Gate-tunable spin waves in antiferromagnetic atomic bilayers

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Remarkable properties of two-dimensional (2D) layer magnetic materials, including spin filtering in magnetic tunnel junctions and gate control of magnetic states, have been recently demonstrated1-12. Whereas these studies have focused on static properties, dynamic magnetic properties such as excitation and control of spin waves have remained elusive. Here we investigate spin-wave dynamics in antiferromagnetic CrI$_3$ bilayers using an ultrafast optical pump/magneto-optical Kerr probe technique. Monolayer WSe$_2$ with strong excitonic resonance is introduced on CrI$_3$ to enhance optical excitation of spin waves. We identify sub-terahertz magnetic resonances under an in-plane magnetic field, from which the anisotropy and interlayer exchange fields are determined. We further show tuning of antiferromagnetic resonances by tens of gigahertz through electrostatic gating. Our results shed light on magnetic excitations and spin dynamics in 2D magnetic materials, and demonstrate their potential for applications in ultrafast data storage and processing.

Spin waves are propagating disturbances in magnetic ordering in a material. The quanta of spin waves are called magnons. The rich spin-wave phenomena in magnetic materials have attracted fundamental interest and impacted on technology of telecommunication systems, radars, and potentially also low-power information transmission and processing. The main magnetic materials of interest have so far been ferromagnets. The operation speed of ferromagnet-based devices is limited by the ferromagnetic (FM) resonance frequency, which is typically in the GHz range. One of the major attractions of antiferromagnets, a class of much more common magnetic materials, is the prospect of high-speed operation. The antiferromagnetic (AF) resonances are in the frequency range of as high as THz because of the spin-sublattice exchange. The antiferromagnets, however, are difficult to access due to the absence of macroscopic magnetization.

The recent discovery of two-dimensional (2D) layered magnetic materials15-17, particularly A-type antiferromagnets, which are antiferromagnetically coupled FM layers15, presents new opportunities to unlock the properties of antiferromagnets. With fully uncompensated FM surfaces, the magnetic state can be easily accessed and controlled18. These materials can be easily integrated into heterostructures with high-quality interfaces19. Their atomic thickness allows the application of strong electric field and large electrostatic doping to control the magnetic properties. Although rapid progress has been made in both fundamental understanding...
and potential applications, spin dynamics, including basic properties such as magnetic resonances and damping, have remained elusive in these atomically thin materials.

Here we investigate spin-wave excitations in bilayer CrI$_3$, a model A-type antiferromagnet, using the time-resolved magneto-optical Kerr effect (MOKE). Magnetic resonances have been recently studied in bulk layered magnets including CrI$_3$ and CrCl$_3$ by neutron scattering and microwave absorption$^{20-22}$. But extending these conventional probes to atomically thin samples, whose typical lateral size is a few microns, is extremely difficult because of the small amount of spins. Raman spectroscopy$^{23-26}$ has been applied to few-layer CrI$_3$ and other layered magnetic materials, but accurate identification of magnetic resonances (which lie in the ultralow Raman frequencies) is challenging. The time-resolved MOKE can access small-size samples and a wide range of resonance frequencies, and is ideally suited for such a study.

The sample is a bilayer CrI$_3$/monolayer WSe$_2$ heterostructure. It is encapsulated in hexagonal boron nitride (hBN) thin layers for protection of air-sensitive CrI$_3$ (Fig. 1a, b). Whereas bulk CrI$_3$ is an FM semiconductor, bilayer CrI$_3$ is an AF semiconductor with a Néel temperature of about 45 K$^{15}$. It consists of two antiferromagnetically coupled FM monolayers with out-of-plane anisotropy. Monolayer WSe$_2$ is a direct gap nonmagnetic semiconductor with strong spin-orbit interactions$^{27}$. It has a type-II band alignment with CrI$_3$ (Fig. 1c). As we discuss below, monolayer WSe$_2$ is introduced to significantly enhance the optical pump absorption and spin-wave excitation in CrI$_3$. It also breaks the layer symmetry in bilayer CrI$_3$ to allow the detection of different spin-wave modes in the polar MOKE geometry. Figure 1d is the magnetization of the heterostructure versus out-of-plane magnetic field at 4 K measured by magnetic circular dichroism (MCD) at 1.8 eV. The AF behavior is fully consistent with the reported results for bilayer CrI$_3$.$^{15}$ The small nonzero magnetization at low fields is a manifestation of the broken layer symmetry. The sharp change of magnetization with hysteresis around 0.75 T corresponds to a spin-flip transition. It provides a measure of the interlayer exchange field $H_E$.

A pulsed laser (200-fs pulse duration) is employed for the time-resolved measurements. The heterostructure is excited by a light pulse centered around the WSe$_2$ fundamental exciton resonance (1.73 eV), and the change in CrI$_3$ magnetization is probed by a time-delayed pulse around 1.54 eV. Both the pump and probe are linearly polarized and at normal incidence. The pump-induced polarization rotation of the probe is detected. It is sensitive only to the out-of-plane component of the magnetization. We apply an in-plane magnetic field $\mu_0 H_\parallel$ ($\mu_0$ denoting the vacuum permeability) to tilt the magnetization from the easy axis and monitor the spin precession dynamics. Unless otherwise specified, all measurements were performed at 1.7 K. (See Methods for details on the sample fabrication and the time-resolved MOKE setup.)

Figure 2a displays the time evolution of the pump-induced change in MOKE of bilayer CrI$_3$ under $H_\parallel$ ranging from 0 – 6 T. For all fields, the MOKE signal shows a sudden change at time zero, followed by an exponential decay on the scale of 10’s – 100’s ps. This reflects the incoherent demagnetization process, in which the magnetic order is disturbed instantaneously by the pump pulse and slowly goes back to equilibrium. Oscillations on the MOKE signal are also instantaneous. The amplitude, frequency and damping of the oscillations evolve systematically with $H_\parallel$. 
Figure 2b is the fast Fourier transform (FFT) of the oscillatory part of the time traces after subtraction of the exponentially decaying demagnetization dynamics. At low $H_{∥}$, a resonance around 70 GHz is observed. As $H_{∥}$ increases, the resonance splits into two. The low-energy mode redshifts significantly and the high-energy one barely shifts until 3.3 T. Above this field, both modes blueshift with increasing $H_{∥}$. While the amplitude of the low-energy mode diminishes quickly, the amplitude of the high-energy mode does not depend strongly on field.

We perform a more detailed analysis of the MOKE dynamics directly in the time domain by fitting them with two damped harmonic waves (red lines, Fig. 3a). Two sample time traces (after subtraction of the demagnetization dynamics) are shown for $H_{∥}$ at 1.5 and 3.75 T. The extracted resonance frequencies, damping rates and amplitudes versus $H_{∥}$ of these two modes are summarized in Fig. 3c, 3d and Supplementary Fig. S8, respectively. The results are in good agreement with peak fittings to the FFT spectra (Fig. 2b and Supplementary Fig. S9). We first focus on the resonance frequencies. The field dependence of the modes ($\omega_h$ for high-energy mode and $\omega_l$ for the low-energy mode) shows two distinct regimes. Below about 3.3 T, the two initially degenerate modes split. While both modes soften with increasing field, the low-energy mode drops nearly to zero frequency. Above 3.3 T, both modes show a linear increase in frequency with a slope equal to the electron gyromagnetic ratio $\gamma/2\pi \approx 28$ GHz/T. The latter is characteristic of a FM resonance in high fields.

The observed field dispersion of the modes is indicative of their magnon origin with a saturation field $H_S \approx 3.3$ T (the field required to fully align the magnetization in-plane) \(^8\). The two modes are the spin precession eigenmodes of the coupled top- and bottom-layer magnetizations under $H_{∥}$ (Fig. 3b). Above $H_S$, the spins are aligned in-plane and the spin waves become FM-like. This interpretation is further supported by the temperature dependence of the resonances (Supplementary Fig. S4-6). Clear mode softening is observed with increasing temperature and the resonance feature disappears near the Néel temperature.

We briefly consider the microscopic mechanism for the observed ultrafast excitation of spin waves in bilayer CrI\(_3\) (see Methods for more discussions). The most plausible process involves exciton generation in WSe\(_2\) by the optical pump, followed by ultrafast exciton dissociation and electron transfer at the CrI\(_3\)-WSe\(_2\) interface \(^{28}\) and an impulsive perturbation to the magnetic interactions \(^{29}\) in CrI\(_3\) by the hot carriers (Fig. 1c). This picture is supported by several control experiments, particularly the pump wavelength dependence of the spin-wave amplitude, which agrees with the WSe\(_2\) absorption spectrum (Supplementary Fig. S1). We have also obtained results on a thicker CrI\(_3\) sample without WSe\(_2\) layer (Supplementary Fig. S10), in which the MOKE signal is weaker than bilayer CrI\(_3\), illustrating the importance of WSe\(_2\) enhancement. Other possible processes such as photon angular momentum transfer and thermal effects have also been considered. The lack of pump polarization dependence (Supplementary Fig. S2) and the instantaneous excitation of the spin waves (Fig. 2a) exclude these processes.

We model the field-dependent spin-wave dynamics using the coupled Landau-Lifshitz-Gilbert (LLG) equations. They describe precession of antiferromagnetically coupled top- and bottom-layer magnetizations under $H_{∥}$ (Details are provided in Methods) \(^{30}\). The effective magnetic field at each layer includes contributions from the applied field $H_{∥}$, intralayer anisotropy field $H_A$, and interlayer exchange field $H_E$. In the simple case of negligible damping and symmetric layers, the
precession eigenmode frequencies are found to be \( \omega_h = \gamma \left[ H_A(2H_E + H_A) + \frac{2H_E - H_A}{2H_E + H_A} H_{||}^2 \right]^{1/2} \),

\[
\omega_l = \gamma \left[ H_A(2H_E + H_A) - \frac{H_A}{2H_E + H_A} H_{||}^2 \right]^{1/2} \text{ (before saturation)}, \quad \omega_h = \gamma \sqrt{H_{||}(H_{||} - H_A)}
\]

\[
\omega_l = \gamma \sqrt{(H_{||} - 2H_E)(H_{||} - 2H_E - H_A)} \text{ (after saturation)}. \]

The low-energy mode \( \omega_l \) corresponds to the net moment oscillations along the applied field (the \( y \)-axis). It drops to zero at the saturation field \( H_S = 2H_E + H_A \). The high-energy mode \( \omega_h \) corresponds to the net moment oscillations in the \( x\)-\( z \) plane (Fig. 3b).

The simple solution describes the experimental result well for the entire field range (dashed lines, Fig. 3c) with \( H_A \approx 1.77 \text{ T} \) and \( H_E \approx 0.76 \text{ T} \). Since \( H_A > H_E \), a spin-flip transition is expected under a field along the easy axis (Fig. 1d) \(^{30} \). The extracted \( H_E \) is in good agreement with the observed spin-flip transition field \( H_D \approx 3.3 \text{ T} \), vertical dotted lines in Fig. 3c, d) is slightly below the reported value for bilayer CrI\(_3\) (3.8 T at 2 K) \(^8 \). This could arise from the different doping levels in different samples (Fig. 4c). The extracted \( H_A \) is smaller than the bulk value (~ 2.5 T) \(^{23} \), which is consistent with the lower magnetic transition temperature observed in atomically thin samples than in bulk samples \(^{15} \).

We examine the normalized damping rate \( (2\pi/\omega \tau) \) of the modes as a function of \( H_{||} \) in Fig. 3d. Overall, damping is higher below or near saturation. In this regime, the mode frequencies are strongly dependent on internal magnetic interactions, which are prone to local doping and strain. Inhomogeneous broadening of the magnetic resonances and spin-wave dephasing is thus likely the dominant damping mechanism. For instance, a variation in \( H_E \) at the 10% level (typical for bilayer CrI\(_3\) \(^3 \)) could account for the observed damping rate for the high-energy mode below \( H_S \). Inhomogeneous broadening also explains the seemingly higher damping for the low-energy mode near \( H_S \) since its frequency has a steeper dispersion with \( H_{||} \) near \( H_S \). Above saturation, the mode frequencies are basically determined by the applied field. Inhomogeneous broadening becomes insignificant especially in the high-field limit (e.g. at 6 T). Our observation of weak layer number dependence of damping from experiment on few-layer CrI\(_3\) (Supplementary Fig. S10) suggests that interfacial damping is also not significant. More systematic studies are required to identify the dominant damping mechanism in this regime.

Finally we demonstrate control of the spin waves by electrostatic gating using a dual-gate device (Fig. 1b). Equal top and bottom gate voltages are applied to the two symmetric gates to induce doping in the heterostructure (see Methods for details). Figure 4a shows the FFT amplitude spectra of coherent spin oscillations under an in-plane field of 2 T at different gate voltages. The high-energy mode shifts continuously from ~ 80 GHz to ~ 55 GHz when the gate voltage is varied from -13 V to +13 V (corresponding to from ‘hole doping’ to ‘electron doping’). Figure 4b shows the magnetic-field dispersion of the high-energy mode (symbols) at representative gate voltages (the low-energy mode is not studied because of its small amplitude). Similar to the zero-gate voltage case, they all show an initial redshift followed by a blueshift with increasing \( H_{||} \). The turning point (i.e. the saturation field) is varied by as much as 1 T. The mode dispersion is strongly modified by gating below saturation and remains nearly unchanged above it.

The observed mode dispersion at different gate voltages can also be described by the above simple solution of the LLG equations (solid lines, Fig. 4b). The corresponding interlayer
exchange and intralayer anisotropy fields in the model are shown in Fig. 4c as a function of gate voltage. Both decrease linearly with increasing gate voltage, with $H_A$ at a higher rate than $H_E$. Similar gate dependence for $H_E$ has been reported from the study of the spin-flip transition under an out-of-plane field in bilayer CrI$_3$. The effect can be understood as a consequence of doping dependent electron occupancy of the magnetic Cr$^{3+}$ ions and their wavefunction overlap. Increasing electron density weakens the magnetic interactions and the applied field responsible for spin precession below $H_S$. Above $H_S$, the magnetization is fully saturated in-plane. The mode frequency is mainly determined by the applied field and is nearly doping independent. However, a quantitative description of the experimental result would require ab initio calculations and is beyond the scope of the current study.

In conclusion, we have demonstrated generation, detection, and gate-tuning of spin waves in a model 2D antiferromagnet bilayer CrI$_3$. Our results allow the characterization of internal magnetic interactions and damping. The combination of the time-resolved MOKE and type-II heterostructures that facilitate ultrafast interlayer charge transfer for efficient spin-wave excitation can be applied to a broad class of magnetic thin films. Local gate control of spin dynamics may also have implications for reconfigurable spin-based devices.

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**Methods**

**Sample and device fabrication**

The measured sample is a stack of 2D materials composed of (from top to bottom) few-layer graphite, hBN, monolayer WSe$_2$, bilayer CrI$_3$, hBN, and few-layer graphite (Fig. 1b). The top
and bottom graphite/hBN pairs serve as gates. An additional stripe of graphite is attached to the WSe₂ flake for grounding and charge injection. The thickness of hBN layers is ~30 nm, and the graphite layers, about 2-6 nm. All layer materials were first exfoliated from their bulk crystals onto SiO₂/Si substrates and identified by their color contrast under an optical microscope. The heterostructure was built by the layer-by-layer dry transfer technique. It was then released onto a substrate with pre-patterned gold electrodes, which contact the bottom gate, top gate, and grounding graphite flake. The steps involving CrI₃ before its full encapsulation in hBN layers were performed inside a nitrogen-filled glovebox because CrI₃ is air sensitive.

Bulk crystals of graphite, hBN and WSe₂ were from HQ graphene. Bulk CrI₃ crystals were synthesized by chemical vapor transport following methods described in previous reports. They crystallize into the $C2/m$ space group with typical lattice constants of $a = 6.904 \, \text{Å}$, $b = 11.899 \, \text{Å}$, $c = 7.008 \, \text{Å}$ and $\beta = 108.74^\circ$, and Curie temperatures of 61 K.

In the gating experiment, equal top and bottom gate voltages of the same sign were applied to the two nearly symmetric gates. The gate voltage shown in Fig. 4 is the voltage on each gate. This geometry allows electrostatic doping into the heterostructure without introducing a vertical electric field. The total carrier density in the CrI₃-WSe₂ heterostructure can be calculated from the gate voltage and the gate capacitances. Because CrI₃ has a much higher density of states than WSe₂, majority of the gate-induced carriers goes into the CrI₃ layer. We measured the doping density in the WSe₂ layer independently by monitoring the photoluminescence energy of charged excitons (trions), which is a sensitive function of doping density. Doping density in CrI₃ was obtained by subtracting the carrier density in WSe₂ from the total carrier density. Supplementary Figure S3 shows Fig. 4c with the calibrated doping density in CrI₃ shown in the top axis.

**Time-resolved magneto-optical Kerr effect (MOKE) and magnetic circular dichroism (MCD)**

In the time-resolved MOKE setup, the probe beam is the output of a Ti:Sapphire oscillator (Coherent Chameleon with a repetition rate of 78 MHz and pulse duration of 200 fs) centered at 1.54 eV, and the pump beam is the second harmonic of an optical parametric oscillator (OPO) (Coherent Chameleon compact OPO) output centered at 1.73 eV. The time delay between the pump and probe pulses is controlled by a motorized linear delay stage. Both the pump and probe beam are linearly polarized. The pump intensity is modulated at 100 kHz by a combination of a half-wave photoelastic modulator (PEM) and a linear polarizer whose transmission axis is perpendicular to the original pump polarization. The pump and probe beam impinge on the sample at normal incidence. The reflected light is first filtered to remove the pump, passed through a half-wave Fresnel rhomb and a Wollaston prism, and detected by a pair of balanced photodiodes. The pump-induced change in Kerr rotation is determined as the ratio of the intensity imbalance of the photodiodes obtained from a lock-in amplifier locked at the pump modulation frequency and the intensity of each photodiode.

For the MCD measurements, a single beam centered at 1.8 eV is used. The light beam is modulated at 50 kHz between the left and right circular polarization using a PEM. The reflected light is focused onto a photodiode. The MCD is determined as the ratio of the ac component of
the photodiode signal measured by a lock-in amplifier at the polarization modulation frequency and the dc component of the photodiode signal measured by a voltmeter.

For all measurements samples were mounted in an optical cryostat (attoDry2100) with a base temperature of 1.7 K and a superconducting solenoid magnet up to 9 Tesla. For measurements under an out-of-plane field, the sample was mounted horizontally and light was focused onto the sample at normal incidence by a microscope objective. For measurements under an in-plane field, the sample was mounted vertically and the light beam was guided by a mirror at 45° and focused onto the sample at normal incident by a lens.

Landau-Lifshitz-Gilbert (LLG) equations
We model the field-dependent spin dynamics in AF bilayer CrI$_3$ using coupled Landau-Lifshitz-Gilbert (LLG) equations $^{30}$,

$$\frac{\partial M_i}{\partial t} = -\gamma M_i \times H_i^{\text{eff}} + \frac{\alpha}{M_S} M_i \times \frac{\partial M_i}{\partial t}. \quad (1)$$

Here $M_i \ (i = 1, 2)$ is the magnetization of the top or bottom layer (which is assumed to have a magnitude $M_S$), $\gamma/2\pi \approx 28$ GHz/T is the electron gyromagnetic ratio, $\alpha$ is the dimensionless damping factor, and $H_i^{\text{eff}}$ is the effective magnetic field at the $i$-th layer that is responsible for spin precession. In the absence of applied magnetic field, $M_1$ and $M_2$ are anti-aligned along the easy axis (z-axis). When an in-plane field $H_\parallel$ (along the y-axis) is applied, $M_1$ and $M_2$ are tilted symmetrically towards the y-axis, before fully turned into the applied field direction at the saturation field $H_S = 2H_E + H_A$. Here $H_E$ and $H_A$ are the interlayer exchange and intralayer anisotropy fields, respectively. The canting angle $\theta$ of magnetization (with respect to the anisotropy axis) is given by $\sin \theta = H_\parallel/H_S$ (Supplementary Fig. S11). The effective field has contributions from the applied field, the interlayer exchange field, and the intralayer anisotropy field $H_i^{\text{eff}} = H_\parallel - \frac{H_E}{M_S} M_{z,1} + \frac{H_A}{M_S} (M_{1,2})_z \hat{z}$. We search for solution in the form of a harmonic wave $e^{i\omega t}$ with angular frequency $\omega$. For the simple case of zero damping ($\alpha = 0$), two eigenmode frequencies ($\omega_h \geq \omega_l$) are given in the main text.

In case of finite but weak damping, we simplify the LLG equations for the high-energy and low-energy modes. Before saturation ($H_\parallel < H_S$),

$$\omega_h^2 (1 + \alpha^2) - i\alpha \omega_h \gamma \left( \frac{\omega_h^2}{2H_E + H_A} + 2H_E + H_A \right) - \omega_h^2 = 0; \quad (2)$$

$$\omega_l^2 (1 + \alpha^2) - i\alpha \omega_l \gamma \left( \frac{\omega_l^2}{H_A} + H_A \right) - \omega_l^2 = 0. \quad (3)$$

After saturation ($H_\parallel > H_S$),

$$\omega_h^2 (1 + \alpha^2) - i\alpha \omega_h \gamma (2H_\parallel - H_A) - \omega_h^2 = 0; \quad (4)$$

$$\omega_l^2 (1 + \alpha^2) - i\alpha \omega_l \gamma (2H_\parallel - 4H_E - H_A) - \omega_l^2 = 0. \quad (5)$$
Here \( \omega_{h,0} \) and \( \omega_{l,0} \) correspond to the solution at zero damping (\( \alpha = 0 \)). If \( \alpha \ll 1 \), the oscillation frequency (the real part of \( \omega_h \) and \( \omega_l \)) becomes \( \frac{\omega_0}{\sqrt{1+\alpha^2}} \), where \( \omega_0 \) is the undamped solution for the two modes. With the correction from finite but very small \( \alpha \), the eigenmode frequencies are reduced, and the two modes are no longer degenerate at zero field.

**Mechanism for ultrafast excitation of coherent magnons**

We have investigated the mechanism for the observed ultrafast excitation of magnons in bilayer CrI\(_3\). A plausible picture involves exciton generation in WSe\(_2\) by the optical pump, ultrafast exciton dissociation and charge transfer at the CrI\(_3\)-WSe\(_2\) interface, and an impulsive perturbation to the magnetic anisotropy and exchange fields in CrI\(_3\) by the injected hot carriers. For a typical pump power used in the experiment and assuming \( \sim 20\% \) WSe\(_2\) light absorption at the pump energy, we estimate a transferred charge density of \( \sim 10^{12} \text{ cm}^{-2} \) at time zero for \( 10\% \) charge transfer efficiency from WSe\(_2\) to CrI\(_3\). Such a density is on par with the electrostatic doping density that induces significant changes in the magnetic interactions (Supplementary Fig. S3).

Several control experiments were performed to test this picture. Pump-probe measurements were performed on both monolayer WSe\(_2\) and bilayer CrI\(_3\) areas alone (non-overlapped regions in the heterostructure) under the same experimental conditions. Negligible pump-induced MOKE signal was observed. Measurements were also done on the heterostructure at different pump energies. The mode frequencies were found unchanged, but the amplitudes are consistent with the absorption spectrum of WSe\(_2\) (Supplementary Fig. S1). These two experiments show that magnons are generated through optical excitation of excitons in WSe\(_2\). It has been reported that CrI\(_3\)-WSe\(_2\) heterostructures have a type-II band alignment, which can facilitate ultrafast exciton dissociation and charge transfer at the interface \(^{28}\). Moreover, the onset of coherent oscillations is instantaneous with optical excitation. This excludes lattice heating in CrI\(_3\) as a dominant mechanism for the generation of magnons, which typically takes a longer time to build up. Moreover, the resonance amplitude is independent of the pump laser polarization (Supplementary Fig. S2). It indicates that hot carriers, rather than the angular momentum of the carriers, are responsible for the excitation of magnons. These experiments are all consistent with the proposed mechanism of ultrafast excitations of magnons in CrI\(_3\)-WSe\(_2\) heterostructures.

**Temperature dependence of magnon modes**

We have performed the optical pump/MOKE probe experiment in CrI\(_3\)-WSe\(_2\) heterostructures at temperature ranging from 1.7 K to 50 K. No obvious oscillations can be measured above 50 K when bilayer CrI\(_3\) is close to its Néel temperature (\( \sim 45 \) K). The results at 1.7 K are presented in the main text. Supplementary Fig. S4 and S5 show the corresponding measurements and analysis for 25 K and 45 K, respectively. With increasing temperature, the magnon frequency decreases and the saturation field (estimated from the minimum of the frequency dispersion) decreases. A systematic temperature dependence is shown in Supplementary Fig. S6 for the high-energy mode \( \omega_h \) at a fixed in-plane field of 2 T. The frequency has a negligible temperature dependence well below the Néel temperature (\( < 20 \) K), and decreases rapidly when the temperature approaches the Néel temperature.

**Magnetic-field dependence of mode amplitudes**
The magnetic-field dependence of the high-energy and low-energy mode amplitudes (Supplementary Fig. S8) can be qualitatively understood. Under zero magnetic field, the equilibrium magnetization is in the out-of-plane direction. Assuming that the magnetization amplitudes do not change, the out-of-plane magnetization oscillation is zero for both modes. With an increasing in-plane field, spins cant towards the in-plane direction, and the out-of-plane magnetization oscillation grows till the field reaches the saturation field. At this point, the magnetizations are fully aligned in-plane. With a further increase in the in-plane field, spins stiffen and the oscillation amplitude decreases. However, many details of Supplementary Fig. S8 remain not understood. For instance, the maximum amplitude of the two modes occurs at different in-plane fields, and the amplitude of the low-energy mode decreases much faster than the high-energy mode above saturation. Future systematic studies are required to better understand the field dependence of the mode amplitudes.

Measurements on few-layer CrI3

We have measured the magnetic response from a few-layer CrI3 (6-8 layer) sample. Because of the larger MOKE signal and higher optical absorption in thicker samples, magnetic oscillations can be measured without the enhancement from monolayer WSe2. The results are shown in Supplementary Fig. S10. The comparison of results from samples of different thicknesses provides insight into the origin of magnetic damping. For instance, in the high-field limit (6 T), few-layer and bilayer CrI3 show a similar level of damping. This indicates that interfacial damping is not the dominant contributor to damping.

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Data availability

The data that support the findings of this study are available within the paper and its Supplementary Information. Additional data are available from the corresponding authors upon request.

Acknowledgments
This work was supported by the National Science Foundation under award DMR-1807810 (time-
resolved spectroscopy), the Center for Emergent Materials: an NSF MRSEC under award
number DMR-1420451 (bulk CrI₃ crystal growth and device fabrication), and the Air Force
Office of Scientific Research under award number FA9550-19-1-0390 (data analysis). This work
was also partially supported by the Cornell Center for Materials Research with funding from the
NSF MRSEC program under DMR-1719875 (optical characterization). The growth of hBN
crystals was supported by the Elemental Strategy Initiative conducted by the MEXT, Japan and
the CREST(JPMJCR15F3), JST. D.W. gratefully acknowledges the financial support by the
German Science Foundation (Deutsche Forschungsgemeinschaft, DFG) under the fellowship
number WE6480/1. X.Z. acknowledges Postdoctoral Fellowship from the Kavli Institute at
Cornell (KIC). K.F.M. acknowledges support from David and Lucille Packard Fellowship.

Author contributions
X.Z., K.F.M. and J.S. designed the study. X.Z. developed the time-resolved spectroscopy setup
and performed the measurements. L.L. fabricated the devices and assisted X.Z. in the
measurements. D.W. and J.E.G. grew the bulk CrI₃ crystals. K.W. and T.T. grew the bulk hBN
crystals. X.Z., K.F.M. and J.S. co-wrote the manuscript. All authors discussed the results and
commented on the manuscript.

Competing interests
The authors declare no competing interests.

Figure captions

Figure 1 | Bilayer CrI₃/monolayer WSe₂ heterostructures. a, Optical microscope image of the
heterostructure. Bilayer CrI₃ is outlined with a purple line, monolayer WSe₂ with a black line, and
one graphite gate layer with a gray line. The CrI₃/WSe₂ stack is encapsulated by hBN on both
sides, and graphite layers are used to electrically connect to the lithographically defined metal
electrodes (bottom and top right). The dark spots are air bubbles trapped in the 2D stack. Scale
bar is 5 μm. b, Schematic sideview of the dual-gated device employed in the gating experiment.
c, Schematic of a type-II band alignment between monolayer WSe₂ and CrI₃. Optically excited
exciton in WSe₂ is dissociated at the interface and electron is transferred to CrI₃. 28 d, Magnetic
 circular dichroism (MCD) of the heterostructure as a function of out-of-plane magnetic field
(μ₀Hₐ) at 4 K. Hysteresis is observed for field sweeping along two opposing directions. Insets
are schematics of the corresponding magnetization in the top and bottom layers of blayer CrI₃.
The dashed lines indicate the spin-flip transition around 0.75 T.

Figure 2 | Time-resolved magnon oscillations. a, Pump-induced Kerr rotation as a function of
pump-probe delay time in bilayer CrI₃ under different in-plane magnetic fields. b, Fast Fourier
transform (FFT) amplitude spectra of the time dependences shown in a after the removal of the
demagnetization dynamics (exponential decay). The black dashed lines are Voigt peak fitting of
the resonance features centered at frequencies of Fig. 3c. The curves in a and b are vertically
displaced for clarity. The keys between the panels are for both panels.
Figure 3 | Magnon dispersion and damping. a, Pump-induced magneto-optical Kerr effect (MOKE) dynamics in bilayer CrI$_3$ under two representative in-plane fields of 1.5 T (upper panel) and 3.75 T (lower panel). Grey lines are experiment after subtracting the demagnetization dynamics, and red lines, fits to two damped harmonic oscillations. b, Illustration of two spin-wave eigenmodes under an in-plane field (y-axis): the high-energy mode (left) and the low-energy mode (right). The dotted lines indicate the equilibrium top and bottom layer magnetization $M_1$ and $M_2$, which are titled symmetrically from the z-axis towards the applied field. The magnetizations precess following the green and blue arrows in the order 1 through 4. c, d, Magnetic-field dependence of frequencies (c) and damping rates (d) of the high-energy and low-energy modes extracted from the fit shown in a. The damping coefficient $\alpha = 2\pi/\omega\tau$ is normalized by the resonance frequency $\omega$. The error bars are the fit uncertainties. Dashed lines in c are fits to the Landau-Lifshitz-Gilbert (LLG) equations as described in the text. The vertical dotted lines indicate the in-plane saturation field from the fits to the LLG equations.

Figure 4 | Gate tunable magnon frequencies. a, Fast Fourier transform (FFT) amplitude spectra of the magnons as a function of gate voltage under a fixed in-plane field of 2 T. Dashed lines are Voigt peak fits. The gray line is a guide to the eye of the evolution of the mode frequency with gate voltage and triangles indicate the peak of the resonance. b, Magnetic-field dispersion of the high-energy mode at representative gate voltages. The solid lines are fits to the Landau-Lifshitz-Gilbert (LLG) equations and symbols are experimental data. Fig. S12 shows the magnetic dispersion for the measured gate voltages in a. c, Anisotropy and exchange field extracted from the fits in b at different gate voltages. Error bars are the standard deviation from the fitting. Dashed lines are linear fits.
