{g U_r}{\omega - \omega_r + i\eta_r} \right) h,
\]

along with Eqs. (4) and (5) in the main text. It further reduces to a real cubic equation for \( |c_0|^2 \), but the closed-form solution is not very illuminating. We ultimately resorted to numerical solutions, which were used to generate Figs. 4(a) and 4(b) in the main text. However, one can guess the qualitative behavior of the solution as a function of \( h \) by inspection. For \( h \to 0 \), there is clearly only one positive root for \( |c_0|^2 \propto |h|^2 \) that represents the usual FMR solution. It remains a good approximation as long as \( |c_0|^2 \ll |c_{cr}|^2 \). When the latter condition ceases to be valid, the third-term on the left-hand-side starts growing. Note that this term grows indefinitely as \( |c_0| \) approaches \( |c_{cr}| \) from below. Therefore, however large \( |h| \) becomes on the right-hand-side, there is always a solution \( |c_0| \) that is close to but no greater than \( |c_{cr}| \). It is easy to convince oneself that this solution continuously evolves out of the usual FMR solution at low power. Thus we are led to conclude that the Kittel mode amplitude saturates at \( |c_{cr}| \) in the high-power limit. It also tells that at nonzero temperatures, the instability is not a clear threshold process, but it occurs continuously as the input power is increased. The phase information can also be read off from Eq. (S17) and Eq. (4) in the main text, which yields Eq. (3) in the main text.

We have so far been ambiguous about the driving field amplitude \( h \), and it has to be specified for simulations. Here by convention \( h \) is taken to have the dimension of frequency, which means \( U_{r,0} \) are dimensionless. The threshold value of \( h \) for zero power is a mess in the general setup. To provide a theoretically convenient normalization, we introduce the threshold field \( h_{cr} \) defined for \( g = 0 \):

\[
h_{cr} = \frac{\omega - \omega_0 + i\eta_0}{U_0} \frac{\omega/2 - \omega_k + i\eta_k}{V_k}.
\]

It gives an order of magnitude estimate for the relevant input power range, but we do not attach a direct physical meaning to it since the absolute value of \( U_0 \) is not easily obtainable from the experimental data. The ratio \( U_0/U_r \) affects the lineshape in an essential way, however. After experimenting with different values, we fixed \( U_0/U_r = -0.3i \) which gave a reasonable comparison with Fig. 3(c) in the main text.

To compare the theory with the experimentally measured \( S_{11} = -10 \log_{10} R \), we need an estimate for the reflection coefficient \( R \) that is the reflected power divided by the input power. Since there is no transmission line in our device, the reflected power is equal to the input power \( P_{in} \) minus the power dissipated \( P_d \) in the device, namely \( R = 1 - P_d/P_{in} \). By definition \( 0 < R < 1 \) and therefore \( S_{11} < 0 \). The experimental data suggest that \( R \approx 1 \) away from the resonance.
and $R \approx 0$ at the peak of the resonance. The former implies that the loss from dynamical entities other than the magnons and photons is very small so that one can reasonably estimate $P_d$ by the dissipation rate of the magnons and photons:

$$P_d^{\text{theoretical}} \sim \eta_0 |c_0|^2 + \eta_r |c_r|^2 + 2\eta_k n_k.$$  \hfill (S19)

All the quantities on the right-hand-side can be readily computed as functions of $H, \omega$ and $h$ once the model parameters are fixed. To compare the calculation with the experiment, however, we need an expression for $P_d$ in, which is difficult to model theoretically. Here we take a phenomenological approach by observing that in the experimental data the minimum value of $S_{11}$ is roughly -30 dB regardless of the input power. We choose $P_{\text{in}}$ for each $h$ such that the computed minimum for $S_{11}$ is equal to -30 dB:

$$S_{11}^{\text{theoretical}} = -10 \log_{10} \left( 1 - \frac{(1 - 10^{-3}) P_d^{\text{theoretical}}}{\max_{H,\omega} P_d^{\text{theoretical}}} \right).$$  \hfill (S20)

As explained after Eq. (S16), the wave vector $k$ of the magnon pair that become unstable is determined by minimizing $|c_{cr}|$ for each $\omega$. Therefore, in computing the $\omega$-dependence of $P_d^{\text{theoretical}}$, one in principle has to treat $k$ as a function of $\omega$. To do it, one would need to know the $k$-dependence of $V_k$ and $\eta_k$. However, the former could significantly differ from the bulk expression (S8) for our 5 $\mu$m thickness film while it is practically impossible to model the $k$-dependence of $\eta_k$ in any systematic way. Since we do not aim at being quantitative, we set both $V_k$ and $\eta_k$ to be independent of $k$ and fix their values as

$$V_k = \omega M \sqrt{\frac{2g_{\text{eff}} \mu_B}{VM_s}}, \quad \eta_k = \alpha \frac{\omega_0}{2},$$  \hfill (S21)

where the magnon Gilbert damping $\alpha = 0.01$ has been introduced and the sample volume is taken to be

$$V = \frac{2g_{\text{eff}} \mu_B}{M_s} \times 10^{11}.$$  \hfill (S22)

Note that the prefactor on the right-hand-side would equal twice the volume of the unit cell if the magnet was a simple ferromagnet. With $V_k$ and $\eta_k$ being constants, the minimum of $|c_{cr}|$ occurs always at $\omega_k = \omega/2$. For Kittel mode, we decided to use a constant damping $\eta_0$ independent of $\omega$ instead of the Gilbert type as usually assumed, since the FMR linewidth away from the photon-magnon hybridization appears very weakly dependent of $\omega$, indicating that the disorder-induced two-magnon contribution dominates $\eta_0$. Finally, the reference driving field $h_{\text{ref}}$ has been taken equal to $h_{cr}$ evaluated at $\omega = \omega_0$ and $\mu_0 H = 15$ mT.

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