

Coherent spin-wave transport in an antiferromagnet

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Magnonics is a research field complementary to spintronics, in which the quanta of spin waves (magnons) replace electrons as information carriers, promising less energy dissipation¹⁻³. The development of ultrafast nanoscale magnonic logic circuits calls for new tools and materials to generate coherent spin waves with frequencies as high, and wavelengths as short, as possible^{4,5}. Antiferromagnets can host spin waves at THz frequencies and are therefore seen as a future platform for the fastest and the least dissipative transfer of information⁶⁻¹¹. However, the generation of short-wavelength coherent propagating magnons in antiferromagnets has so far remained elusive. Here we report the efficient emission and detection of a nanometer-scale wavepacket of coherent propagating magnons in antiferromagnetic DyFeO₃ using ultrashort pulses of light. The subwavelength confinement of the laser field due to large absorption creates a strongly non-uniform spin excitation profile, enabling the propagation of a broadband continuum of coherent THz spin waves. The wavepacket features magnons with detected wavelengths down to 125 nm that propagate with supersonic velocities of more than 13 km/s into the material. The long-sought source of coherent short-wavelength spin carriers demonstrated here opens up new prospects for THz antiferromagnetic magnonics and coherence-mediated logic devices at THz frequencies.

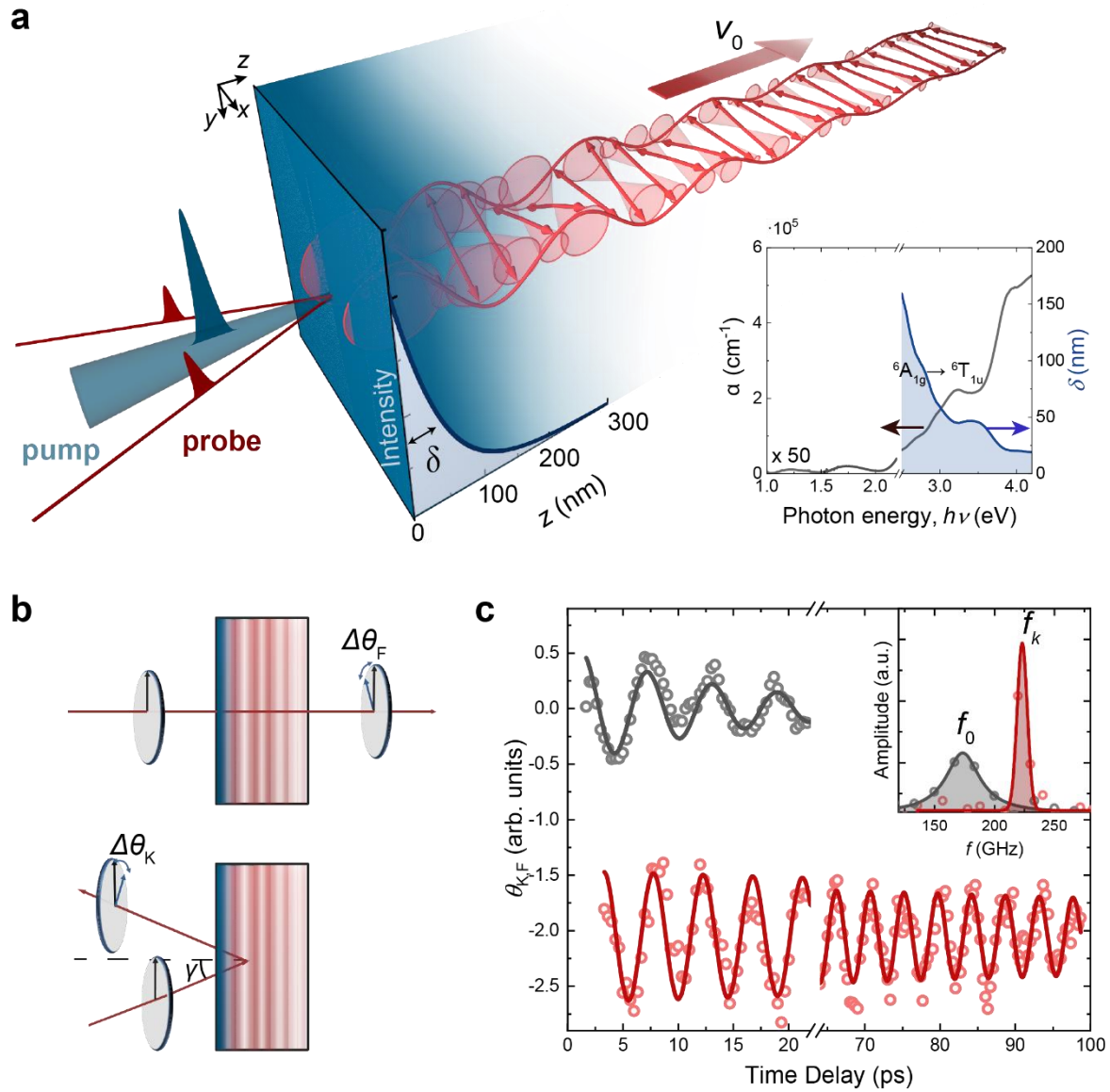
Antiferromagnetic insulators (AFMs) are prime candidates to replace ferromagnets (FMs) as active media in the quest towards high-speed spin transport and large spectral bandwidth operation⁶⁻⁸. Integration of AFMs in future wave-based technologies³ crucially requires the realization of coherent (ballistic) transport of antiferromagnetic spin waves over large distances⁵. In this regard, non-uniform spin-wave modes with short wavelengths ($\lambda \lesssim 100$ nm) are of particular importance: they can operate at THz clock rates, exhibit high propagation velocities and enable the miniaturization of devices down to the nanoscale. Phase-coherent ballistic spin transport in AFMs

40 is also interesting from a fundamental point of view, as it is predicted to be a prerequisite for the
41 occurrence of exotic phenomena such as magnetic solitons¹², Bose-Einstein condensates^{13,14} and
42 spin-superfluidity¹⁵⁻¹⁷. These prospects call for efficient methods for the excitation, manipulation,
43 and detection of short-wavelength coherent antiferromagnetic magnons.

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45 Conventional methods of linear spin-wave excitation use spatially varying oscillating magnetic
46 fields. However, the high-frequency THz resonances inherent to antiferromagnetic dynamics make
47 traditional field sources based on microstrip lines or coplanar waveguides impractical to be used
48 in antiferromagnetic media. As a result, recent demonstrations of magnon-mediated spin transport
49 in antiferromagnets were represented either by diffusive propagation of incoherent magnons⁹⁻¹¹ or
50 by evanescent spin-wave modes¹⁸, and the generation of coherent propagating short-wavelength
51 magnons leading to phase-coherent transport in an antiferromagnet has yet to be experimentally
52 realized.

53
54 Ultrashort pulses of light have been routinely used to generate and to control large-amplitude THz
55 spin precession¹⁹⁻²¹ in antiferromagnets. The small photon momentum, however, poses a problem:
56 it gives rise to a large momentum mismatch with short-wavelength spin waves. Consequently,
57 optical techniques have so far been restricted to the generation of $k = 0$ uniform antiferromagnetic
58 magnons and/or pairs of mutually coherent magnons at the edges of the Brillouin zone²², for which
59 group velocities are (near-)zero and no spatial transport of energy and angular momentum takes
60 place. Here we overcome this problem and present an all-optical method to excite and detect a
61 broadband wavepacket of short-wavelength coherent propagating magnons in an insulating
62 antiferromagnet. Optical excitation of intense charge-transfer electronic transitions using
63 ultrashort pulses in the prototypical antiferromagnet DyFeO₃ provides strong confinement of the
64 optical field, which creates a narrow exponential profile of deflected spins near the sample surface.
65 This magnetic non-uniformity extends over the nanoscale penetration depth of the light and serves
66 as a source of short-wavelength coherent spin waves propagating into the sample bulk, as
67 illustrated in Figure 1a. Using k -selective magneto-optical Bragg detection we map out spectral
68 components of the magnon wavepacket and reveal magnon modes with nanoscale wavelengths,
69 supersonic group velocities and an estimated propagation length of more than 1 μm .

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73 **Figure 1: All-optical generation and detection of coherent antiferromagnetic spin waves.** (a) Schematic
 74 illustration of the generation and detection of propagating antiferromagnetic spin waves after excitation of strongly
 75 absorbing electronic transitions. The optical penetration depth δ of the light defines the excited region and the width
 76 of the magnetic non-uniformity. Inset: Absorption coefficient (left axis) and corresponding penetration depth (right
 77 axis) for DyFeO_3 as function of photon energy (see methods). ${}^6A_{1g} \rightarrow {}^6T_{1u}$: the charge-transfer transition of interest.
 78 (b) Schematics for the optical detection mechanisms of spin waves in transmission (top) and reflection (bottom)
 79 geometries, measuring transient changes in the Faraday rotation (θ_F) and Kerr rotation (θ_K) respectively; γ is the angle
 80 of incidence. (c) Time-resolved measurements of the polarization rotation of a near-infrared probe pulse after
 81 excitation with pump pulses with a photon energy of 3.1 eV in the detection geometries shown in (b). The thick solid

82 lines are exponentially damped sine fits. Inset: Fourier spectra of the oscillations with Lorentzian fits (thick solid
83 lines), with central frequencies f_0 (transmission geometry) and f_k (reflection geometry). a.u.: arbitrary units.

84

85 Dysprosium orthoferrite (DyFeO_3) is a charge-transfer antiferromagnetic insulator with the Néel
86 temperature $T_N = 645$ K, exhibiting one of the strongest observed interactions between spins and
87 laser pulses^{19,23,24}. The optical spectrum of DyFeO_3 is dominated by a set of intense electronic O-
88 Fe ($2p$ - $3d$) charge-transfer (CT) transitions. The absorption due to these transitions sets in above
89 2 eV, and promptly brings the absorption coefficient to values as high as $5 \times 10^5 \text{ cm}^{-1}$ (see inset Fig.
90 1a)²⁵. Such strong absorption enables confinement of light down to penetration depths (δ) of less
91 than 50 nm.

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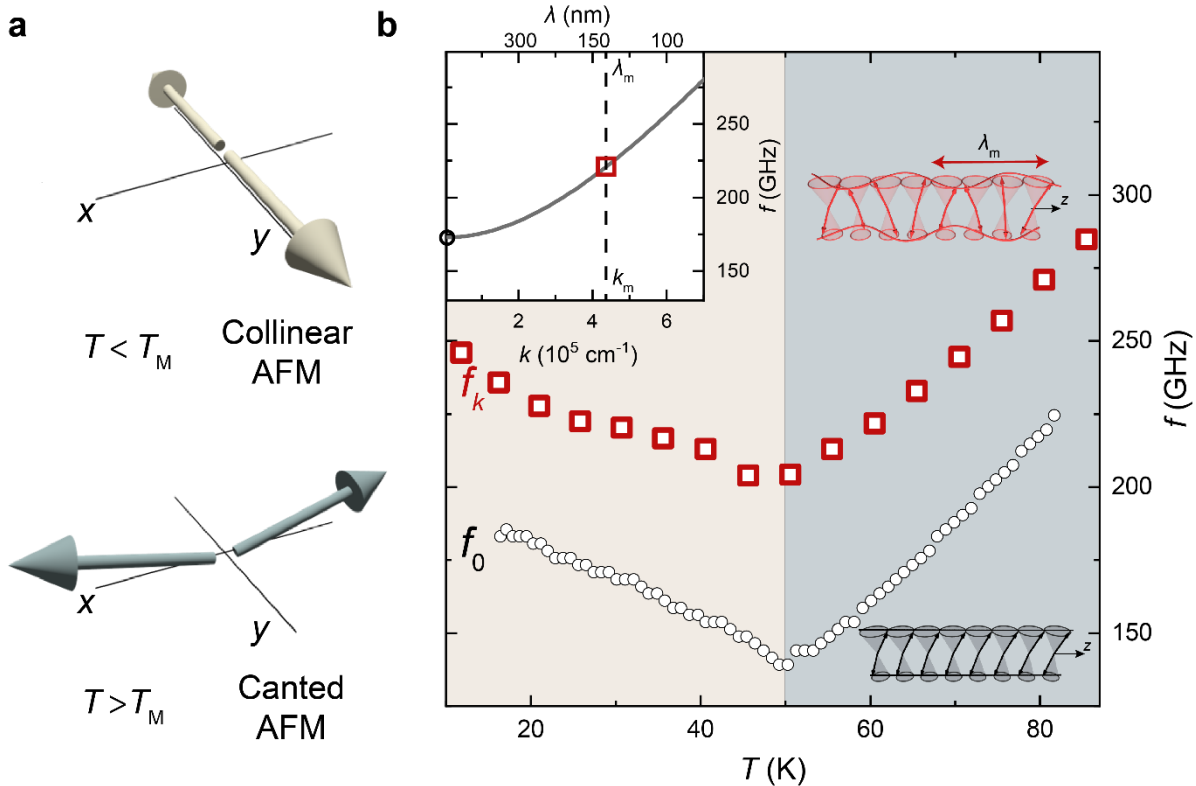
93 In our experiments we study a 60 μm thick slab of z -cut DyFeO_3 . The sample is excited with 100 fs
94 pump pulses which have photon energy tunable in the spectral range of 1.5 - 3.1 eV, covering the
95 lowest energy ${}^6\text{A}_{1g} \rightarrow {}^6\text{T}_{1u}$ charge-transfer electronic transition²⁵. We use time-delayed probe pulses
96 at various photon energies below the fundamental absorption gap ($h\nu < 2$ eV) to detect the photo-
97 induced magnetic dynamics, see Extended Data Fig. 1. The probing is simultaneously performed
98 in two complementary experimental geometries: a transmission geometry using the Faraday effect
99 and a reflection geometry, using the magneto-optical Kerr effect (MOKE) (see Fig. 1b). In both
100 geometries, the pump-induced rotation of the probe polarization plane, originating from the
101 Faraday effect (θ_F) or the MOKE (θ_K), is tracked as a function of the pump-probe time delay. Note
102 that while the Faraday transmission geometry is routinely used in pump-probe experiments for
103 detecting uniform ($k = 0$) spin precession in antiferromagnets¹⁹, the reflection geometry has been
104 shown to enable detection of finite- k coherent excitations such as propagating acoustic
105 wavefronts^{26,27}. As shown below we demonstrate that the reflection geometry can be also used to
106 probe the dynamics of short-wavelength propagating coherent spin waves.

107

108 Following the optical pumping in the regime of strong absorption ($h\nu = 3.1$ eV, $\delta = 50$ nm) the
109 time-resolved dynamics of the probe polarization reveal high-frequency oscillations in the
110 hundreds of GHz range (see Fig. 1c). The frequencies f_0 and f_k of the oscillations observed in the
111 transmission and reflection geometry respectively, are substantially different: $f_k > f_0$ (see inset Fig.
112 1c). Notably, the decay time of the oscillations also differ by nearly an order of magnitude.

113

114 To identify the origin of the oscillations, we track their central frequency as a function of
 115 temperature. The antiferromagnetic state in DyFeO₃ adopts two distinct spin arrangements, sharply
 116 separated by a first-order phase transition at the so-called Morin temperature $T_M \approx 50$ K²⁸. At
 117 $T < T_M$, the antiparallel iron spins are oriented along the y -axis and arranged in a compensated
 118 collinear AFM pattern. Above T_M , the spins experience a reorientation towards the x -axis
 119 accompanied by the mutual canting and stabilization of a canted AFM phase (see Fig. 2a). The
 120 experimentally acquired temperature dependence of the oscillation frequency exhibits a
 121 characteristic cusp-like softening with a minimum at T_M (see Fig. 2b and Extended Data Fig. 2).
 122 This frequency softening is an unambiguous hallmark of the so-called quasi-antiferromagnetic (q -
 123 AFM) magnon branch in DyFeO₃ and is caused by strong temperature variations of the magneto-
 124 crystalline anisotropy in the vicinity of the spin-reorientation phase transition²⁹. Indeed, the
 125 frequencies f_0 of the oscillations observed in the transmission geometry perfectly match values
 126 reported in literature for the zone-centre ($k = 0$) q -AFM magnon²⁹.



127
 128 **Figure 2: Generation and detection of coherent finite- k antiferromagnetic spin waves.** (a) The spin orientation in
 129 the collinear AFM (top panel) and canted AFM phase (bottom panel). (b) Temperature (T) dependence of the central
 130 frequency of the oscillatory dynamics as measured in the reflection geometry (red square markers) after excitation
 131 with strongly absorbed pump pulses ($h\nu = 3.1$ eV, $\delta = 50$ nm) compared with the $k = 0$ magnon T -dependence, as

132 measured in standard transmission geometry (black circle markers). See also Extended Data Fig. 2. Left top inset: The
 133 antiferromagnetic magnon dispersion of DyFeO₃ and the wavenumbers of the magnons observed in the reflection and
 134 transmission experimental geometries. Right insets: Schematic illustration of the spin wave corresponding to the
 135 oscillatory dynamics at the different frequencies.

136

137 To explain the physical origin of the oscillation at frequency f_k seen in the MOKE experiment, we
 138 refer to the dispersion relation for magnon modes in antiferromagnets. In both magnetic phases,
 139 below and above T_M , the magnon spectrum ω_k in DyFeO₃, is given by³⁰:

$$140 \quad \omega_k = \sqrt{\omega_0^2 + (v_0 k)^2}, \quad (1)$$

141 where $v_0 \approx 20$ km/s is the limiting group velocity of the spin waves³⁰. This dispersion relation is
 142 schematically shown as an inset to Fig. 2b. At small wavenumbers $kv_0 \ll \omega_0$, it has a quadratic
 143 form due to the magnon gap $\omega_0 = 2\pi f_0$, arising from magneto-crystalline anisotropy. At larger
 144 wavenumbers ($v_0 k \gg \omega_0$), the dispersion relation becomes dominated by the exchange interaction
 145 (exchange regime), and thus takes a linear form typical for antiferromagnets³⁰. We identify the
 146 MOKE signal at f_k as a finite- k magnon on the q -AFM branch: it follows the characteristic
 147 temperature dependence of the f_0 zone-center magnon mode and has a nearly temperature-
 148 independent blueshift. The detection geometry implies that the magnon wavevector \mathbf{k} is
 149 perpendicular to the sample surface, and its magnitude can be deduced from Eq. (1) to be
 150 $k = 4.2 \times 10^5 \text{ cm}^{-1}$ ($\lambda \approx 140$ nm), as marked on the inset to Figure 2b.

151

152 By considering the modulation of the material's dielectric tensor due to the propagating coherent
 153 spin waves, the attribution of the f_k oscillation to a finite- k magnon on the q -AFM branch can be
 154 further supported. A spin wave with a propagation vector along the z -axis results in a perturbation
 155 of the magnetic order and a corresponding periodic modulation of the off-diagonal components of
 156 the dielectric tensor³¹. In this way the spin waves in the medium produce the magneto-optical
 157 analogue of a dynamical volume phase grating for the probe light wave. The polarization state of
 158 the reflected probe is explained by the Bragg reflection of light from this diffraction grating, an
 159 approach similar to the one used in Brillouin light scattering studies on spin waves³². As a result,
 160 the polarization rotation of the reflected probe beam with wavenumber k_0 becomes subject to a
 161 Bragg condition:

$$162 \quad k_m = 2k_0 n(\lambda_0) \cos \gamma', \quad (2)$$

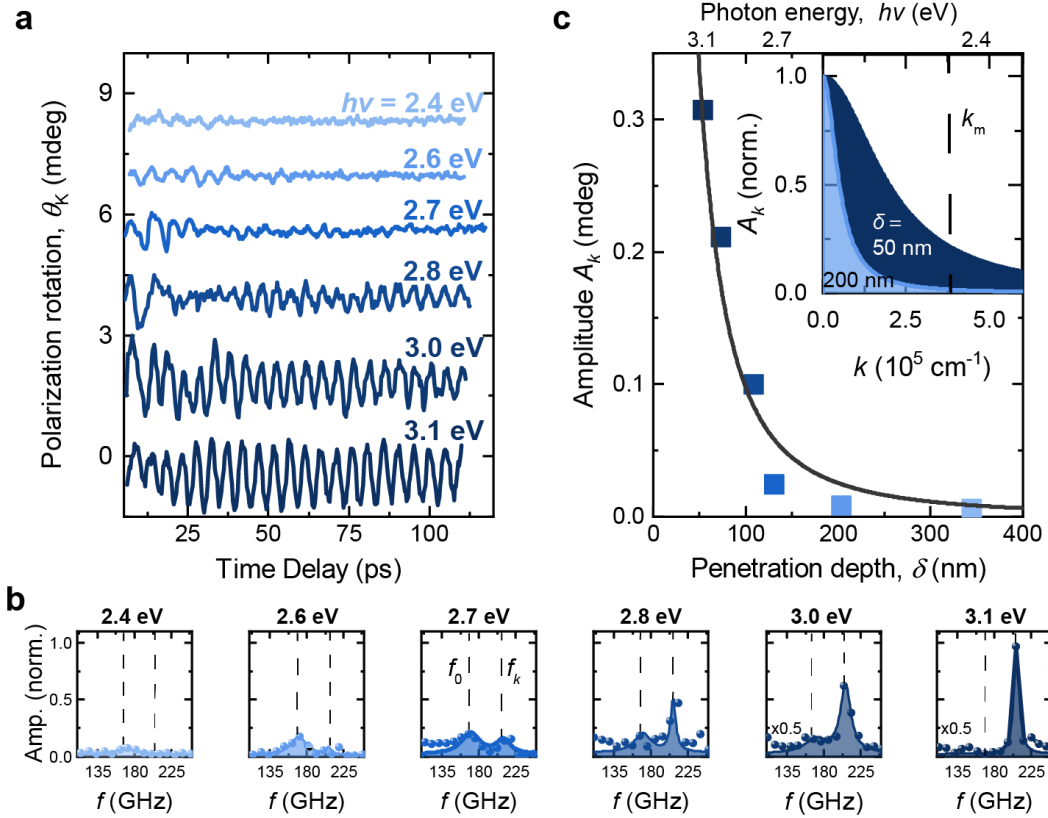
163 where $n(\lambda_0)$ is the optical refractive index of the medium at the probe wavelength λ_0 , γ' is the
164 refracted angle of incidence of the probe, and k_m is the normal projection of the \mathbf{k} -vector of the
165 magnon to which the probe pulse is sensitive (see Supplementary section S1). Using Eq. (2) we
166 find that a probe pulse at a central wavelength of 680 nm ($n \approx 2.39$)²⁵ and normal incidence ($\gamma' = 0$)
167 is sensitive to propagating magnons with wavenumber $k \approx 4.2 \times 10^5 \text{ cm}^{-1}$. Note that this independent
168 estimation perfectly matches the magnon wavenumber retrieved using the measured frequency and
169 known dispersion relation (Eq. (1)).

170

171 The generation and detection of finite- k coherent magnons is anticipated to rely strongly on the
172 confinement provided by the optical penetration depth δ , which is highly dispersive near the
173 charge-transfer band. In particular, changing the pump photon energy between 2.4 eV and 3.1 eV
174 provides a variation in the penetration depth between 300 and 50 nm, while the real part of the
175 refractive index (influencing the pump wavelength) changes by only 5% (see Extended Data Fig.
176 3). Therefore, we expect that by changing the photon energy of the pump pulses, the amplitude of
177 the finite- k magnon will vary strongly. The time-resolved MOKE signals obtained in the reflection
178 geometry for different photon energies of the pump excitation, are shown in Fig. 3a. The Fourier
179 transforms of the signals (Fig. 3b) show that the spectra are composed of two components,
180 corresponding to the zone-centre and finite- k ($k = 3.7 \cdot 10^5 \text{ cm}^{-1}$) magnon modes. We observe that
181 with increasing photon energy (i.e. decreasing penetration depth), the amplitude of the finite- k
182 magnon mode increases dramatically. The magnon amplitude extracted for the pump excitation at
183 different photon energies, as a function of the corresponding penetration length, is shown in Fig.
184 3c. The obtained dependence shows that the finite- k magnon is nearly absent for penetration depths
185 larger than 150 nm, a value close to the wavelength of the detected magnons, and grows
186 dramatically for shorter penetration lengths.

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Figure 3: Confinement of the light as a necessary condition for the generation of high- k spin waves. (a) Time-resolved signals of the polarization rotation of the near-infrared probe pulse after excitation with pump pulses with increasing photon energy in a reflection geometry. (b) Fourier amplitude spectra of the time-resolved signals from panel (a). (c) Amplitude of sine fit oscillations corresponding to the AFM propagating spin wave to the data from panel (a) vs penetration depth of the excitation pulse (color markers correspond to traces in panel (a)). The solid line is a fit using $I_0/(1 + (k\delta)^2)$. Inset: Magnon frequency distribution after excitation with pump pulses with $\delta = 50 \text{ nm}$ (dark blue, broadband distribution) and 200 nm (light blue, narrowband). The probe is sensitive to $k_m = 3.7 \cdot 10^5 \text{ cm}^{-1}$, indicated by the dashed line.

We model this observation using a simple assumption: the ultrashort light pulse promotes a spin excitation that is strongly non-uniform along the direction of incidence z . The excitation leads to a nearly instantaneous deflection of spins by an angle $\varphi(z)$ with the spatial distribution following the optical absorption profile given by the Beer-Lambert law: $\varphi(z, t=0) = \varphi_0 e^{-z/\delta}$, where $\varphi_0 \sim I_0/\delta$ is proportional to the intensity of the pump pulse I_0 , and inversely proportional to the light penetration depth δ (see Supplementary section S2). The strongly non-uniform spin excitation distributes the initial deflection among magnon modes at different k -wavenumbers, with the amplitudes given by the reciprocal space image A_k of the initial excitation set up by the penetration

208 depth (see inset Fig. 3c). As a result, the finite- k mode is expected to have spectral amplitude A_k
 209 (see Supplementary Section S2):

$$210 \quad A_k \sim \frac{I_0}{1+(k\delta)^2} \quad (3)$$

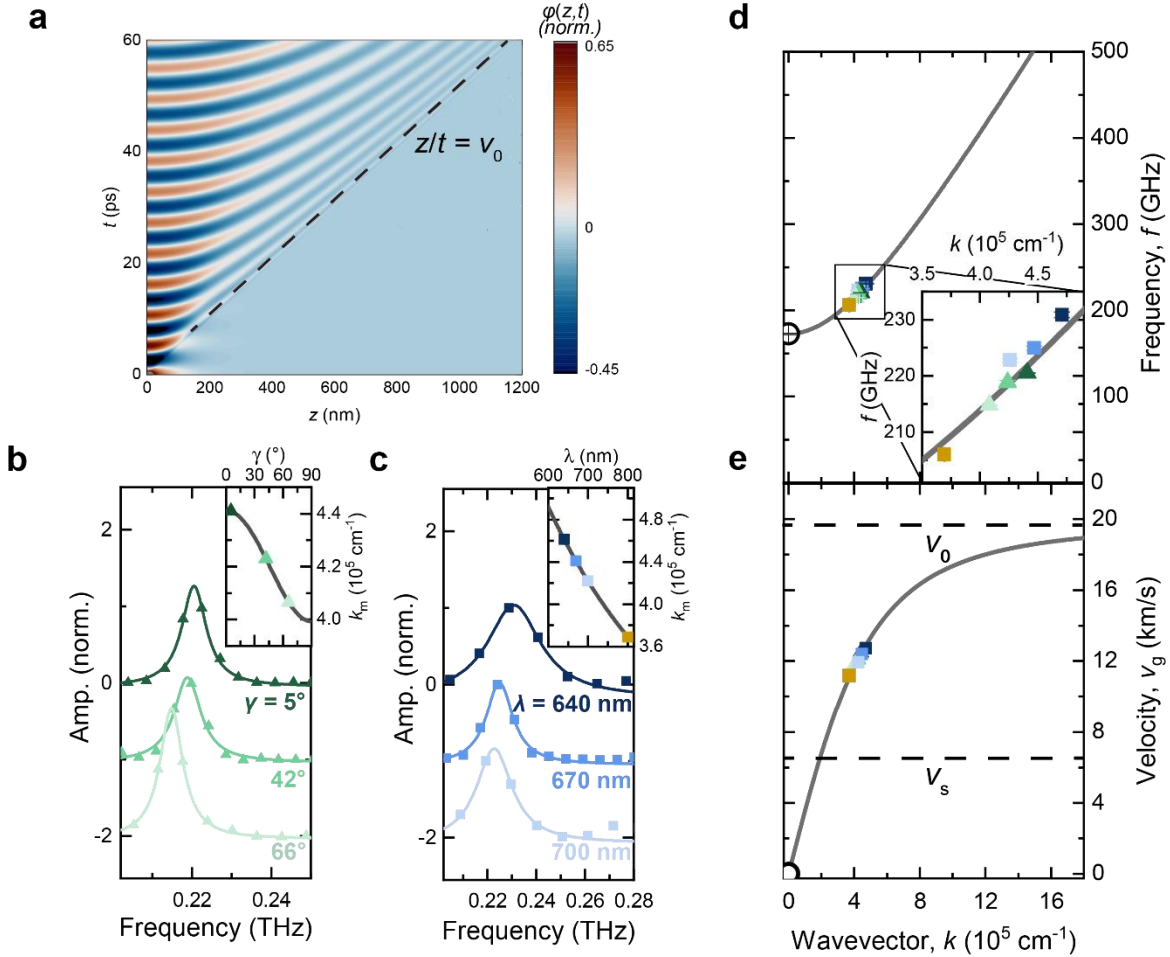
211 This expression not only agrees well with the observations of Fig. 3c ($k \approx 3.7 \times 10^5 \text{ cm}^{-1}$,
 212 $\lambda = 170 \text{ nm}$), where it is plotted as a best fit to the pump intensity I_0 , but also confirms the intuitive
 213 interpretation that a stronger confinement shifts the spectral amplitude of the excited magnon wave
 214 packet towards larger k .

215
 216 The excitation of a continuum of coherent antiferromagnetic spin waves forms a broadband
 217 magnon wavepacket, in which individual spectral components propagate independently, each
 218 adhering to the dispersion relation $\omega_k = 2\pi f_k$ (Eq. (1)). In order to visualize the time evolution of
 219 the excited magnon wavepacket, we make use of the linearized sine-Gordon equation for the space-
 220 (z) and time- (t) dependent amplitude of the spin deflections $\varphi(z,t)^{30}$. We find that the evolution of
 221 the spin dynamics is described by the following simple formula (see Supplementary section S2):

$$222 \quad \varphi(z, t) = \frac{2}{\pi} \int_{-\infty}^{\infty} dk [A_k \cos(kz) \cos(\omega_k t)] \quad (4)$$

223 By integrating this equation, under the assumption of the exponential distribution of the time-zero
 224 spin deflections and $\delta = 50 \text{ nm}$, we obtain the complete dynamics of the magnetic excitation. As
 225 shown in Fig. 4a and Extended Data Fig. 4, the strong dispersion promptly smears out the initial
 226 exponential profile of the spin excitation simultaneously forming a spin-wave front after $\sim 10 \text{ ps}$
 227 that propagates into the bulk. This front is composed by the short-wavelength magnons with
 228 $k \gtrsim 20 \times 10^5 \text{ cm}^{-1}$ propagating with the limiting group velocity v_0 .

229
 230 Applying the Bragg condition of Eq. (2), we can experimentally map out the spectral components
 231 of the excited broadband magnon wave packet, as well as determine the group velocity and
 232 propagation length of individual magnon modes. First, we vary the incidence angle γ of the probe
 233 pulse, (inset Fig. 4b) and find that the central frequency of the oscillations is



234

235 **Figure 4: Revealing spectral components of the broadband antiferromagnetic magnon wavepacket.** (a)

236 Simulation of the time- and position-dependent spin deflection $\phi(z,t)$ after optical excitation at 3.1 eV with a

237 penetration depth of 50 nm, as determined by Eq. (3). (b,c) Fourier spectra of time-resolved measurements of the

238 polarization rotation of a near-infrared probe pulse obtained in the reflection geometry after excitation with pump

239 pulses with a photon energy of 3.1 eV at the temperature $T = 60$ K for different probe incidence angles γ ($\lambda = 680$ nm)

240 (a) and probe wavelengths λ ($\gamma = 5^\circ$) (b). The solid superimposed lines are Lorentzian fits of the Fourier peaks. Insets:

241 The magnon wavenumber k_m to which the probe is sensitive, as a function of the angle γ (b) and probe wavelength λ

242 (c), with the measured points indicated by coloured markers. (d,e) The extracted central magnon frequencies (d) and

243 the calculated group velocity (e) from the data in panel (b) and (c) at their respective calculated wavenumbers plotted

244 with a best fit of the spin-wave dispersion curve (d) and the group velocity corresponding to the dispersion fit (e). The

245 marker colours and shapes correspond to the measurements in panel (b) and (c).

246

247 reduced upon increasing γ' (Fig. 4b), in perfect agreement with Eq. (2) and the magnon dispersion

248 of Eq. (1). Next, upon decreasing the probe wavelength, we observe a systematic increase in the

249 magnon frequency, as shown in Fig. 4c, once again in accordance with the Bragg condition. To

250 summarize our observations, we plot the extracted central frequencies as a function of the
 251 corresponding wavenumbers k_m (see Fig. 4d). These points are fit to the dispersion relation given
 252 by Eq. (1), with the spin-wave gap ω_0 defined by the experimentally obtained zone-center magnon
 253 frequency. The best fit of the experimental data yields a limiting group
 254 velocity $v_0 = 19.7 \pm 0.1$ km/s. This value stands in good agreement with the limiting group
 255 velocity of ~ 20 km/s extracted from the speed limit of the magnetic domain walls in DyFeO₃, as
 256 reported in Ref. [30]. Using the extracted value of the limiting group velocity we evaluate the
 257 group velocities $v_g = \frac{\partial \omega_k}{\partial k} |_{k=k_m}$ of the optically detected magnons given by $v_g = v_0^2 \frac{k_m}{\omega_k}$. These
 258 values, shown in Fig. 4e, indicate that while the zone-center magnons do not support propagation,
 259 the shortest-wavelength components of the magnon wavepacket detected in our experiment
 260 ($\lambda = 125$ nm) propagate at a supersonic ($v_s = 6$ km/s, see Supplementary Section S3) velocity of
 261 nearly 13 km/s. We note that magnons with these wavelengths already approach the exchange
 262 wave regime characterized by the limiting group velocity v_0 . This remarkable feature, inherent to
 263 antiferromagnets, stands in sharp contrast with the situation in ferromagnets, where the quadratic
 264 dispersion relation dictates that the exchange value of the group velocity is reached only for
 265 magnons with $\lambda_m \lesssim 10$ nm. Although the shortest magnon wavelength detected in our experiments
 266 is 125 nm, magnons at even shorter wavelengths, down to the penetration depth limit of 50 nm,
 267 are anticipated, and could be detected using probe pulses at higher photon energies or other means
 268 to measure non-local ultrafast spin excitations³³⁻³⁵. Using the extracted lifetime of the oscillations
 269 $\tau = 85$ ps (see Extended Data Fig. 5), we estimate the coherence length l_c of the spin-wave transport
 270 $l_c = v_g \tau = 1.1$ μ m. We note that this length, dramatically enhanced as compared to metallic
 271 antiferromagnets^{8,36}, also agrees with studies of diffusive spin transport in other insulating
 272 antiferromagnets^{9,37}. One can anticipate even longer propagation lengths for the coherent (ballistic)
 273 regime reported here: our estimate of the coherence length is only a lower limit, as the propagating
 274 spin wave is likely to escape from the region that is probed by the reflected probe light ($\sim \lambda/2$).
 275 These striking observations make antiferromagnetic insulators such as DyFeO₃ a promising
 276 platform for the realization of high-speed wave-based magnonic devices.

277

278 By optical pumping of above-bandgap electronic transitions, we have explored an efficient and
 279 virtually universal route to launch coherent propagating spin waves in insulating antiferromagnets.
 280 The strong optical absorption provides an opportunity to spatially confine the light to a

281 subwavelength scale, inaccessible by any other means, e.g. focusing³⁸⁻⁴⁰, enabling the emission of
282 a broadband continuum of short-wavelength antiferromagnetic magnons. The universal
283 mechanism opens up prospects for terahertz coherent AFM magnonics and opto-spintronics⁷
284 providing a long-sought source of coherent high-velocity spin waves. We anticipate even higher
285 propagation velocities to be observed in the broad class of easy-plane antiferromagnets (e.g.
286 hematite³⁷ and FeBO₃), in which the spin-wave gap ω_0 is reduced and the high-velocity exchange
287 wave regime can be achieved at significantly smaller wavenumbers k . The demonstrated approach
288 holds promise for a wide range of fundamental studies exploiting the excitation and propagation
289 of non-linear spin waves such as magnetic solitons^{12,41} as well as the investigation of the giant
290 magneto-elastic coupling between antiferromagnetic magnons and acoustic phonons⁴² directly in
291 the time-domain.

292

293 **References**

294

- 295 1 Kruglyak, V. V. & Hicken, R. J. Magnonics: Experiment to prove the concept. *J. Magn.*
296 *Magn. Mater.* **306**, 191-194 (2006).
- 297 2 Kruglyak, V. V., Demokritov, S. & Grundler, D. Magnonics. *J. Phys. D* **43**, 264001 (2010).
- 298 3 Lenk, B., Ulrichs, H., Garbs, F. & Münzenberg, M. The building blocks of magnonics.
299 *Phys. Rep.* **507**, 107-136 (2011).
- 300 4 Chumak, A. V., Vasyuchka, V. I., Serga, A. A. & Hillebrands, B. Magnon spintronics. *Nat.*
301 *Phys.* **11**, 453-461 (2015).
- 302 5 Sander, D. *et al.* The 2017 magnetism roadmap. *J. Phys. D* **50**, 363001 (2017).
- 303 6 Jungwirth, T., Marti, X., Wadley, P. & Wunderlich, J. Antiferromagnetic spintronics. *Nat.*
304 *Nanotechnol.* **11**, 231-241 (2016).
- 305 7 Němec, P., Fiebig, M., Kampfrath, T. & Kimel, A. V. Antiferromagnetic opto-spintronics.
306 *Nat. Phys.* **14**, 229-241 (2018).
- 307 8 Baltz, V. *et al.* Antiferromagnetic spintronics. *Rev. Mod. Phys.* **90**, 015005 (2018).
- 308 9 Lebrun, R. *et al.* Tunable long-distance spin transport in a crystalline antiferromagnetic
309 iron oxide. *Nature* **561**, 222-225 (2018).
- 310 10 Li, J. *et al.* Spin current from sub-terahertz-generated antiferromagnetic magnons. *Nature*
311 **578**, 70-74 (2020).

- 312 11 Vaidya, P. *et al.* Subterahertz spin pumping from an insulating antiferromagnet. *Science*
313 **368**, 160-165 (2020).
- 314 12 Galkina, E. & Ivanov, B. Dynamic solitons in antiferromagnets. *Low. Temp. Phys.* **44**, 618-
315 633 (2018).
- 316 13 Giamarchi, T., Rüegg, C. & Tchernyshyov, O. Bose–Einstein condensation in magnetic
317 insulators. *Nat. Phys.* **4**, 198-204 (2008).
- 318 14 Johansen, Ø., Kamra, A., Ulloa, C., Brataas, A. & Duine, R. A. Magnon-mediated indirect
319 exciton condensation through antiferromagnetic insulators. *Phys. Rev. Lett.* **123**, 167203
320 (2019).
- 321 15 Bunkov, Y. M. *et al.* High- T_c spin superfluidity in antiferromagnets. *Phys. Rev. Lett.* **108**,
322 177002 (2012).
- 323 16 Takei, S., Halperin, B. I., Yacoby, A. & Tserkovnyak, Y. Superfluid spin transport through
324 antiferromagnetic insulators. *Phys. Rev. B* **90**, 094408 (2014).
- 325 17 Qaiumzadeh, A., Skarsvåg, H., Holmqvist, C. & Brataas, A. Spin superfluidity in biaxial
326 antiferromagnetic insulators. *Phys. Rev. Lett.* **118**, 137201 (2017).
- 327 18 Dąbrowski, M. *et al.* Coherent transfer of spin angular momentum by evanescent spin
328 waves within antiferromagnetic NiO. *Phys. Rev. Lett.* **124**, 217201 (2020).
- 329 19 Kimel, A. *et al.* Ultrafast non-thermal control of magnetization by instantaneous
330 photomagnetic pulses. *Nature* **435**, 655-657 (2005).
- 331 20 Duong, N., Satoh, T. & Fiebig, M. Ultrafast manipulation of antiferromagnetism of NiO.
332 *Phys. Rev. Lett.* **93**, 117402 (2004).
- 333 21 Kampfrath, T. *et al.* Coherent terahertz control of antiferromagnetic spin waves. *Nat.*
334 *Photonics* **5**, 31-34 (2011).
- 335 22 Bossini, D. *et al.* Macrospin dynamics in antiferromagnets triggered by sub-20
336 femtosecond injection of nanomagnons. *Nat. Commun.* **7**, 1-8 (2016).
- 337 23 Afanasiev, D. *et al.* Ultrafast control of magnetic interactions via light-driven phonons.
338 *Nat. Mater.*, 1-5 (2021).
- 339 24 Afanasiev, D. *et al.* Control of the ultrafast photoinduced magnetization across the Morin
340 transition in DyFeO₃. *Phys. Rev. Lett.* **116**, 097401 (2016).
- 341 25 Usachev, P. *et al.* Optical properties of thulium orthoferrite TmFeO₃. *Phys. Solid State* **47**,
342 2292-2298 (2005).

343 26 Thomsen, C., Grahn, H. T., Maris, H. J. & Tauc, J. Surface generation and detection of
344 phonons by picosecond light pulses. *Phys. Rev. B* **34**, 4129 (1986).

345 27 Hortensius, J. R., Afanasiev, D., Sasani, A., Bousquet, E. & Caviglia, A. D. Ultrafast strain
346 engineering and coherent structural dynamics from resonantly driven optical phonons in
347 LaAlO₃. *npj Quantum Mater.* **5**, 1-6 (2020).

348 28 Afanasiev, D., Zvezdin, A. & Kimel, A. Laser-induced shift of the Morin point in
349 antiferromagnetic DyFeO₃. *Opt. Express* **23**, 23978-23984 (2015).

350 29 Balbashov, A., Volkov, A., Lebedev, S., Mukhin, A. & Prokhorov, A. High-frequency
351 magnetic properties of dysprosium orthoferrite. *Zh. Eksp. Teor. Fiz. [Sov. Phys. JETP]* **88**,
352 974-987 (1985).

353 30 Bar'yakhtar, V. G., Ivanov, B. & Chetkin, M. V. Dynamics of domain walls in weak
354 ferromagnets. *Sov. Phys. Usp.* **28**, 563-588 (1985).

355 31 Zvezdin, A. K. & Kotov, V. A. *Modern magneto-optics and magneto-optical materials*.
356 (CRC Press, 1997).

357 32 Demokritov, S. O., Hillebrands, B. & Slavin, A. N. Brillouin light scattering studies of
358 confined spin waves: linear and nonlinear confinement. *Phys. Rep.* **348**, 441-489 (2001).

359 33 Qiu, H. *et al.* Ultrafast spin current generated from an antiferromagnet. *Nat. Phys.*, 1-7
360 (2020).

361 34 Rzdolski, I. *et al.* Nanoscale interface confinement of ultrafast spin transfer torque driving
362 non-uniform spin dynamics. *Nat. Commun.* **8**, 1-5 (2017).

363 35 Melnikov, A. *et al.* Ultrafast transport of laser-excited spin-polarized carriers in
364 Au/Fe/MgO (001). *Phys. Rev. Lett.* **107**, 076601 (2011).

365 36 Siddiqui, S. A. *et al.* Metallic antiferromagnets. *J. Appl. Phys.* **128**, 040904 (2020).

366 37 Lebrun, R. *et al.* Long-distance spin-transport across the Morin phase transition up to room
367 temperature in the ultra-low damping single crystals of the antiferromagnet α -Fe₂O₃ *Nat.*
368 *Commun.* **11**, 6332 (2020).

369 38 Hashimoto, Y. *et al.* All-optical observation and reconstruction of spin wave dispersion.
370 *Nat. Commun.* **8**, 1-6 (2017).

371 39 Satoh, T. *et al.* Directional control of spin-wave emission by spatially shaped light. *Nat.*
372 *Photonics* **6**, 662-666 (2012).

- 373 40 Au, Y. *et al.* Direct excitation of propagating spin waves by focused ultrashort optical
374 pulses. *Phys. Rev. Lett.* **110**, 097201 (2013).
- 375 41 Bonetti, S. *et al.* Direct observation and imaging of a spin-wave soliton with p -like
376 symmetry. *Nat. Commun.* **6**, 1-6 (2015).
- 377 42 Ozhogin, V. & Preobrazhenskiĭ, V. Anharmonicity of mixed modes and giant acoustic
378 nonlinearity of antiferromagnetics. *Sov. Phys. Usp.* **31**, 713-729 (1988).

379 **Methods**

380

381 **Sample.**

382 A single crystal of DyFeO₃, 60 μm thick, grown by a floating zone melting technique was used
383 in this work. The sample is cut perpendicularly to the crystallographic z-axis.

384

385 **Time-resolved experiment.**

386 The experimental setup is schematically shown in Extended Data Figure 1.

387 An amplified 1 kHz Ti:Sapphire laser system (Astrella, Coherent, central wavelength 800 nm,
388 pulse energy: 7 mJ, pulse duration: 100 fs) forms the basis of the experimental setup. A large
389 fraction of this output is used to pump a dual optical parametric amplifier (OPA, TOPAS-Twins,
390 Light Conversion). The OPA delivers linearly polarized, 100 fs output pulses, with photon
391 energies $\hbar\omega$ in the range 0.45-1 eV ($\lambda = 2.7 - 1.4 \mu\text{m}$). The photon energy of these output pulses
392 was doubled or tripled using a β -barium borate (BBO) single crystal in order to obtain tunable
393 excitation pulses which cover the photon energies in the optical range of 1.5-3.1 eV (wavelength
394 400-800 nm). A small portion of the amplifier pulses was sent through a mechanical delay line
395 and used as probe of the spin dynamics in the reflection and transmission geometries.

396 Pump and probe pulse were focused onto the DyFeO₃ sample (pump spot diameter: 300 μm,
397 typical fluence 2 mJ/cm², probe spot diameter: 80 μm), which was kept in a dry-cycle cryostat
398 (Montana Instruments) that allowed to cool it down to 10 K and vary the temperature with high
399 stability in a wide temperature range (10-250 K). The pump-induced changes in the polarization
400 $\theta_{K,F}$ of the reflected or transmitted probe pulse were measured using an optical polarization
401 bridge (Wollaston prism) and a pair of balanced Si photodetectors.

402

403 **Experimental determination of the absorption coefficient.**

404 The unpolarised absorption spectrum of DyFeO₃ was directly obtained with light propagating
405 along the crystal z-axis in the spectral region 1-2.2 eV. The resulting absorption is shown in
406 Figure 1b. In addition, we performed spectroscopic ellipsometry measurements using a
407 Woollam M5000 ellipsometer over a wide energy range to obtain the real and imaginary parts
408 of the refractive index. In the photon energy region 2.5-4 eV, where the transmission
409 measurements are not possible for thick samples, we estimated the absorption using the
410 acquired complex refractive index. These values are shown in Figure 1b.

411

412

413

414 **Data Availability:**

415 All data needed to evaluate the conclusions in the paper are present in the paper and/or
416 Supplementary Materials. The source data for figures are publicly available with identifier
417 (DOI) 10.5281/zenodo.4716539

418 **Code Availability:**

419 The code used to simulate the magnon dynamics is available upon reasonable request.

420

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431

432 **Author contributions:**

433 D.A. and A.D.C. conceived the project. J.R.H., D.A. and M.M performed the experiments,
434 analysed the data. B.A.I. developed the general theoretical framework describing the spin wave
435 propagation. R.L and R.V.M. developed the theoretical formalism of the spin wave detection.
436 B.A.I., R.C., R.V.M. and A.V.K. contributed to discussion and theoretical interpretation of the
437 results. A.D.C. supervised the project. The manuscript was written by J.R.H., D.A. and A.D.C.,
438 with feedback and input from all co-authors.

439

440 **Competing interests:**

441 The authors declare no competing interests.

442

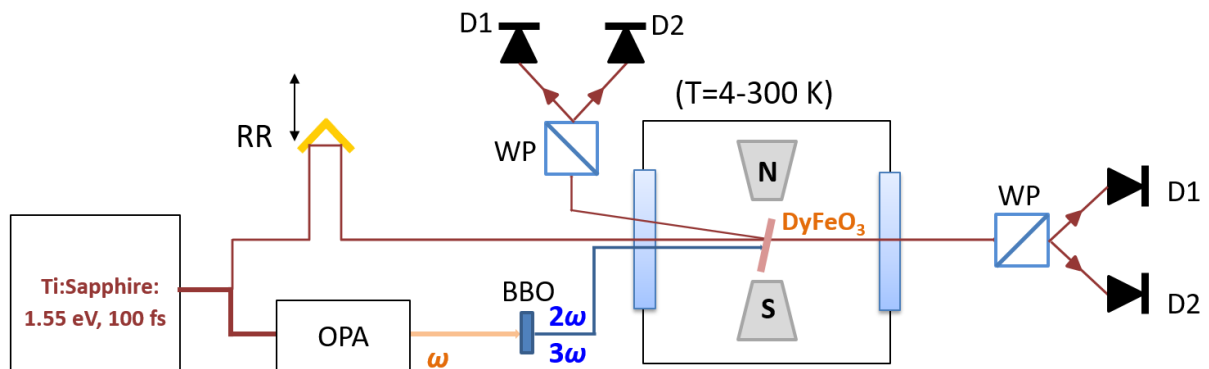
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445 **Extended Data Figures**

446

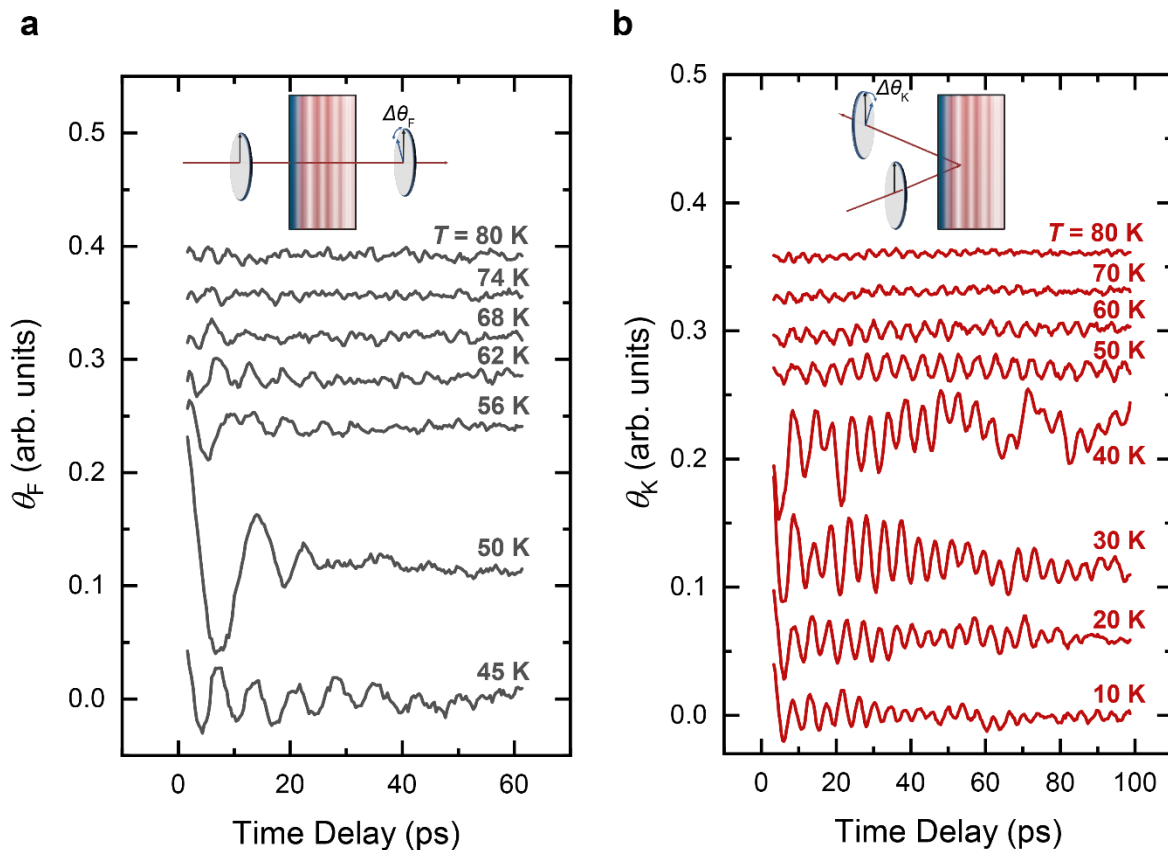
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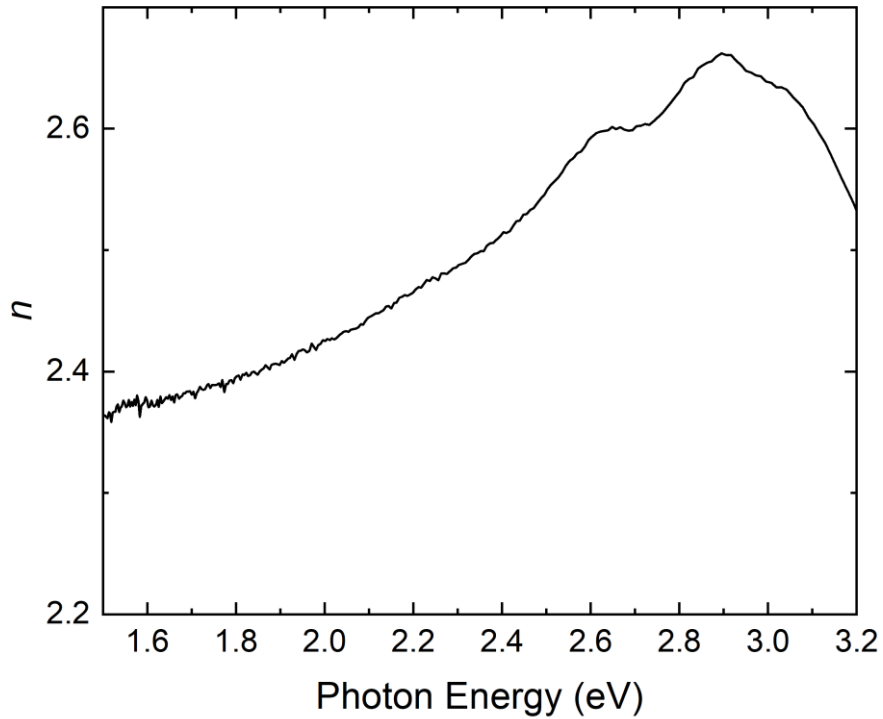
449 **Extended Data Figure 1: Experimental setup.** RR: gold retroreflector mounted on a motorized precision delay
 450 stage, OPA: optical parametric amplifier, BBO: β -barium borate crystal, WP: Wollaston Prism, D1, D2: pair of
 451 balanced silicon photodetectors.

452



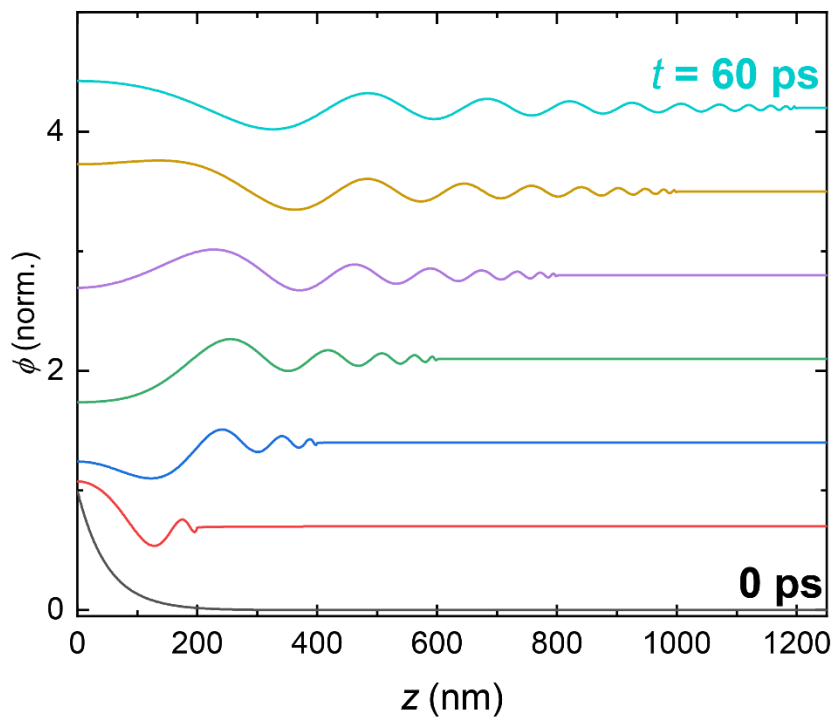
453

454 **Extended Data Figure 2: Magnon time traces at different temperatures.** (a,b) Time resolved polarization
 455 rotation in the transmission (a) and reflection geometry (b) following excitation at $h\nu = 3.1$ eV at different
 456 temperatures. The probe incidence angle is near-normal, with $\lambda = 640$ nm and $\lambda = 700$ nm for the measurements
 457 in panel a and b, respectively.



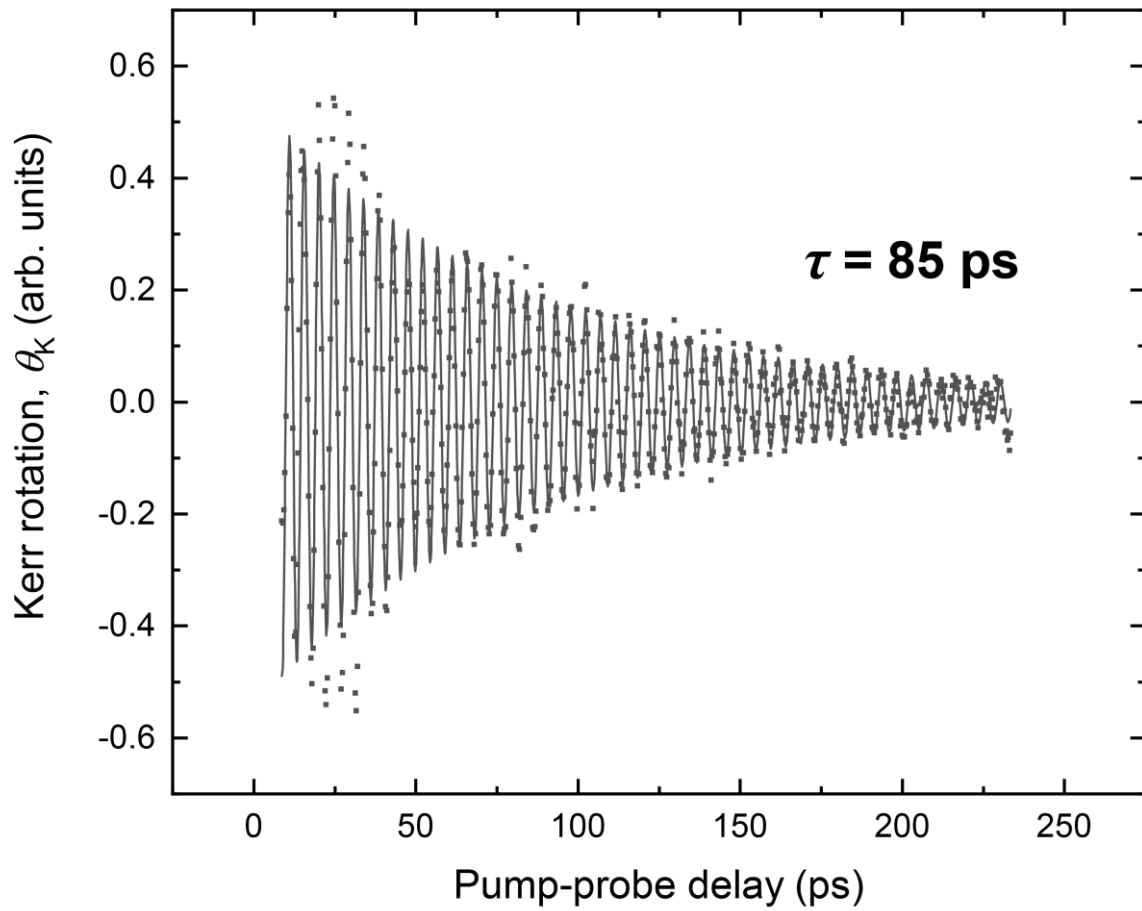
458

459 **Extended Data Figure 3: Real part of the refractive index as a function of the pump photon energy.** Real
 460 part n of the refractive index, as extracted using spectroscopic ellipsometry measurements.



461

462 **Extended Data Figure 4: Simulations of the light-induced spin wave dynamics.** Real-space distribution of
 463 the magnon spin deflection at different times t , after optical excitation at $h\nu = 3.1$ eV with a penetration depth of
 464 50 nm, as determined by Eq. (3).



465

466 **Extended Data Figure 5: Extracting the magnon propagation distance.** Time-resolved polarization rotation
 467 originating from a propagating magnon, as obtained in the reflection geometry. The solid line represents a best fit
 468 of a damped sine, giving a lifetime of ~ 85 ps. With the largest estimated group velocities v_g of the measured
 469 magnons of about 13 km/s, this gives a propagation distance $l_c = v_g \tau = 1.1 \mu\text{m}$.

470