Néel Spin Orbit Torque driven antiferromagnetic resonance in Mn$_2$Au probed by time-domain THz spectroscopy

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We observe the excitation of collective modes in the THz range driven by the recently discovered Néel spin-orbit torques (NSOT) in the metallic antiferromagnet Mn$_2$Au. Temperature dependent THz spectroscopy reveals a strong absorption mode centered near 1 THz, which upon heating from 4 K to 450 K softens and looses intensity. Comparison with the estimated eigenmode frequencies implies that the observed mode is an in-plane antiferromagnetic resonance (AFMR) mode. The AFMR absorption strength exceeds those found in antiferromagnetic insulators, driven by the magnetic field of the THz radiation, by three orders of magnitude. Based on this and the agreement with our theory modelling, we infer that the driving mechanism for the observed mode is the current induced NSOT. This electric manipulation of the Néel order parameter at high frequencies makes Mn$_2$Au a prime candidate for AFM ultrafast memory applications.

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Antiferromagnetic (AFM) materials are attracting significant interest in the field of spintronics [1-5]. The magnetic order consisting of alternating magnetic moments on neighboring atoms results in zero net magnetization and makes the AFMs insensitive to external magnetic fields. At the same time, it also leads to the absence of stray fields in AFMs. Compared to ferromagnets, this property can enable a significant reduction of the minimum volume necessary to store one bit of information. On the other hand, insensitivity of AFMs to external magnetic field makes an efficient manipulation and detection of the magnetic state of an AFM challenging.

A key advance in overcoming this challenge is the recent proposal [7] of an all-electrical switching of the staggered magnetization of metallic AFMs via an electrically induced Néel spin-orbit torque (NSOT), which has been experimentally realized recently [8-11]. The NSOT takes place in AFMs with a lack of local inversion symmetry. Here, charge currents produce a staggered Néel spin-orbit torque (NSOT) [7], which presents a novel route of manipulating metallic AFMs. Two materials, CuMnAs [8, 12] and Mn$_2$Au [13-15], that meet the indicated requirements are known to date. In both materials, switching of the Néel vector by pulsed DC currents was demonstrated [8,10,16]. In CuMnAs, Néel vector switching was also realised recently using intense THz pulses [17]. Provided that frequencies of collective modes, i.e., antiferromagnetic resonances (AFMR), in these metallic AFMs indeed lie in the terahertz (THz) range [18-20], ultrafast switching of the Néel vector on the picosecond timescale can be achieved.

Among the metallic AFM materials Mn$_2$Au is of special interest due to its high Néel temperature ($\approx 1500$ K) [13, 14], strong spin-orbit coupling, and high conductivity [15]. Bringing the recently demonstrated switching of the Néel vector by pulsed DC currents [9,10] towards the THz regime is a necessary key advance for the possible realisation of ultrafast switching and a bridge materials to overcome the THz communication gap.

In this Letter, we report on the temperature dependent THz conductivity data of c-axis epitaxial Mn$_2$Au thin films. In addition to the Drude free carrier response, the complex THz conductivity data reveal the presence of a mode, centered near 1 THz at 4 K. The mode displays significant softening and loss of intensity upon increasing the temperature to 450 K, as expected for an AFMR. By comparing the experimentally determined mode frequency to the theoretically estimated values we attribute the 1 THz mode to an in-plane AFMR. The mode’s absorption strength is found to be about three orders of magnitude larger that in typical insulating antiferromagnets, and cannot be accounted for by the coupling to the magnetic field component of the THz pulse. By comparing the magnitudes of the Zeeman torque with expected values of the Néel spin-orbit torque in Mn$_2$Au, we conclude that the in-plane AFMR mode in Mn$_2$Au is driven by the AC currents via the NSOT.

We have grown C-axis (001) oriented Mn$_2$Au thin films, with the crystal structure depicted in Fig. 1(a), on a 530 µm thick r-cut Al$_2$O$_3$ substrate with the lateral size of 10×10 mm$^2$ by radio–frequency magnetron sputtering [15]. To ensure epitaxial growth, 40 nm thick Mn$_2$Au films are deposited on a 8 nm thick (001) Ta buffer layer. The films are capped by ~ 2 nm of Al, forming an insulating aluminum oxide, to protect Mn$_2$Au from oxidation. One half of the metallic film is etched off to serve as a reference in time-domain THz spectrometer (TDTS) in transmission configuration (see Fig. 1(b)). The home-
The measurements of $\sigma(\nu)$ were performed between 4 K and 450 K. The real and imaginary part of the THz conductivity, recorded at 50 and 300 K, are presented in Fig. 2(a). $\sigma(\nu)$ is clearly dominated by the Drude free carrier response. The low frequency limit of $\sigma_1(\nu, 300$ K) matches the room temperature DC conductivity, recorded on the same sample by the Van der Pauw method. Dashed black lines in Figure 2(a) present the best fit to the data obtained by the simple Drude model, $\sigma^D(\nu) = 2\pi\varepsilon_0\nu_D^2/(\gamma_D - i\nu)$, with the plasma frequency $\nu_D \approx 600$ THz, and the free carrier scattering rate, $\gamma_D (300$ K) $\approx 12$ THz. In addition, a distinct spectral feature, consistent with a resonance centered around 1 THz, is observed for all temperatures in both $\sigma_1(\nu)$ and $\sigma_2(\nu)$. To account for both, the free carrier response and the $\approx 1$ THz mode, we use the Drude-Lorentz model solid blue lines, where a finite frequency mode with a linewidth of $\approx 0.3$ THz is centered near $\approx 1$ THz. There is an upturn in $\sigma_2$ at $\nu \lesssim 0.5$ THz, which might suggest a possible second low-frequency mode centered much below 0.2 THz [27]. Since the use of a 3 mm aperture limits our spectral range to $\nu > 0.3$ THz, we do not include additional modes in the data analysis.

Figure 2(b) presents the temperature dependence of the Drude scattering rate (plasma frequency was kept constant). It coincides with the temperature dependence of the DC resistivity [15]. To track the temperature dependence of the $\approx 1$ THz mode, we subtracted the Drude part, $\sigma^D(\nu, T)$, from the experimental $\sigma(\nu, T)$. As shown by $\sigma_1(\nu, T) - \sigma^D_1(\nu, T)$, presented in Fig. 2(c), the mode frequency clearly red shifts with increasing temperature. While the linewidth shows no temperature dependence within the measured temperature range, the mode’s spectral weight, $S_{\text{spectral}} \propto \int (\sigma_1(\nu, T) - \sigma^D_1(\nu, T))\, d\nu$, is substantially reduced upon increasing the temperature.

Based on the low frequency of the mode and its temperature dependence we attribute the mode to one of
the two $q = 0$ antiferromagnetic eigenmodes of Mn$_2$Au. The parameters of this AFMR, obtained by analyzing $\sigma(q, T)$ with the Drude-Lorentz model, are presented in Fig. 3. The mode frequency, $\nu_{\text{AFMR}}$, clearly shows a shift from 1.03 THz to 0.88 THz as the temperature increases from 5 K to 450 K (Fig. 3(a)). For Mn$_2$Au only the extrapolated value of the Néel temperature, $T_n$, is known (1300 K $<$ $T_n$ $<$ 1600 K) [14], since the crystal becomes structurally unstable around 1000 K. Furthermore, little is known of the temperature dependence of anisotropy fields in Mn$_2$Au. Thus, we use a simple Ginzburg-Landau model, $\nu_{\text{AFMR}} \propto \sqrt{T_n - T}$, to evaluate the temperature dependence of $\nu_{\text{AFMR}}$. As shown in the inset of Fig. 3(a), the resulting extrapolated value of $T_n$ is 1500 K, which is consistent with previously reported estimates [14]. We observe a pronounced loss of the mode’s spectral weight, $S_{\text{AFMR}}$, signifying a gradual decrease of the order parameter (i.e., the mode should disappear at $T_n$). The mode damping, $\Gamma_{\text{AFMR}}$, shows no measurable temperature dependence, suggesting that the damping is due to the interaction with free carriers which are mostly unaffected in this temperature range.

The experimentally determined $\nu_{\text{AFMR}}$ should be compared to the expected eigenmode frequencies of Mn$_2$Au. Since Mn$_2$Au is an easy plane AFM, with a strong out-of-plane and very weak in-plane magnetic anisotropy, two nearly linearly polarized AFMR modes exist, similar to NiO [18]. The first mode, sketched in Figure 4 is an in-plane ($\parallel$) mode, where the two sub-lattice magnetization vectors, $M_1$ and $M_2$, with $|M_1| = |M_2| = M_s$, precess in opposite directions. The bases of the resulting cones are narrow ellipses whose long axes lie in the easy plane. While in equilibrium $M_1$ and $M_2$ are anti-parallel and fully compensate each other, the dynamics result in a small oscillating net magnetization $\mathbf{m} = M_1 + M_2 \neq 0$ pointing in the direction of the hard axis (Figure 4). The second eigenmode is the out-of-plane ($\perp$) mode with the long axes of ellipses pointing along the hard-axis ($\perp$) and the oscillating net magnetization lying in the easy-plane.

Following Kittel’s approach [28], we estimate the two eigenfrequencies of Mn$_2$Au as

$$\nu_{\text{AFMR}}^{\parallel} = \frac{\gamma}{\sqrt{H_{ex} H_{an}^{\parallel}}},$$

where $H_{ex}$ is the exchange field, $H_{an}^{\parallel}$ and $H_{an}^{\perp}$ are the in-plane and the out-of-plane anisotropy field, respectively, and $\gamma = 1.76 \cdot 10^{11} \text{s}^{-1} \text{T}^{-1}$ is the gyro-magnetic ratio. Using $H_{ex} \approx 1300$ T [14], $H_{an}^{\parallel} \approx 0.3$ T, and $H_{an}^{\perp} \approx 10$ T [29] we obtain $\nu_{\text{AFMR}}^{\parallel} \approx 0.6$ THz and $\nu_{\text{AFMR}}^{\perp} \approx 5$ THz, where $\nu_{\text{AFMR}}^{\parallel, \perp} \equiv \omega_{\text{AFMR}}^{\parallel, \perp}/2\pi$. These estimates suggest that we observe the in-plane AFMR (Fig. 4).

As noted above, the mode’s absorption strength is much higher than in insulating AFMs like NiO [18] [19], MnO [18] and α-RuCl$_3$ [20], where sample thicknesses of several 100 μm are required for THz absorption measurements [18] [20] [30] [31]. For quantitative assessment, we compare the THz absorption coefficients at the center-frequencies of AFMRs, $\alpha_{\text{AFMR}}$, for several AFMs. In Mn$_2$Au the coefficient is $\alpha_{\text{AFMR}} \approx 880 \text{ mm}^{-1}$ at 50 K and 300 mm$^{-1}$ at 300 K, in NiO $\alpha_{\text{AFMR}} \approx 0.18 \text{ mm}^{-1}$ at 300 K [32] and in α-RuCl$_3$ $\alpha_{\text{AFMR}} \approx 0.2 \text{ mm}^{-1}$ at 4 K [20].

In insulating AFMs, like NiO, the Zeeman torque exerted by the magnetic field component of the THz pulse is
commonly ascribed to drive the AFMR mode [18, 19, 28]. However, the in-plane AFMR in Mn$_2$Au does not couple to the magnetic field component of the THz pulse (in-plane polarized). This fact, together with the three orders of magnitude larger absorption compared to insulating AFMs, indicates that an alternative driving mechanism of the AFMR in Mn$_2$Au must be present. Recent reports on current induced switching of the Néel vector in both CuMnAs [8, 10] and Mn$_2$Au [9, 10], where in the former even switching with intense THz pulses was demonstrated [17], demonstrate the effectiveness of NSOT.

Next we analyze if this same scenario involving NSOT is able to quantitatively account for both, the polarization and strength of the observed AFMR. Due to the strong exchange coupling between the two magnetic sublattices, given by $H_{\text{ex}}$, the oscillating net magnetization $\mathbf{m}$ of the AFMR is very small and the dynamics can be fully described by the Néel vector, $\mathbf{n} = \mathbf{M}_1 - \mathbf{M}_2$. Interaction of a system with broken local inversion symmetry, like CuMnAs and Mn$_2$Au, with an electromagnetic wave results in two external field-like torques: i) a torque created by a time-dependent magnetic field component, $\propto \mathbf{n} \times \hat{B}_{\text{em}} \times \mathbf{n}$ [33], and ii) a Néel spin orbit torque $\propto \mathbf{n} \times \hat{z} \times \mathbf{E}_{\text{em}}$, which is driven by current $\mathbf{j} = \sigma \mathbf{E}_{\text{em}}$, induced by an electric field component of an electro-magnetic wave, $\mathbf{E}_{\text{em}}$, and $\sigma$ is the optical conductivity. The resulting equation of motion for the Néel vector is given by [33, 35]:

$$\mathbf{n} \times (\mathbf{\dot{n}} + 2\alpha_G \gamma H_{\text{ex}} \mathbf{n} - 2\gamma^2 H_{\text{ex}} M_s \mathbf{H}_n) = \gamma \mathbf{n} \times (\hat{B}_{\text{em}} \times \mathbf{n} + 2\lambda_{\text{NSOT}} \sigma H_{\text{ex}} M_s \mathbf{E}_{\text{em}} \times \hat{z}).$$

(1)

Here $\alpha_G$ is the Gilbert damping constant, $H_n = -\partial w_{\text{an}}/\partial \mathbf{n}$ is the internal effective field, determined by the magnetic anisotropy energy landscape, $w_{\text{an}}$, and $\lambda_{\text{NSOT}}$ is a constant proportional to the NSOT strength $\approx \sqrt{10}$ cm$^{-1}$. With the electromagnetic wave polarized within the $x-y$ plane, where $\mathbf{E}_{\text{em}}[0] \parallel \hat{x}$ and $\mathbf{E}_{\text{em}} \parallel \hat{y}$ (see Figure 4), the resulting equations for small deviations of the Néel vector from its equilibrium state, $\dot{n}_{y,z}$ (in dimensionless form), are

$$\dot{n}_y + 2\alpha_G \gamma H_{\text{ex}} \dot{n}_y + (\omega_{\text{AFMR}}^\parallel)^2 \dot{n}_y = -\gamma \lambda_{\text{NSOT}} \sigma H_{\text{ex}} \mathbf{E}_{\text{em}},$$

$$\dot{n}_z + 2\alpha_G \gamma H_{\text{ex}} \dot{n}_z + (\omega_{\text{AFMR}}^\parallel)^2 \dot{n}_z = -\gamma \dot{B}_{\text{em}}.$$  

(2)

The above equations show that the NSOT drives the in-plane mode, while the torque created by the magnetic field component of the THz pulse couples to the out-of-plane mode. Moreover, taking the theoretically estimated $\lambda_{\text{NSOT}} = 5 - 50$ s$^{-1}$Å$^{-1}$cm$^2$ [2, 33], $\omega_{\text{AFMR}}^\parallel \approx 6.3 \cdot 10^{12}$ s$^{-1}$ and $\sigma$ (1 THz, 4 K) $\approx 3.4 \cdot 10^8$ Ω$^{-1}$cm$^{-1}$, and assuming $E_{\text{em}}/B_{\text{em}} = c$, a comparison of the strengths of the two driving fields gives $(2\lambda_{\text{NSOT}} \sigma E_{\text{em}})/B_{\text{em}} \omega_{\text{AFMR}}^\parallel / \approx 10^2 - 10^3$, accounting for the anomalously large absorption strength in Mn$_2$Au compared to insulating AFMs.

Based on the measured linewidth $\gamma_{\text{AFMR}}$ and $\nu_{\text{AFMR}}^\parallel$ and Eq (2) we can also estimate the value of the Gilbert damping. From $\gamma_{\text{AFMR}}/\nu_{\text{AFMR}}^\parallel = 2\alpha_G \sqrt{H_{\text{ex}}/H_{\text{an}}^\parallel}$, where $\gamma_{\text{AFMR}}$ = 0.3 THz and $\nu_{\text{AFMR}}^\parallel$ = 1 THz, we obtain a low value of $\alpha_G$ $\approx$ 2.5 · 10$^{-3}$, which is typical for metallic antiferromagnets [33].

Finally, from the experimentally determined frequencies we obtain the spin-flop field, $H_{s-f} \equiv \omega_{\text{AFMR}}^\parallel/\gamma = \sqrt{H_{\text{ex}}/H_{\text{an}}^\parallel}$, to be 35 T at 4 K and 30 T at 300 K, consistent with recent measurements on identical films exposed to pulsed magnetic fields [37].

In summary, time-domain THz spectroscopy of Mn$_2$Au thin films reveals the presence of a strong mode near 1 THz. The comparison of the mode’s frequency to the estimated AFMR frequencies of Mn$_2$Au shows that the mode is likely an in-plane AFMR. Compared to previous reports on insulating AFMs, the mode has an anomalously high absorption strength. Since the (in-plane) magnetic field component of the THz pulse only weakly couples to the in-plane AFMR, we suggest the in-plane AFMR in Mn$_2$Au is driven by the AC current producing a Néel spin-orbit torque. The high frequency of the mode, its driving mechanism, and the recently demonstrated DC-current switching of the Néel vector [9] make Mn$_2$Au a prime candidate for AFM ultrafast memory applications.

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[1] I. Žutić, J. Fabian, and S. D. Sharma, Rev. Mod. Phys. 76, 323 (2004).
[2] A. H. MacDonald and M. Tsoi, Phil. Trans. R. Soc. A 369, 3098–3114 (2011).
[3] E. V. Gomonay and V. M. Loktev, Low Temp. Phys. 40, 17 (2014).
[4] T. Jungwirth, X. Marti, P. Wadley, and J. Wunderlich, Nature Nanotechnology 11, 231-241 (2016).
[5] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono and Y. Tserkovnyak, arXiv:1606.04284 (2017).
[6] A. S. Núñez, R. A. Duine, Paul Haney, and A. H. MacDonald, Phys. Rev. B 73, 214426 (2006).
[7] J. Železný, H. Gao, K. Výborný, J. Zemen, J. Mašek, A. Manchon, J. Wunderlich, J. Sinova, and T. Jungwirth, Phys. Rev. Lett. 113, 157201 (2014).
[8] P. Wadley, B. Howells, J.Železný, C. Andrews, V. Hills, R. P. Campion, V. Novák, K. Olejník, F. Maccherozzi,
S. S. Dhesi, S. Y. Martin, T. Wagner, J. Wunderlich, F. Freimuth, Y. Mokrousov, J. Kunes, J. S. Chauhan, M. J. Grzybowski, A. W. Rushforth, K. W. Edmonds, B. L. Gallagher, T. Jungwirth, *Science* **351**, 587-590 (2016).

[9] S. Yu. Bodnar, L. Smejkal, I. Turek, T. Jungwirth, O. Gomonay, J. Sinova, A. A. Sapozhnik, H.-J. Elmers, M. Kläui, and M. Jourdan, *Nature Commun.* **9**, 348 (2018).

[10] M. Meinert, D. Graulich, T. Matalla-Wagner, arXiv:1706.06983 (2017).

[11] V. Saidl, et al., *Nature Photonics* **11**, 91–96 (2017).

[12] P. Wadley, et al., *Nature Commun.* **4**, 2322 (2013).

[13] S. Khmelevskyi, and P. Mohn, *Appl. Phys. Lett.* **93**, 162503 (2008).

[14] V. M. T. S. Barthem, C. V. Colin, H. Mayaffre, M.-H. Julien, and D. Givord, *Nature Commun.* **4**, 2892 (2013).

[15] M. Jourdan, H. Bräuning, A. Sapozhnik, H.-J. Elmers, H. Zabel and M. Kläui, *J. Phys. D: Appl. Phys.* **48**, 385001 (2015).

[16] P. Wadley, et al., arXiv:1711.08444 (2017).

[17] K. Olejnik, et al., arXiv:1711.08444 (2017).

[18] A. J. Sievers, and M. Tinkham, *Phys. Rev.* **129**, 1566 (1963).

[19] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer and R. Huber, *Nature Photonics* **5**, 31–34 (2011).

[20] A. Little, L. Wu, P. Lampen-Kelley, A. Banerjee, S. Patankar, D. Rees, C. A. Bridges, J.-Q. Yan, D. Mandrus, S. E. Nagler, and J. Orenstein, *Phys. Rev. Lett.* **119**, 227201 (2017).

[21] M. Beck, M. Klammer, S. Lang, P. Leiderer, V. V. Kabanov, G. N. Gol’tsman, and J. Demsar, *Phys. Rev. Lett.* **107**, 177007 (2011).

[22] M. Beck, et al., *Phys. Rev. B* **95**, 085106 (2017).

[23] M. Beck, H. Schäfer, G. Klatt, J. Demsar, S. Winnerl, M. Helm, and T. Dekorsy, *Opt. Express* **18**, 9251 (2010).

[24] P. N. Baker, *Thin Solid Films* **14**, 3-25 (1972).

[25] J. Lloyd-Hughes and T.-I. Jeon, *J. Infrared Milli. Terahz. Waves* **33**, 871–925 (2012).

[26] Supplementary Online Information...

[27] M. Arana, F. Estrada, D. S. Maior, J. B. S. Mendes, L. E. Fernandez-Outon, W. A. A. Macedo, V. M. T. S. Barthem, D. Givord, A. Azevedo, and S. M. Rezende, *Appl. Phys. Lett.* **111**, 192409 (2017).

[28] C. Kittel, *Physical Review* **82**, 565 (1951).

[29] A. B. Shick, S. Khmelevskyi, O. N. Myrasov, J. Wunderlich, and T. Jungwirth, *Phys. Rev. B* **81**, 212409 (2010).

[30] R. V. Mikhailovskiy, E. Hendry, F. Y. Ogrin, and V. V. Kruglyak, *Phys. Rev. B* **87**, 094414 (2013).

[31] S. G. Chou, P. E. Stutzman, S. Wang, E. J. Gaborczi, W. F. Egelhoff, and D. F. Plusquellic, *J. Phys. Chem. C* **116**, 16161 (2012).

[32] T. Higuchi, N. Kanda, H. Tamaru, and M. Kuwata-Gonokami, *Phys. Rev. Lett.* **106**, 047401 (2011).

[33] T. Satoh, S. J. Cho, R. Iida, T. Shimura, K. Kuroda, H. Ueda, Y. Ueda, B. A. Ivanov, F. Nori, and M. Fiebig, *Phys. Rev. Lett.* **105**, 077402 (2010).

[34] O. Gomonay, T. Jungwirth, and J. Sinova, *Phys. Rev. Lett.* **117**, 017202 (2016).

[35] O. Gomonay, T. Jungwirth, and J. Sinova, arXiv:1712.02686 (2017).

[36] J. Zelezný, H. Gao, A. Manchon, F. Freimuth, Y. Mokrousov, J. Zemen, J. Mašek, J. Sinova, T. Jungwirth, *Phys. Rev. B* **95**, 014403 (2017).

[37] A. A. Sapozhnik, et al., in preparation.