Antiferromagnetic resonance in $\alpha$-Fe$_2$O$_3$ up to its Néel temperature

M. Bialek,$^1$ J. Zhang,$^2$ H. Yu,$^{1,3}$ and J.-Ph. Ansermet$^{1,3}$

$^1$Institute of Physics, École Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland
$^2$Fert Beijing Institute, MIT $^*$ Key Laboratory of Spintronics, School of Integrated Circuit Science and Engineering, Beihang University, Beijing, China
$^3$International Quantum Academy, Shenzhen 518048, China

(Dated: 12 July 2022)

Hematite ($\alpha$-Fe$_2$O$_3$) is an antiferromagnetic material with a very low spin damping and high Néel temperature. The temperature dependence of the antiferromagnetic resonance in a bulk single crystal of hematite was characterized from room temperature up to the Néel temperature in the frequency range of 0.19–0.5 THz. From these data, the Néel temperature was estimated as 966 K.

Hematite ($\alpha$-Fe$_2$O$_3$) is a very common room-temperature antiferromagnet of the Néel temperature, reported as $T_N = 946$ K$^{[12]}$ $T_N = 950$ K$^{[2]}$ $953$ K$^{[3]}$ $960$ K$^{[4]}$ $972$ K$^{[4]}$. It is characterized by a very low spin damping that make it promising for spintronics, and strong light-mater coupling.$^{[6,11]}$ Above the spin-reorientation transition (Morin phase transition) at about $T_M = 260$ K$^{[21]}$ the superexchange Dzialoshinskii-Moriya interaction leads to canting of the two sublattices that gives rise to net magnetisation $\mathbf{m}$, i.e. making this material a weak ferromagnet.

Hematite crystallizes in an approximately hexagonal structure with space group R3c. Precise measurements show that the actual symmetry is monoclinic C2/c.$^{[13]}$ In the weak ferromagnetic state the ferromagnetic moment $\mathbf{m}$ is along $a$ axis$^{[16,17]}$ (magnetic symmetry C2'/c') or $b$ axis$^{[20]}$ (magnetic symmetry C2/c$^{[18]}$). Owing to the spin canting, the antiferromagnetic resonance has two modes: at a higher frequency, the quasi-antiferromagnetic resonance (qAFMR) mode which is excited when the dynamical magnetic field $\mathbf{h}$ is parallel to the magnetization ($\mathbf{h} \parallel \mathbf{m}$), and at low frequencies, the quasi-antiferromagnetic resonance (qFMR) mode, excited by a dynamical magnetic field perpendicular to the magnetization ($\mathbf{h} \perp \mathbf{m}$). In this work, we characterize the qAFMR mode up to the Néel temperature, which was observed previously up to about 600 K$^{[2]}$ and below room temperature.$^{[44]}

The qFMR mode has non-zero frequency only with external field applied$^{[19,49]}$ and falls below our experimental range. To our knowledge, the qFMR mode was investigated only at room temperature and below$^{[20,22]}$.

Development of frequency extenders for vector network analyzers (VNA) allows continuous-wave spectroscopic measurements up to 1.5 THz to be conducted with a high frequency resolution and with a very high dynamic range.$^{[12,23,26]}$. Our sample was a natural single crystal of $\alpha$-Fe$_2$O$_3$ of $d = 0.5$ mm in thickness and $10 \times 10$ mm$^2$ in lateral dimensions. The normal to the sample surface was (11-20).

---

$^a$Electronic mail: marcin.bialek@fuw.edu.pl

![Schematic of the THz spectrometer](image-url)
between components of the experimental setup. Since this pattern very weakly depends on temperature, we eliminated it by calculating temperature-derivative spectra, that is, by subtracting successive spectra from one another. We calculated magnitude temperature-derivative spectra

\[
\frac{d|S_{21}|}{dT} = \frac{|S_{21}(f, T + \Delta T)| - |S_{21}(f, T)|}{\Delta T}, \tag{1}
\]

and phase derivative:

\[
\frac{d\arg(S_{21})}{dT} = \frac{\arg(S_{21}(f, T + \Delta T)) - \arg(S_{21}(f, T))}{\Delta T}. \tag{2}
\]

Our main results are presented in Fig. 2a (magnitude) and Fig. 2c (phase). They were obtained with the THz magnetic field acting in the basal plane of the crystal, as determined from polarization-dependent results in Fig. 3. At a perpendicular polarization, with THz magnetic field along c axis, the resonance is almost not excited. We can notice the the resonance peak shows a tiny splitting of about 1 GHz. Result of angular dependence obtained with a c-cut sample shows a more complex dependence on polarization angle that is different for a higher and a lower modes of the qAFMR (green circles and blue rectangles in Fig. 3b). This angular dependence suggests that the observed splitting is related to different magnetic domains in our samples.

We observed a strong and narrow resonance (Fig. 2b and c), the frequency of which is rising with temperature at above room temperature (Fig. 2a), reaching a maximum of about 484 GHz at about 685 K and then dropping sharply as \( T_N \) is approached. The periodic, almost horizontal pattern is related to the Fabry–Pérot type cavity modes inside the sample. We can account for these temperature-differential spectra, as shown in Fig. 2 (b, d), by using an electrodynamics-based model:

\[
\frac{d|S_{21}(f, T)|}{dT} = \frac{20}{\Delta T} \log_{10} \frac{|t(f, T + \Delta T)|}{|t(f, T)|}, \tag{3}
\]

where \( t \) is the transmittance of a plane electromagnetic
wave at normal-incident on a parallel-plane slab of infinite lateral dimensions and thickness $d$

$$t = \frac{(1 - r^2)e^{ikd}}{1 - r^2e^{2ikd}}.$$  

(4)

Here, $r = (\sqrt{\epsilon} - \sqrt{\mu})/(\sqrt{\epsilon} + \sqrt{\mu})$ and $k = 2\pi f\sqrt{\mu}/c$. We assume an isotropic slab material characterized by its permeability $\mu(f, T)$ and permittivity $\epsilon(f, T)$. Our crystal is not isotropic, but in the case of incidence at a normal angle to the crystal surface, the crystal being cut in one of its principal axes, we approximate it as isotropic, that is, we assume that there is no rotation of polarization in the crystal. At other angles of incident beam polarization, we get different values of $\epsilon$. However, as long as we were interested in the behaviour of the antiferromagnetic resonance, we determined permittivity at the polarization angle where the resonance is excited the strongest. Since this occurs when the dynamical magnetic field is in the basal plane, the $\epsilon$ that we determined in this communication describes response of hematite when exposed to dynamical electric field acting in the $c$ axis.

The assumptions made for both $\mu$ and $\epsilon$ are the following. We found that our data are sensitive to a temperature dependence of the dielectric response function. The simplest model that accounts for our data is a real second order polynomial of temperature. This approximation is justified because our frequency range is far from optical phonons (the lowest at about 7.5 THz) and dielectric absorption is negligible. Thus, we assumed

$$\epsilon = \epsilon_90 + a(T - T_0) + b(T - T_0)^2,$$  

(5)

where $T_0 = 900$ K, $\epsilon_90 = 23.405$, $a = 1.139 \cdot 10^{-2} K^{-1}$, $b = 6.434 \cdot 10^{-6} K^{-2}$. To account for the magnetic resonance, we assumed that the permittivity has the form of two Lorentzian functions to account for the observed splitting of the qAFMR mode

$$\mu = 1 + \frac{\Delta \mu_1 f_l^2}{f^2 - f_l^2 - i f g_1} + \frac{\Delta \mu_2 (f_r + \Delta f)^2}{(f_r + \Delta f)^2 - f^2 - i f g_2}.$$  

(6)

Here, $\Delta \mu_1 + \Delta \mu_2 = \Delta \mu$ gives the total static magnetic susceptibility, $f_l$ is the lower qAFMR frequency, $g_1$ and $g_2$ are widths of the two split resonances and $\Delta f$ is the splitting of about 1 GHz. In the next paragraphs we describe these parameters.

In order to describe the temperature dependence of the antiferromagnetic resonance, we first fitted the resonance frequency, amplitude, and line widths in temperature intervals of 2 K around some temperature $T_0^{(l)}$, where $l$ is the interval number. Within each such interval, we put amplitudes and widths independent of temperature and we fit the resonance frequency with a first order polynomial $f(T)^{(l)} = f_{T_0}^{(l)} + f_{T_0}^{(l)}(T - T_0)$. Results of these temperature interval fittings are shown as green crosses in Fig. 2(a,b,c). We show in Fig. 2(a) middle frequency $f_{T_0}^{(l)}$ obtained in given temperature interval.

These results allowed us to assume some simple functional dependences that are approximately valid in the entire experimental temperature range. The global fitting result, using the above assumptions, are displayed in Fig. 2(b) and (d). The fit parameter values are given in Tab. 1. The functions used to describe modes frequencies, amplitudes and widths are drawn as solid yellow lines in Fig. 2.
To describe the observed temperature dependence of the resonant frequency (Fig. 4a), we took the dependence of the resonant frequency to follow a modified power law

$$f_r(T) = \sqrt[3]{\sum_{j=2}^{j=5} a_j T^j \left(1 - \frac{T}{T_N}\right)^\beta},$$

(7)

where $T_N$ is a parameter close to the Néel temperature and the power factor $\beta$ is expected to be about $1/2$. We modified the classical power law adding a 5-th order polynomial that lets us heuristically describe its non-trivial dependence on temperature. This non-monotonic dependence of resonance frequency on temperature is caused by the strong temperature dependence of the magnetic anisotropy field $\Delta T$ that governs the frequency of this mode. It is related to the spin-reorientation transition at about 260 K, with one of its effects being that the qAFMR mode frequency has a minimum at this temperature. The fit value of $T_N \approx 966$ K is within the spread of literature values of $T_N = (946 - 972)$ K. These inconsistencies in the literature might stem from errors in temperature measurement in different experiments. However, it may be also due to different models used to determine $T_N$, when it is determined by extrapolation.

We assumed that the observed splitting (Fig. 4b) can be described in the following way

$$\Delta f = \delta f_r + f_r(T_{TN} = T_N + \delta T_N/2) - f_r(T_{TN} = T_N - \delta T_N/2),$$

(8)

which express our expectation that the splitting is caused by presence of two types of domains characterized by slightly different anisotropy fields (factor $\delta$) and slightly different critical temperatures (factor $\delta T_N$). The second term dominates at high temperatures. Inhomogeneous nature of this splitting is justified by the measurements for 0001 cut sample (Fig. 4b) that show that upper and lower modes have different dependence on polarization angle. The observed temperature dependence of the splitting can be well fitted (Fig. 4b) with the Eq. 8 using reasonable values of $\delta$ and $\delta T_N$ (Table I).

Taking into account the scatter in Fig. 4b, we took amplitudes of the resonance $\Delta \mu_1 = \Delta \mu_2 = \frac{1}{2} \Delta \mu$ to be independent of temperature. This scatter is probably due to imperfect interval fits, especially pronounced at around 700 K and close to $T_N$, owing to low signal obtained by temperature-differential transmission in these temperature ranges.

We assumed that the temperature dependence of the observed width (Fig. 4c) of both modes is described by the same function consisting of a sum of a constant and an exponential,

$$g(T) = g_{400} + g_e e^{\delta g_1 (T - T_N)},$$

(9)

where the latter dominates close to $T_N$. It could be due to either: 1) temperature inhomogeneity, 2) inhomogene-
We observed weak interaction of magnetic resonance with electromagnetic standing waves, as found previously\cite{ref1}. These effects are readily taken into account by a classical electrodynamic model. Light-matter interaction when the sample is its own cavity is in a weak regime, because the quality factor of modes of dielectric cavity is low. Nevertheless, these electromagnetic cavity modes already have a dramatic and nontrivial effect on the observed lineshapes, as shown in Fig. 3\textsuperscript{a} and Fig. 4\textsuperscript{a}.

In Fig. 4\textsuperscript{a} we also included the magnetic anisotropy factor, which we deduced from equation $K = \mu_0 H_A M_0$, where $H_A$ is anisotropy field and $M_0$ is a saturation magnetisation of a sublattice. To do so, we took into account that the static magnetic susceptibility is given by $\chi_\perp = M_0/2 H_E$, where $H_E$ is exchange field, and that $\chi_\perp = \Delta \mu_2/\rho_m$, where $\rho_m = 5.27 \cdot 10^3$ kg/m$^3$ is the hematite mass density\cite{ref2}. Then, using the Kittel’s equation

$$2\pi f_r = \gamma \mu_0 \sqrt{2H_E H_A} = \gamma \sqrt{\frac{K_\perp}{\chi_\perp}}, \quad (10)$$

we found the mass density of the magnetic anisotropy energy. We see in Fig. 4\textsuperscript{a} that $K$ has a complex behaviour, which is known to be caused by the spin-reorientation transition that occurs at about 260 K.

Hematite single crystals present a very narrow resonance around room temperature, about 1 GHz. The resonance frequency increases sharply above room temperature and reaches a maximum of 484 GHz at about 685 K and then, the mode softens steeply when approaching the Néel temperature $T_N$. Above 690 K, the observed width of the resonance rises exponentially with temperature. Weak light-matter coupling was observed between the qAFMR mode and the standing waves inside the slab itself. The qAFMR mode presents a splitting that was ascribed to domains in the sample.

Support by the Sino-Swiss Science and Technology Cooperation (SSSTC) grant no. EG-CN\_02\_032019 is gratefully acknowledged. The VNA and frequency extenders were funded by EPFL and the SNF R’Equip under Grant No. 206021\_144983.

The authors have no conflicts to disclose. The data that support the findings of this study are available from the corresponding author upon reasonable request.

1. L. Oravova, Z. Zhang, N. Church, R. J. Harrison, Ch. J. Howard, and M. A. Carpenter. Corrigendum: Elastic and anelastic relaxations accompanying magnetic ordering and spin-flop transitions in hematite, Fe$_2$O$_3$. Journal of Physics: Condensed Matter, 25(25):255001, may 2013.
2. K. S. Aleksandrov, L. N. Bezmaternykh, G. V. Kozlov, S. P. Lebedev, A. A. Mukhin, and A. S. Prokhorov. Anomalies of high-frequency magnetic permeability of hematite at the Morin phase transition. Journal of Experimental and Theoretical Physics, 65(3):591, 1986.
3. F. J. Morin. Magnetic Susceptibility of $\alpha$Fe$_2$O$_3$ and $\alpha$Fe$_3$O$_4$ with Added Titanium. Phys. Rev., 78:819–820, Jun 1950.
4. N. B. Nešković, B. Babić, and J. Konstantinović. High temperature anomalous behaviour of the crystal lattice of hematite. physica status solidi (a), 41(2):K133–K136, 1977.
12. M. Bialek, J. Zhang, H. Yu, and J.-Ph. Ansermet. Spin-wave coupling of antiferromagnetic resonance with subterahertz cavity fields. Phys. Rev. Applied, 15:044018, Apr 2021.

13. I. Boventer, H. T. Simensen, A. Anane, M. Kläui, A. Brataas, and R. Lebrun. Antiferromagnetic cavity magnon polaritons in collinear and canted phases of hematite, 2022.

14. Sh. G. Chou, P. E. Stutzman, Sh. Wang, E. J. Garboczi, W. F. Egelhoff, and D. F. Plusquellic. High-Resolution Terahertz Optical Absorption Study of the Antiferromagnetic Resonance Transition in Hematite (α-Fe₂O₃). The Journal of Physical Chemistry C, 116(30):16161–16166, 2012.

15. R. Przeniosło, I. Sosnowska, M. Stekiel, D. Wardecki, A. Fitch, and J. B. Jasinski. Monoclinic deformation of the crystal lattice of hematite α-Fe₂O₃. Physica B: Condensed Matter, 449:72 – 76, 2014.

16. Dzyaloshinsky. A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics. Journal of Physics and Chemistry of Solids, 4(4):241 – 255, 1958.

17. A. H. Morrish. Antiferromagnetism, chapter 8, pages 432–485. John Wiley & Sons, Ltd, 2001.

18. L. V. Velikov and E. G. Rudashevskii. Antiferromagnetic resonance in hematite in the weakly ferromagnetic state. Soviet Physics JETP, 29(5):836, May 1969.

19. Zavadskiy and H. Chumak. Tunable reflective structures based on weak ferromagnetics and their application as tunable subterahertz resonators. Radioelectronics and Communications Systems, 62(8):377–380, Aug 2019.

20. B. R. Morrison, A. H. Morrish, and G. J. Troup. High-Field Antiferromagnetic Resonance in α-Fe₂O₃. Physica status solidi (b), 56(1):183–195, 1973.

21. I. Boventer, H. T. Simensen, A. Anane, M. Kläui, A. Brataas, and R. Lebrun. Room-temperature antiferromagnetic resonance and inverse spin-hall voltage in canted antiferromagnets. Phys. Rev. Lett., 126:187201, May 2021.

22. Hailong Wang, Yuxuan Xiao, Mingda Guo, Eric Lee-Wong, Gerald Q. Yan, Ran Cheng, and Chunhui Rita Du. Spin pumping of an easy-plane antiferromagnet enhanced by dzyaloshinskii-moriya interaction. Phys. Rev. Lett., 127:117202, Sep 2021.

23. C. Caspers, V. P. Gandhi, A. Magrez, E. de Rijk, and J.-P. Ansermet. Sub-terahertz spectroscopy of magnetic resonance in BiFeO₃ using a vector network analyzer. Applied Physics Letters, 108(24):241109, 2016.

24. M. Bialek, A. Magrez, A. Murk, and J.-Ph. Ansermet. Spin-wave resonances in bismuth orthoferrite at high temperatures. Phys. Rev. B, 97:054410, Feb 2018.

25. M. Bialek, T. Itø, H. Rønnow, and J.-Ph. Ansermet. Terahertz-optical properties of a bismuth ferrite single crystal. Phys. Rev. B, 99:064429, Feb 2019.

26. M. Bialek, A. Magrez, and J.-Ph. Ansermet. Spin-wave coupling to electromagnetic cavity fields in dysprosium ferrite. Phys. Rev. B, 101:024405, Jan 2020.

27. D. L. a de Faria, Venâncio Silva, and M. T. de Oliveira. Raman microspectroscopy of some iron oxides and oxyhydroxides. Journal of Raman Spectroscopy, 28(11):873–878, 1997.

28. I. Chamristski and G. Burns. Infrared- and raman-active phonons of magnetite, maghemite, and hematite: A computer simulation and spectroscopic study. The Journal of Physical Chemistry B, 109(11):4965–4968, 2005. PMID: 16863155.

29. A. M. Jubb and H. C. Allen. Vibrational spectroscopic characterization of hematite, maghemite, and magnetite thin films produced by vapor deposition. ACS Applied Materials & Interfaces, 2(10):2804–2812, 2010.

30. M. Eibschütz, S. Shtrikman, and D. Treves. Internal field in orthoferrites and the one third power law. Solid State Communications, 4(3):141 – 145, 1966.

31. G. S. Ovchinnikov, V. V. Rudenko, and V. I. Tugarinov. Temperature dependence of the uniaxial magnetic anisotropy of rhombohedral antiferromagnetic crystals with ions in the s state. Physics of the Solid State, 52(1):112–116, Jan 2010.

32. N. Pailhè, J. Majimel, S. Pechev, P. Gravereau, M. Gaudon, and A. Demourgues. Investigation of Nanocrystallized α-Fe₂O₃ Prepared by a Precipitation Process. The Journal of Physical Chemistry C, 112(49):19217–19223, 2008.