Controlling the anisotropy of a van der Waals antiferromagnet with light

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trigonal distortion perpendicular to this plane (see Fig. 1B). Below the Néel temperature, $T_N = 155$ K, the magnetic moments of Ni$^{2+}$ ions arrange into a complex compensated antiferromagnetic pattern. The pattern is formed by zigzag ferromagnetic spin chains along the $a$ axis, which are coupled antiferromagnetically within the single layer (28) (see Fig. 1A). A large spacing, $c = 6.63$ Å, between adjacent layers leads to a negligible orbital overlap between the magnetic ions of different layers, thereby suppressing interlayer exchange such that the antiferromagnetic order acquires a 2D character already in the bulk form.

The orientation of magnetic moments in NiPS$_3$ is governed by a biaxial magnetocrystalline anisotropy consisting of two distinct contributions: a dominant easy-plane anisotropy that locks the orientation of the spins to a magnetic plane (xy), slightly inclined from the crystallographic $ab$ plane, and a secondary weaker anisotropy that orients the spins in the magnetic (xy) plane along the $a$ axis. Microscopically, the easy-plane anisotropy develops as a result of a zero-field splitting [$D \approx -1.1$ meV (29)] of the $^3A_{2g}$ ground state of the Ni$^{2+}$ ion ($S = 1$) in the crystal field of the trigonally distorted NiS$_6$ octahedra (see Fig. 1B). Note that $^3A_{2g}$ is an orbital singlet and alone cannot develop the splitting. The splitting and anisotropy arise indirectly as a consequence of spin-orbit-driven intermixing of the ground state with the first excited orbital triplet state $^3T_{2g}$, which is split by the trigonal lattice distortion (29, 30) into a set of low-symmetry doublets $^3E_g$ and singlets $^3A_{1g}$ separated by an energy gap of around 110 meV, as schematically shown in Fig. 1B. Although there are no reports on the origin of the in-plane magnetic anisotropy along the $a$ axis in NiPS$_3$, it likely stems from a rhombic distortion of the NiPS$_3$ octahedra, which further splits the $^3A_{2g}$ levels. Hence, an anisotropic Hamiltonian considering not only the axial distortion of the octahedron but also an in-plane distortion may be needed to take this observation into account (see section S6).

The orbital resonances in NiPS$_3$ correspond to a pair of $d$-$d$ transitions $^3A_{2g} \rightarrow ^3T_{2g}$ and $^3A_{2g} \rightarrow ^3T_{1g}$ emerging within the $^3F$ ground state multiplet of the Ni$^{2+}$ ion split by the octahedral crystal field ($O_h$) (see fig. S1). In NiPS$_3$, these transitions result in a pair of two broad optical absorption bands centered at 1.07 ($^3A_{2g} \rightarrow ^3T_{2g}$) and 1.73 eV ($^3A_{2g} \rightarrow ^3T_{1g}$). Note that in contrast to other transition metal ions, the $d$-$d$ resonances in Ni$^{2+}$ are spin-parity allowed ($\Delta S = 0$), i.e., they do not involve a spin-flip, and thus cannot directly affect the exchange interaction between adjacent spins. To selectively address these resonances, we used ultrashort (~100 fs) pump pulses with photon energy tunable in a broad spectral range of 0.1 to 1.9 eV. The pump-induced dynamics were measured by tracking the intensity $I$ and the rotation of the polarization plane $\theta$ of time-delayed copropagating near-infrared probe pulses at a photon energy of 1.55 eV, as schematically shown in Fig. 1C. Whereas $I$ is considered as a measure of the nonmagnetic components of the dielectric tensor, $\theta$ is sensitive to the magnetic order via magneto-optical effects, such as the Faraday effect and magnetic linear birefringence.

The sample was cooled down to 10 K, well below $T_N$, and pumped using linearly polarized pulses at variable photon energies. The time-resolved polarization rotation $\theta$ reveals a notable sensitivity of the pump-induced dynamics to the photon energy of the excitation (see Fig. 1, D and E). When excited at the $^3A_{2g} \rightarrow ^3T_{2g}$ resonance ($h\nu = 1.8$ eV), $\theta$ displays a damped oscillation as a function of the pump-probe time delay $\Delta t$, with frequency $f_1 = 0.30$ THz (see fig. S2 for the Fourier spectra). No coherent oscillations were observed
when exciting at the higher photon energy ($h\nu = 0.97$ eV) corresponding to the $^3A_{2g} \rightarrow ^3T_{1g}$ resonance. Detuning the photon energy below the absorption lines of the resonances ($h\nu = 0.8$ eV) shows no signal associated with the frequency $f_1$ but reveals instead another higher-frequency mode at $f_2 = 0.92$ THz. We note that the oscillations seen in the polarization rotation dynamics were not observed in the probe intensity dynamics $I$ (see fig. S3), thus hinting at their magnetic origin. In addition, we found no match for the frequencies of these oscillations in the phonon spectrum of NiPS$_3$, which was well studied in recent years (26, 31–33).

To understand the significance of the orbital resonances, we tracked the ultrafast dynamics while varying the pump photon energies across the subgap states down to the phonon Reststrahlen band edge at 0.1 eV. The amplitudes of both oscillations at $f_{1,2}$ were retrieved, and their relative values (normalized on the pump fluence) were plotted as a function of the pump photon energy. Figure 2A shows that the $f_2$ mode is excited in the broad window of optical transparency 0.1 to 0.9 eV, indicating the off-resonant character of the excitation and impulsive stimulated Raman scattering (ISRS) process as a plausible mechanism. In notable contrast, the excitation of the lower-frequency $f_1$ mode only occurs in a relatively narrow photon energy range, showing a pronounced resonance with the $^3A_{2g} \rightarrow ^3T_{2g}$ transitions (see Fig. 2A). The lineshape of the resonance reveals a fine structure indicative of the trigonal splitting of the $^3T_{2g}$ manifold ($^3E_g$-$^3A_{1g}$) (see fig. S1). Despite a nearly order of magnitude stronger optical absorption, no oscillations were seen upon resonant pumping of the $^3A_{2g} \rightarrow ^3T_{1g}$ higher-energy orbital resonance, underscoring the exceptional sensitivity of the oscillations to the photoexcitation of the $^3T_{2g}$ states. Whereas the amplitude of the mode at $f_2$ reveals a linear dependence on the pump fluence (Fig. 2B), the amplitude of the $f_2$ mode saturates above 5 mJ/cm$^2$ (Fig. 2C), indicating a possible saturation of the $^3A_{2g} \rightarrow ^3T_{2g}$ transition.

The temperature ($T$) dependence of the frequencies $f_{1,2}$ evidences that these modes are sensitive to the magnetic ordering. Figure 3A shows that as $T$ increases, the damping of the first mode goes up, while the frequency $f_1$ gradually decreases and ultimately converges to zero at a temperature close to $T_N$. Although the application of a relatively weak in-plane magnetic field $H$ up to 7 kOe produced no observable shift in $f_1$ (see fig. S4), the observation of the critical softening is a strong indication that the oscillation is of magnetic origin (34, 35). The softening can be characterized by a power law $f_1(T) \propto (T_N - T)^\beta$ (see Fig. 3C and fig. S5), with $T_N \approx 155 \pm 1$ K, in full agreement with literature data, and a critical exponent $\beta = 0.23 \pm 0.01$ valid down to the temperature $T = 0.65 \pm 0$ K (see fig. S5). Note that the $\beta$ value also matches remarkably well with the critical exponent of the XY model ($\beta_{XY} = 0.23$) previously proposed to describe the temperature evolution of the 2D magnetic ordering in NiPS$_3$ (36–38). This remarkable observation is an unambiguous and, at the same time, unexpected indication of the intrinsically 2D character of the mode observed in the bulk form of NiPS$_3$.

The temperature evolution of the higher-frequency oscillation at $f_2$ is substantially different. As $T$ increases, the central frequency $f_2$ shows a slight increase, which above 75 K is followed by a steep, nearly linear, softening. A linear extrapolation of the frequency decrease versus $T$ suggests that a complete softening of the mode occurs at $T = 170$ K, in proximity to $T_N$ (see Fig. 3D). The softening indicates that the oscillation is either of magnetic origin itself or strongly sensitive to the magnetic ordering. This is further corroborated by the significant growth of the damping constant upon heating. Such highly damped behavior is typical for soft modes in the vicinity of their associated phase transitions (39).

We now analyze the spin dynamics possible in NiPS$_3$ from a phenomenological theory perspective. Two magnon modes are expected in a compensated antiferromagnet, featuring a biaxial magnetic anisotropy (40, 41). The modes correspond to orthogonal deflections of the Néel vector defined as $\mathbf{L} = S(S_1 - S_2)$, where $S = S(T)$ is the average value of the Ni$^{2+}$ spin, and $S_{1,2}$ is a pair of antiferromagnetically coupled spins. In equilibrium, $\mathbf{L}$ is oriented along the $x$ axis, and deflections are expected in ($\parallel$) and out of ($\perp$) the magnetic easy plane (xy), in such a way that the dynamical components $\Delta L_x$ and $\Delta L_z$ emerge (see Fig. 3, E and F). The frequencies $f_{\parallel,\perp}$ of the magnons are defined by the geometric mean of the respective magnetic anisotropy ($D_{\parallel,\perp}$) and exchange energy $J_{xx}$ (see section S6) and, in addition, proportional to $S(T)$. Hence, both should experience a power-law temperature scaling inherent to the magnetic order parameter $L$ similarly to the one observed for the $f_1$ mode. Because the out-of-plane anisotropy is typically more substantial for easy-plane antiferromagnets such as NiPS$_3$, $f_{\perp} \ll f_1$ is expected. Note that although there is no net magnetization in the ground state: $M = S(S_1 + S_2) = 0$, a finite magnetization component $M \propto \mathbf{L} \cdot \mathbf{L}$ emerges due to the dynamics of the Néel vector $\mathbf{L}$ (42). As a consequence, the in-
out-of-plane magnetic modes can be fully described by the orthogonal pairs \((L_y, M_z)\) and \((L_z, M_y)\), respectively.

In (43), it was recently shown that the application of an in-plane magnetic field larger than \(H_{sf} = 100 \text{ kG}\) promotes a spin-flop transition in NiPS\(_3\), during which the spins suddenly rotate in the easy-plane and, in addition, cant along the field orientation. It can be easily shown (see section S7) that the magnitude of the spin-flop field \(H_{sf}\) is a direct measure of the frequency of the in-plane dynamics \(f_\parallel = \gamma H_{sf} = 280 \text{ GHz}\), where \(\gamma = 28 \times 10^{-4} \text{ GHz/G}\) is the gyromagnetic ratio. This estimate agrees particularly well with \(f_1\) and thus provides another strong indication that the coherent oscillation excited upon resonant pumping of the \(^3A_{2g} \rightarrow ^3T_{2g}\) transition is the in-plane magnon mode characterized by \(L_y\) and \(M_z\) and a 2D critical scaling. We note that even though there is an oscillating out-of-plane magnetic component \(M_z\), the experimentally observed oscillations show a strong phase and amplitude dependence on the orientation of the probe polarization plane with respect to the crystal axes, indicating that the detection of this mode is given by...
linear magnetic birefringence due to the $L_y$ component rather than Faraday rotation, sensitive to $M_z$ (44).

Having identified $f_\parallel = f_1$, we put forward the assumption that the higher-frequency oscillation at $f_2$ can be assigned to the complementary out-of-plane magnon ($f_\perp = f_2$). Our phenomenological theory (see section S8) suggests that excitation of the out-of-plane magnon mode with linearly polarized light is possible in NiPS$_3$ due to the low-symmetry (monoclinic) distortion of the crystal lattice. However, these assumptions do not agree with the recently reported, although mutually conflicting, values for the zone-center magnon at the significantly higher frequencies of 1.69 and 2.4 THz from (45) and (25), respectively. To unambiguously establish the origin of the coherent mode $f_2$, time-resolved measurements in high magnetic fields, $H \geq H_{\text{sf}}$, are of primary importance.

To further our understanding of the excitation mechanism of the in-plane ($f_1$) magnon and its relation to the light-induced magnetic anisotropy, we varied the orientation of the pump polarization plane, set by the azimuthal angle $\phi$ (see Fig. 4A). Although the optical absorption at the $3A_{2g} \rightarrow 3T_{2g}$ orbital resonance is nearly independent of $\phi$, the amplitude and phase of the induced magnetic oscillations are strongly affected by variation of the angle. Figure 4B shows that the amplitude of the excited magnon follows a clear $\pi$-periodic sinusoid with maxima corresponding to the polarization oriented at $\pm 45^\circ$ with respect to the orientation of the Néel vector $L$. This dependency can be simply understood: The linearly polarized light incident at normal to the $(ab)$ crystal plane promptly induces a magnetic anisotropy axis, directed along the orientation of the pump polarization plane. The axis breaks the magnetic symmetry in the basal plane $(xy)$, providing an in-plane magnetic torque sufficiently short to impulsively trigger the planar motion of the spins (see Fig. 4C). The validity of this scenario is further supported by a phenomenological theory based on symmetry considerations and general principles of light-matter interactions in a magnetic medium (see section S8).

**DISCUSSION**

The azimuthal dependence of the oscillation amplitude on the orientation of the pump polarization allows us to attribute the excitation of the in-plane magnon mode to an ultrafast light-induced change of the magnetocrystalline anisotropy. This anisotropy emerges in response to resonant optical excitation of the Ni$^{2+}$ electrons to the $3T_{2g}$ orbital state, characterized by an unquenched net angular momentum. To estimate the lifetime of the photoinduced anisotropy, we performed a time-resolved analysis of the frequency of the spin precession at various fluences of the incident pump. A long-living...
Crystals of NiPS$_3$ were grown by chemical vapor transport. First, polycrystalline NiPS$_3$ was synthesized by mixing powders of S (99.998%, from Sigma-Aldrich), P (> 99.99%, from Sigma-Aldrich), and Ni (99.99%, from Sigma-Aldrich) in a stoichiometric ratio, pressed into a pellet, and sealed in a quartz ampoule (P $\approx$ 5-10$^{-5}$ mbar, length = 50 cm, internal diameter = 1.5 cm). The ampoule was kept at 400°C for 20 days and cooled down slowly (0.07°C/min). Next, the previous material was mixed with iodine (99.999% anhydrous beads from Sigma-Aldrich; [12] $\approx$ 5 mg/cm$^3$), sealed in an evacuated quartz ampoule (P $\approx$ 5-10$^{-5}$ mbar, length = 50 cm, internal diameter = 1.5 cm), and placed in a three-zone furnace in a gradient of temperatures of 700/650/675°C for 28 days. Phase and compositional purity were verified by powder x-ray diffraction and inductively coupled plasma optical emission spectrometry. The materials were handled inside an argon glove box to avoid any possible oxidation. Exact details about temperature gradients and characterization of crystals from the same batch as the ones used in this work can be found in (6).

The pump pulses at the photon energies of 0.8 to 1.9 eV ($\sim$100 fs) were obtained using an optical parametric amplifier (OPA), and to access energies below 0.4 eV ($\sim$200 fs), we used difference frequency generation by mixing the outputs of two OPAs in a GaSe crystal (52). The pump pulses at a 500-Hz repetition rate were focused on the sample surface to a spot with a diameter of 200 μm. The time-delayed copropagating near-infrared probe pulses at a photon energy of 1.55 eV were focused to a spot of 130 μm, such that the spatial overlap between pump and probe pulses was satisfied.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/23/eabf3096/DC1

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Controlling the anisotropy of a van der Waals antiferromagnet with light

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