Strong coupling of magnetization waves with sub-THz cavity fields

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Strong coupling of electromagnetic cavity fields with antiferromagnetic spin waves in hematite (\(\alpha\)-Fe\(_2\)O\(_3\)) was achieved above room temperature. A cube of hematite was placed in a metallic tube and transmission was measured, using a continuous-wave THz spectrometer. Spectra, collected as a function of temperature, reveal the formation of magnetic polaritons.

The hybrid nature of strongly coupled light-matter states, were the dissipation rate is lower than the exchange rate (Rabi frequency) \([1]\), has attracted a lot of attention since the late 1980s \([2]\). In the 2000s decade, strong light-matter coupling was investigated in the solid state \([3]\), where the coupling of light interacting with \(N\) resonators increases by a factor of \(\sqrt{N}\) \([4]\). This regime might lead to device elements that may play a role in quantum devices \([1] [5] [9]\). In the 2010s, interest turned toward magnon-photon coupling in ferromagnetic materials \([10] [15]\), as spins benefit from a relatively low coupling to their environment \([6] [19]\).\n
Achieving strong coupling with antiferromagnets has the advantage of operating at frequencies in the terahertz (THz) range. There is a large variety of materials available, and many of them show magnetic ordering above room temperature. The absence of stray fields could in principle allow for extremely dense packing of elements \([20]\). However, there are only a few examples of weak magnon-photon coupling in antiferromagnets \([21] [22]\). Strong coupling was only achieved at low frequencies \([23]\), in bulk samples \([24]\) or via an indirect coupling \([25] [29]\), while direct strong cavity-magnon coupling was not shown until now \([27]\).

In this letter, we show strong coupling between electromagnetic fields of a 3-dimensional cavity and spin waves in hematite (\(\alpha\)-Fe\(_2\)O\(_3\)), a very common room-temperature antiferromagnet. Hematite crystallizes in approximately hexagonal structure with space group R\(3c\). Precise measurements show that the actual symmetry is monoclinic C2/c \([28]\). Below the Neél temperature \(T_N \approx 955\) K \([29]\), the Fe\(^{3+}\) magnetic moments orient in an antiferromagnetic order. Above the spin-reorientation transition (Morin phase transition) at about \(T_M \approx 260\) K \([30]\), the superexchange Dzialoshinskii-Moriya interaction leads to a canting of the two sublattices that gives rise to a net magnetization \(\mathbf{m}\), i.e. making this material a weak ferromagnet. Owing to the spin canting, the antiferromagnetic resonance has two modes, one of which is at a high frequency. This quasi-antiferromagnetic resonance (qAFMR) mode is excited by dynamical magnetic field \(\mathbf{h}\) parallel to the magnetization (\(\mathbf{h} \parallel \mathbf{m}\)). At temperatures above room temperature the width of qAFMR is about 1 GHz only, and its frequency rises sharply with temperature \([30]\).

Thanks to the development of frequency extenders for vector network analyzers (VNA), continuous-wave spectroscopic measurements up to 1.5 THz can be rapidly conducted with a high frequency resolution and with a very high dynamic range \([22] [31] [33]\).

Out of a bulk natural single crystal, we cut a cube of \(l_s = 0.2\) mm edge length, with faces in the \(a\), \(b\) and \(c\) crystalline directions. The cube was placed inside a tube of \(l_c = 2\) mm length and \(2\pi r_c = 0.58\) mm internal diameter, cut out of a gold-plated stainless steel needle. The needle was wrapped with Teflon tape to prevent the cube from dropping out of the tube. The tube with the cube in it was inserted in a copper holder composed of two cones (cone angle, about 60°) which focused the THz beam into the cavity (Fig. 1). The THz beam propagated between the source, sample holder and detector in oversized metallic waveguides, 8 mm in diameter, matching the larger diameter of the cones. The holder was placed between Peltier elements that controlled its temperature, which was monitored with a K-type thermocouple inserted in a hole on its side. The detector measured the transmitted power and phase of the THz electric field. Temperature scans started from the highest temperature with a step of \(\Delta T = -0.25\) K. This temperature step was...
so chosen that the frequency change was smaller than the line width of the resonance. After stabilizing the sample temperature \( T \), we measured transmission as a function of radiation frequency \( f \).

The source emits linearly polarized beam of THz radiation and the detector detects only radiation of linear polarization that matches its waveguide, i.e. it acts like a polarization filter. Rotation of the polarization plane is possible by rotating source and detector about the optical axis. We define the polarization angle \( \delta \) as the angle of the radiation \( \hbar \) field with respect to the plane of the optical table (Fig. 1). We measured transmission in a polarization angle range of \( \delta = -90^\circ \) to \( +90^\circ \).

In Fig. 2, we can see avoided crossing of the cavity mode at \( f_1 \approx 242.5 \) GHz with the qAFMR mode, the frequency of which rises approximately linearly with temperature. This result was obtained at polarization angle \( \delta = 30^\circ \). We fitted the observed spectra using the equation developed in the framework of input-output theory [10, 34]:

\[
S_{21} = 1 + \frac{a_1}{i(f - f_1(T)) - \frac{a_1}{2} + \frac{c_1^2}{2(f - f_1(T))^2 - g/2}},
\]

where, \( a_1 = (0.32 - i0.02) \) GHz is a complex parameter describing the coupling of the cavity with the source and the detector. The parameter \( f_1(T) \approx 242.5 \) GHz is the observed frequency of the 1st cavity mode and \( a_1 = 2.1 \) GHz describes its width, \( 2G_1 = 6.2 \) GHz is the minimum splitting between upper and lower polariton branches, \( f_r(T) \) is the qAFMR resonance frequency and \( g = 0.5 \) GHz describes its width. With these parameters, we can determine the cooperativity factor

\[
C = \frac{4G_1^2}{\kappa_1 g} \approx 40,
\]

that is, the square of a ratio of the Rabi splitting \( (2G_1) \) and the losses of the polariton states. This means that the splitting was about 6 times larger than the linewidth. According to the harmonic coupling model, the upper and lower polariton frequencies are given by [12, 35],

\[
f_{\pm} = \frac{1}{2} \left( f_1 + f_r \pm \sqrt{(f_1 - f_r)^2 + 4q_1^2f_1} \right).
\]

The coupled mode frequencies \( f_{pm} \) are drawn on Fig. 2 as green dotted lines, with \( 2g_1\sqrt{f_1} = 2G_1 = 6.2 \) GHz, as well as the non-interacting mode frequencies \( f_1(T) \) and \( f_r(T) \). Clearly, this prediction matches that of Eq. 1 (Fig. 2b). The discrepancies between fits using Eq. 1 and data (Fig. 2a) may arise from interactions with other cavity modes, as we discuss in further sections of this communication.

The coupling strength can be calculated using a microscopic model [12]

\[
g_i = \frac{g_i \mu_B}{2h} \sqrt{\frac{\mu_0}{2} \frac{V_c}{V_c}},
\]

where, \( g_i = 2, \rho \) is density of resonators in the hematite cube of a volume \( V_c = l_s^3 \) and \( V_c \), the volume of the metal tube, \( V_c = \pi r_s^2 l_s \approx 0.528 \) mm$^3$. In Eq. 4 each magnon is coupled to the electromagnetic cavity mode with a coupling strength \( g_i \mu_B B_0/2h \) [36], where \( B_0 = \sqrt{\mu_0 \hbar f_1/2V_c} \approx 4.4 \times 10^{-10} \) T is the magnetic component of vacuum fluctuations [37] (here \( f_1 = 242.5 \) GHz). This is a few orders of magnitude smaller than the amplitude of the THz field in our experiment. The collective coupling strength of \( N = \rho V_c \) oscillators is increased by a factor \( \sqrt{N} \) [12, 13, 25, 36], thus \( g_i \) depends only on the oscillators density \( \rho \) and the ratio of the crystal volume to the cavity volume [12, 25], that is \( V_c/V_c \approx 1.51 \times 10^{-2} \).

The density of iron atoms in hematite is \( \rho = 3.987 \times 10^{28} \text{ m}^{-3} \) [38]. This value is quite high compared to that of many common antiferromagnets. Hence, hematite is a good material to achieve strong coupling. Taking this into account gives \( g_i = 7.01 \times 10^{27} \text{ Hz}^{1/2} \), i.e. the splitting is predicted to be \( 2G_1 = 2g_i \sqrt{f_1} \approx 6.9 \) GHz for the cavity mode at \( f_1 = 242.5 \) GHz. The observed splitting \( 2G_1 = 6.2 \) GHz is about 90\% of that value. This slight discrepancy may be due to an imperfect matching
of the electromagnetic mode with the antiferromagnetic resonance in the hematite cube, i.e. the cavity magnetic field in the volume of the cube does not excite all the spins with equal amplitude (as confirmed in the simulation discussed below, Fig. 3). We calculated that in the 1st mode, indeed the magnetic field component along the z axis makes about 85% of all the integrated magnetic field strength in the cube volume (Tab. I).

We found that the parameter $a_1$ strongly depends on the polarization angle $\delta$ (Fig. 3a). This parameter describes the coupling between the cavity and the rest of the spectrometer, i.e. constructive or destructive interferences between the cavity and the experimental setup. The result presented in Fig. 3a, obtained at $\delta = 30^\circ$, shows a minimum in transmission that is accounted for with $a_1$ having a small imaginary part and a positive real part. The dependence of $a_1$ on polarization angle (Fig. 3b) is not an intrinsic property of the cavity, but depends on the coupling of the cavity with the experimental setup. As a consequence, after it was rearranged, we obtained a slightly different value for $a_1$ (Fig. 3b). Thus, the result obtained at $\delta = -75^\circ$ (Fig. 3c) under (a) conditions shows a dispersive lineshape that is accounted for by a negative imaginary $a_1$ (Fig. 3c). In contrast, the result obtained at $\delta = -60^\circ$ (Fig. 3d) under (b) conditions shows constructive interference that is explained by an almost entirely real, negative $a_1$ (Fig. 3d). Under the same conditions, at $\delta = 70^\circ$ a weak inverted dispersive lineshape was observed (Fig. 3e) that is account for with a small positive imaginary $a_1$.

The raw data for spectra are dominated by multiple interferences that are not strongly interacting with the cavity and are weakly temperature-dependent. We normalized both the measured spectra and fitted functions to a base frequency that is temperature-dependent and passes though the middle of the interaction. This way, we reduced artifacts at the cavity mode that would occur if instead, we normalized to the first recorded spectrum. We could eliminate this interference background by calculating temperature-derivative spectra, that is subtracting from one another successive spectra, thus having amplitude derivatives:

$$
\frac{d|S_{21}|}{dT}[dB] = \frac{20}{\Delta T} \log_{10} \left| \frac{|S_{21}(f, T + \Delta T)|}{|S_{21}(f, T)|} \right|, 
$$

and phase derivatives:

$$
\frac{d(\text{arg} S_{21})}{dT} = \frac{\text{arg} S_{21}(f, T + \Delta T) - \text{arg} S_{21}(f, T)}{\Delta T}. 
$$

The advantage of looking at temperature-differential spectra is that it is possible to see small features, like a weak middle line passing through the interaction region (Fig. 4ac) that is almost invisible in normalized spectra. This middle line can be qualitatively explained by taking into account the 2nd cavity mode at $f_2 = 258.4$ GHz. We can take it into account by assuming that both modes interact independently with the magnetic resonance mode:

$$
S_{21} = 1 + \sum_{j=1}^{2} \frac{a_j}{i(f - f_j(T)) - \frac{\alpha_j^2}{2} + \frac{\kappa_j^2}{2(f - f_j(T))^2}}, 
$$

FIG. 3. (a) Angular dependence of $a_1$. (b) Angular dependence of $a_1$ under different experimental conditions. (c) Normalized $|S_{21}|$ obtained at $\delta = -75^\circ$ under (a) conditions. (d) fit of $a_1$. (e) Result at $\delta = -60^\circ$ under (b) conditions. (f) fit. (g) Result at $\delta = 70^\circ$ under (b) conditions. (h) fit.

FIG. 4. (a) Temperature-differential magnitude spectra of the cavity. (b) Temperature-differential phase spectra of the cavity.
FIG. 5. (a) To a scale model of the cavity with a dielectric cube in the middle. Arrows show magnetic field of the 1st cavity mode at 247 GHz on x and y planes that intersect the cube. Dependence of the modes frequencies on (b) displacement of the cube from the center of the tube, (c) rotation of the cube about y axis, (d) length of the tube, (e) diameter of the tube, (f) cube edge length, (g) dielectric constant of a cube. In (d)(e)(f) and (g) vertical lines mark expected values of these parameters.

In Fig. 6 we present temperature-differential of Eq. 7, where we put \( \alpha_2 = (4.5 - i1.1) \) GHz, \( G_2 = 0.56 \) GHz and \( \kappa_2 = 14.7 \) GHz. These values are consistent with our idea of a weak interaction with the 2nd cavity mode that has a lower quality factor than the 1st mode. This simple model is not fully sufficient above 250 GHz. We think that a more precise estimate might require taking into account other weakly interacting cavity modes or interaction between cavity modes that are mediated by the magnetic resonance. We found that the observed phase temperature-differential spectra (Fig. 4b) are reproduced by the same set of parameters (Fig. 4f), using argument of Eq. 7 and Eq. 8.

We identified the cavity modes using a numerical electrodynamics field simulation software (CST Microwave Studio). We modeled the cavity as a metallic cylinder containing an isotropic dielectric cube (Fig. 5a). We assumed that the dielectric constant of hematite was 19.1 at 350 K, as determined from our measurements on bulk samples [39]. We calculated the dependence of modes on the size and position of the cube inside the cylinder. The size of the cube was limited by our cutting process. At this smallest size, the mode frequencies are governed mostly by the size of the cube \( l_s \) (Fig. 5f) and, to a smaller extend, by the diameter of the tube \( 2r_c \) (Fig. 5g). The length of the tube \( l_c \) is irrelevant (Fig. 5d). For a given size cube, we chose a diameter of the tube such that the 1st and 2nd modes are well split (Fig. 5e). We found, that a random position of the cube in the needle spans a mode frequency range of at most 15 GHz (Fig. 5c). In other calculations, the cube was assumed to be in the center of the needle \( (z_{off} = 0) \) and straight along it \( (\beta = 0) \). The calculated frequency of the cavity is a few GHz higher than the observed one. This might be due to inaccuracy in the cube dimensions.

We give in Tab. 1 the expected frequencies of the first five modes and dominant directions of the electric field in the cavity, as well as the dominant direction of the magnetic field in the cube. Only the 5th mode has a similar symmetry to that of the 1st mode, and is able to excite antiferromagnetic resonance in the sample. This suggest that the magnetization vector \( \mathbf{m} \) in the cube was aligned along the z axis of the cavity during the experiment. Under this assumption, modes 2–4 do not excite
the magnetic resonance and thus, should only weakly hybridize with it. This agrees with our observation in Fig. 6b, that only the 5th mode produces some strong coupling, which is not as coherent as the coupling to the 1st mode. This may be due to a more complex distribution of the cavity magnetic field in the cube, that is the ratio of $h_z/h_x$ ≈ 0.53 is lower than in the case of the 1st mode (Tab. [I]). This means that some of the spins in the cube are excited out of phase, thus allowing excitation of magnetostatic modes.

Fig. 6b shows the observed temperature dependence of the 1st mode, that we determined as $f_1(T) = f_1^{345} + f_1^T(T - 345 \text{ K})$, were $f_1^{345} = 243 \text{ GHz}$ and $f_1^T = -2.1 \times 10^{-2} \text{ GHz/K}$. This dependence is caused by the dielectric constant of hematite increasing with rising temperature. The dependence on cube dielectric constant is predicted to be small (Fig. 5g).

In conclusion, we observed strong coupling between the quasi-antiferromagnetic resonance (qAFMR) of a hematite cube ($\alpha$-Fe$_2$O$_3$) located in a cavity near room temperature. The sample was cut from a single crystal in the shape of a cube, 0.2 mm on a side. The cavity was a gold-plated cylinder, 2 mm in length and about 0.6 mm in diameter. The avoided crossing, which is characteristic of polariton dynamics, occurred near 243 GHz. The cooperativity $C$, which is a measure of the coupling strength with respect to both the magnetic resonance and the cavity line widths (Eq. 2), was estimated at 40.

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### TABLE I. Predicted modes frequencies, selection rules and mean magnetic field in the cube.

| \(j\) | \(f_j\) [GHz] | \(c\)-field | \(h_x\) | \(h_y\) | \(h_z\) | \(|h_x|\) | \(|h_y|\) | \(|h_z|\) | \(|h|\) |
|------|----------------|------------|--------|--------|--------|--------|--------|--------|------|
| 1    | 247            | \(x\)      | 0.01   | -0.01  | 2.29   | 0.42   | 0.66   | 2.39   | 2.70 |
| 2    | 258            | \(y\)      | 0.12   | 0.00   | 0.00   | 0.96   | 0.09   | 1.30   | 1.76 |
| 3    | 272            | \(z\)      | 2.51   | 0.00   | -0.01  | 2.61   | 0.75   | 0.63   | 3.05 |
| 4    | 297            | -          | 0.00   | 2.30   | 0.01   | 0.72   | 2.51   | 0.90   | 3.15 |
| 5    | 329            | \(x\)      | 0.00   | 0.00   | -1.00  | 0.54   | 0.43   | 1.87   | 2.19 |

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