In this study, we chose CrSBr because of its excellent semiconducting properties\(^\text{18,19}\) and, more importantly, the observation of strong coupling of Wannier excitons in CrSBr to interlayer magnetic order\(^\text{19}\). The CrSBr crystal consists of van der Waals (vdW) layers with rectangular unit cells in the plane \((a-b)\) and stacked along the \(c\) axis to produce an orthorhombic structure. Atomically thin flakes of CrSBr can be produced by mechanical exfoliation, where the bulk magnetic structure is maintained down to the ferromagnetic (FM) monolayer with a Curie temperature \(T_C = 146\) K and to the antiferromagnetic (AFM) bilayer with a Néel temperature \(T_N = 140\) K (ref. \(\text{19}\)), which is higher than the bulk \(T_N\) of 132 K. CrSBr is also a direct-gap semiconductor down to the monolayer, with an electronic gap of 1.5 eV and an excitonic gap of 1.34 eV (ref. \(\text{20}\)). Towards the two-dimensional (2D) limit, the material can be with or without net magnetization for odd or even numbers of layers, respectively\(^\text{19}\). The co-existence of both magnetic and semiconducting properties implies that a spin wave may coherently modulate the electronic structure, which in a 2D semiconductor, is reflected in the dominant excitonic transitions\(^\text{24,25}\). Such 2D magnon–exciton coupling allows the launch and detection of spin waves from strong absorption, emission or reflection of light in the energy range corresponding to excitonic transitions. This is a major advantage over conventional methods to optically access magnons by (1) resonant excitation in microwave spectroscopy based on microwave antenna and waveguides\(^\text{13}\), (2) magnetooptical effects based on precise detection of light polarization rotation\(^\text{12,26}\) or (3) symmetry changes detected in nonlinear optical spectroscopy with high-power pulsed lasers\(^\text{12,26}\).

The recent discoveries of two-dimensional (2D) magnets\(^\text{1–6}\) and their stacking into van der Waals structures\(^\text{7–11}\) have expanded the horizon of 2D phenomena. One exciting application is to exploit coherent magnons\(^\text{12}\) as energy-efficient information carriers in spintronics and magnonics\(^\text{13,14}\) or as interconnects in hybrid quantum systems\(^\text{15–17}\). A particular opportunity arises when a 2D magnet is also a semiconductor, as reported recently for CrSBr (refs. \(\text{18–20}\)) and NiPS\(_3\) (refs. \(\text{21–23}\)) that feature both tightly bound excitons with a large oscillator strength and potentially long-lived coherent magnons owing to the bandgap and spatial confinement. Although magnons and excitons are energetically mismatched by orders of magnitude, their coupling can lead to efficient optical access to spin information. Here we report strong magnon–exciton coupling in the 2D A-type antiferromagnetic semiconductor CrSBr. Coherent magnons launched by above-gap excitation modulate the exciton energies. Time-resolved exciton sensing reveals magnons that can coherently travel beyond seven micrometres, with a coherence time of above five nanoseconds. We observe these exciton-coupled coherent magnons in both even and odd numbers of layers, with and without uncompensated magnetization, down to the bilayer limit. Given the versatility of van der Waals heterostructures, these coherent 2D magnons may be a basis for optically accessible spintronics, magnonics and quantum interconnects.
Coupling of excitons to coherent magnons. a) Detecting in-plane propagating coherent magnons of δ-CrBr$_3$. We obtain exciton energy modulation caused by coherent oscillation for the low-frequency in-phase branch (Fig. 2c), the temperature dependence of which is expected for a ferromagnetic resonance. In agreement with the magnetic resonance results (Fig. 2b), the $B_0$ dependence of the excitonic transitions in transient reflectance exhibits the same dispersion for the low-frequency in-phase branch (Fig. 2c). The temperature dependence of the magnon frequencies from magnetic resonance measurements (Supplementary Fig. 7) are in good agreement with those obtained from optical measurements (Fig. 1f).

Magnon-exciton coupling from transient reflectance

Upon photoexcitation of CrSBr, the strong oscillatory components (Fig. 1c) come from the coherent spin waves because (1) they are observed only below $T_N$ and the frequencies decrease with $T$ (Supplementary Fig. 1 and Fig. 1e discussed below), and (2) the amplitude of the oscillation signal is much weaker in the absence of a canting field (Supplementary Fig. 2) and increases with an external canting field, $B_0$. To obtain the frequencies of the coherent spin waves, we perform a fast Fourier transform (FFT) of the oscillatory response. Figure 1d shows the FFT amplitude (pseudo-colour) as a function of spin-wave frequency ($\nu_{\text{mag}}$) and $h\nu$. There are two peak frequencies at about 24 GHz and about 34 GHz, both independent of probe $h\nu$. This is expected because the coherent spin waves modulate the interlayer electronic hybridization, and thus all excitonic transitions. We use the $h\nu$-integrated FFT spectrum (inset of Fig. 1e for a typical spectrum at $T = 5\, \text{K}$), to quantify the peak frequencies: $\nu_{\text{mag}} = 24.6 \pm 0.7\, \text{GHz}$ and $\nu_{\text{mag}} = 34 \pm 1\, \text{GHz}$. With increasing $T$, the magnetic order decreases and this results in lowering of the spin-wave frequencies (Fig. 1e). Around $T_N$, both $\nu_{\text{mag}}$ and $\nu_{\text{mag}}$ approach zero. The $T$ dependences of $\nu_{\text{mag}}$ and $\nu_{\text{mag}}$ closely follow that of the magnetic-order parameter.

The strong coupling of excitons to coherent magnons is revealed by clear $\pi$-phase flips of the oscillatory signal at $h\nu$, corresponding exactly to the peaks of excitonic transitions (Fig. 1b,c). The $\pi$-phase shift is a signature of an optical transition modulated by coherent oscillation in a coupled mode, as is also known for coherent phonon–exciton coupling. Likewise, the FFT amplitudes of both $\nu_{\text{mag}}$ and $\nu_{\text{mag}}$ (red and blue spectra in Fig. 1f, respectively) track the first derivative (black) of the static reflectance spectrum. The strength of the coupling between excitons and coherent magnons can be calculated by the modulation of the exciton energy (detailed in Supplementary Information and Supplementary Figs. 3–5). We obtain exciton energy modulation caused by coherent magnons of $\delta E_{\text{ex}} = 4.0 \pm 0.5\, \text{meV}$. It is noted that this coupling is perturbative in nature, not due to resonant hybridization between two modes that differ in energies by four orders of magnitude: $E_{\text{ex}} = 1.3\, \text{eV}$ and $h\nu_{\text{mag}} = 0.10\, \text{meV}$ and $h\nu_{\text{mag}} = 0.14\, \text{meV}$ for $v_1$ and $v_2$, respectively.

Magnetic resonance spectroscopy

To support assignments of coherent magnon modes from excitonic sensing, we use magnetic resonance spectroscopy. Figure 2a shows a series of magnetic resonance spectra at selected microwave frequencies ($\nu = 5–21\, \text{GHz}$) for bulk CrSBr at $T = 5\, \text{K}$. The spectra reveal a single resonance in the low-frequency region (≤15 GHz) and two resonances in the high-frequency (>22 GHz) region. We extract peak frequencies ($\nu_{\text{mag}}$) of the resonances (Supplementary Fig. 6) and plot them as function of the magnetic field applied along the $c$ axis ($B_0$) (Fig. 2b). For $B_0$ smaller than a saturation field ($B_{\text{sat}} = 1.7\, \text{T}$), we observe two $\nu_{\text{mag}}$ branches whose frequencies decrease with increasing $B_0$, consistent with reduction in AFM order as spins are progressively canted away from the easy $b$ axis. The frequencies of these two branches are assigned to the in-phase ($\nu_\|)$ and out-of-phase ($\nu_\perp$) spin precessions, similar to those observed in the 2D AFM materials of chromium(III) chloride and chromium(III) iodide. Above $B_{\text{sat}}$, the spins are fully polarized parallel to $B_0$ and $\nu_{\text{mag}}$ increases linearly with $B_0$ (seen here for the out-of-phase magnon), which is expected for a ferromagnetic resonance. In agreement with the magnetic resonance results (Fig. 2b), the $B_0$ dependence of the excitonic transitions in transient reflectance exhibits the same dispersion for the low-frequency in-phase branch (Fig. 2c). The temperature dependence of the magnon frequencies from magnetic resonance measurements (Supplementary Fig. 7) are in good agreement with those obtained from optical measurements (Fig. 1f).

In-plane propagating coherent magnons

We exploit the efficient exciton sensing to implement time and spatially resolved spectroscopic imaging of propagating coherent magnons. We perform this with the pump and probe beam separated by a controlled...
distance, \(d\) (Fig. 3a). The diffraction-limited excitation spot can create a gradient in the driving force for launching coherent magnons in a finite momentum window at \(|k| > 0\). The propagating nature of the coherent magnons is clearly seen in the delayed rise of the oscillatory signal for \(d > 0\), as illustrated for \(d = 2\) µm along the \(a\) axis (Fig. 3b; see Supplementary Fig. 8 for spatially resolved MOKE propagation images). This delayed response is in stark contrast to the prompt rise of the magnon frequency when the pump and probe pulses overlap spatially (\(d = 0\)) in Fig. 1c. FFT analysis of the propagating waves reveals the two coherent magnon modes at 24 GHz and 34 GHz. Along the \(a\) axis, we detect two magnon frequencies (24 GHz and 34 GHz) and along the \(b\) axis, we detect only the 24-GHz component. We perform short-time Fourier transform (STFT) with respect to \(\Delta t\) for different \(d\) values for the 24-GHz and 34-GHz modes along the \(a\) axis (Fig. 3c–e) and for the 24-GHz mode along the \(b\) axis (Fig. 3d). In each case, the peak \(\Delta t\) shifts linearly with \(d\), establishing the propagating nature of these coherent waves. From these shifts, we obtain group velocities of \(V_g = 1.0 \pm 0.1\) km s\(^{-1}\) and \(V_g = 3.0 \pm 0.3\) km s\(^{-1}\) for the 24-GHz and 34-GHz modes along the \(a\) axis, respectively, and \(V_g = 0.7 \pm 0.1\) km s\(^{-1}\) for the 24-GHz mode along the \(b\) axis (Supplementary Fig. 9). From the probe-distance-dependent measurements along the \(b\) axis (Supplementary Fig. 8), we obtain lower bounds for the coherent transport lengths of \(\lambda_{coh} = 6\) µm and \(\lambda_{coh} = 7\) µm for the 24-GHz and 34-GHz modes, respectively. In complementary experiments, we carry out time-resolved imaging based on the magneto-optical Kerr effect (MOKE). Following initial excitation in the centre of the image frame, the MOKE responses expand in space with \(\Delta t\), as expected from the propagating nature of the spin waves (detailed in Supplementary Section 2). Figure 3f shows a representative early time \((\Delta t = 0.1\) ns\) MOKE image, which is discussed below. To understand the origin of propagating spin waves, we calculated magnon dispersions along high-symmetry directions (Fig. 4a,b) from linear spin-wave theory (LSWT) fits to neutron-scattering data\(^{23}\), taking into account the small interlayer spin exchange in the vertical direction (\(c\) axis) and anisotropy exchange constants from fitting the magnetic resonance spectroscopy data in Fig. 2b. The dispersions along the in-plane \(\Gamma–X\) (\(a\) axis) and \(\Gamma–Y\) (\(b\) axis) directions are determined by the strong FM exchange interaction. Along the \(\Gamma–Z\) (\(c\) axis) direction, dispersion is essentially flat on the energy scale for \(\Gamma–X\) and \(\Gamma–Y\) and becomes visible only when we zoom-in by two orders of magnitude (Fig. 4b). The weak interlayer AFM couplings give rise to vertical bandwidths of only about 70 µeV, compared with about 40 meV in the in-plane directions. From the dispersions in Fig. 4a, we obtain group velocities (\(V_g\)) along the \(\Gamma–X\) and \(\Gamma–Y\) directions (Fig. 4c,d). There are two reasons why the observed propagating spin waves cannot be attributed to their intrinsic properties at sufficiently high momentum vectors \((Q)\). First, owing to the negligible photon momentum, the optical pump–probe experiments can only probe magnons at the bottom of the dispersions, that is, near the \(\Gamma\) point. The spatial gradient may provide a parallel momentum vector of the order of about 1 \(\mu\)m\(^{-1}\), as dictated by the size of a diffraction-limited excitation. To visualize \(V_g\) values of 1–3 \(\mathrm{km\ s^{-1}}\), the magnon dispersions dictate that intrinsic spin waves in the 2D plane (Fig. 4c,d) must possess \(Q\) values greater than 1 \(\mathrm{nm^{-1}}\), three orders of magnitude higher than what is experimentally possible. Second, the calculated \(V_g\) values of the IP and OP modes are nearly identical in broad \(Q\) ranges along both the \(a\) and \(b\) axes, contrary to the experimental ratio of \(V_{g,OP}/V_{g,IP} = 3.0 \pm 0.4\) along the \(a\) axis and observation of only the IP mode along the \(b\) axis. A well-known mechanism for fast propagation of magnons at small momentum vectors is attributed to hybridization between magnon and acoustic phonons through magneto-elastic coupling\(^{33,34}\). In this mechanism, the above-gap pump pulse creates thermally induced strain in magnetic materials and launches the hybridized magnon and phonon modes. Confirmation of this coupling mechanism comes from the short-time MOKE image (Fig. 3f) with the distinct quadrupolar shape, which is a signature of coupling of magnons to longitudinal acoustic phonons\(^{33,34}\). Further support for this mechanism comes from peak splitting in the frequency domain (Supplementary Fig. 10), which can be directly attributed to hybridization and the resulting avoided crossing between the two modes. The two different group velocities...
the same conditions as that in Fig. 1c. There is a clear π-phase flip in the studied system attributed to spin waves. Figure 5a shows spectra for 5L CrSBr obtained under the same conditions as those in the bulk crystal, the transient reflectance spectra from two monolayer because it lacks interlayer exciton coupling. Similar to our findings on the bulk crystal, the transient reflectance spectra from two monolayer flakes. It is noted that this approach cannot be used to probe the FM interlayer magnetic order. The coupling of coherent magnons, living presumably originate from crossing between two different acoustic phonon branches with different dispersions in momentum space.

Coherent magnons down to the bilayer limit

The detection of coherent magnons in CrSBr from simple exciton sensing allows us to extend the measurements to the 2D limit on exfoliated flakes. It is noted that this approach cannot be used to probe the FM monolayer because it lacks interlayer exciton coupling. Similar to findings on the bulk crystal, the transient reflectance spectra from two layers (2L) to five layers (5L) feature coherent oscillations attributed to spin waves. Figure 5a shows spectra for 5L CrSBr obtained under the same conditions as that in Fig. 1c. There is a clear n-phase flip in the oscillatory signal at the exciton peak of about 1.362 eV owing to the strong coupling of coherent magnons to the exciton. Figure 5b shows a line cut at $h\nu = 1.359$ eV for the longest delay within our experimental limit, $\Delta t = 7.6$ ns. The coherent oscillation clearly persists beyond the experimental time window. A Lorentzian fit to its FFT (Supplementary Fig. 11) gives a full-width at half-maximum of $0.20 \pm 0.02$ GHz, corresponding to the coherence time, $\tau_{coh} = 5$ ns. Similar measurements for 2L–4L are shown in Supplementary Figs. 12–15. Supplementary Fig. 16 shows a quantitative analysis of coherence time from the 5L sample. These nanosecond coherence times are similar to those in the bulk crystal, $\tau_{coh} = 2.5\text{–}5.0 \pm 0.5$ ns. It is noted that the measured $\tau_{coh}$ and $k_{coh}$ represent lower bounds in the intrinsic coherence times and lengths, as the measurement with finite excitation pulse width, gradient in excitation profiles and inhomogeneity in sample environment introduces extrinsic decoherence.

Figure 5c summarizes the dependence of the spin-wave frequencies on the layer number. With increasing thickness, the frequency of the in-phase mode gradually decreases by about 10% (from 24.0 $\pm$ 1.0 GHz to 24.6 $\pm$ 0.7 GHz), whereas that of the out-of-phase mode increases by about 10% (from 31.1 $\pm$ 1.0 GHz to 34.1 $\pm$ 1.0 GHz). Figure 5d plots the modulation in exciton energies, $\delta E_{exc}$, by the coherent magnons for layer thicknesses ranging from 3L to thin bulk. The lower signal-to-noise ratio from the 2L sample prohibits a quantitative analysis of $\delta E_{exc}$. Within experimental uncertainty, the magnon–exciton coupling energies from 3L to 5L (Supplementary Fig. 14) are the same as that from the bulk, $\delta E_{exc} = 4.0 \pm 0.5$ meV. An attractive attribute of the coherent magnons approaching the 2D material limit is that they can have net magnetization for an odd (not even) number of layers. Having a net magnetization is essential for application as a quantum interconnect

Summary and prospects

Our findings of exciton-coupled coherent magnons represent several key advances and may be applicable to vdW layered magnetic semiconductors in general, as long as the 2D excitons are of the Wannier type and interlayer electronic coupling is sufficiently influenced by interlayer magnetic order. The coupling of coherent magnons, living
in the sub-millielectronvolt energy range, to excitons living in the electronvolts region, can enable direct coupling between the exciton and magnon. This coupled exciton–magnon also offers an opportunity to control the exciton properties of CrSBr by magnons, or vice versa, using optical or microwave cavities and electrostatic gating. In addition, the efficient exciton–magnon coupling opens the door to exciting applications, including integration with optoelectronics using on-chip light-emitting diodes and photodetectors. In this context, our observations of coherent magnons with excellent coherent properties down to the 2D bilayer limit suggest that CrSBr could be combined with other 2D materials through vdW heterojunctions, leading to novel device architectures and applications.

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focus on how the latter modulates the former. The coherent magnons

... above on exciton-coupled coherent magnons...
the easy axis is in the $b$ direction. Solving this equation for the magnon modes using the Holstein–Primakoff formalism\(^{31}\) gives the formulae in the main text for the two low-energy modes. It is noted that these modes are the acoustic intraplane magnon mode, split by the presence of interplane exchange and magnetic anisotropy. It is also noted that the presence of an intermediate axis is crucial to the observation of two modes at zero applied magnetic field: in a single easy-axis case, $A_x = A_y$, the two $B = 0$ modes are degenerate.

We quantitatively analyse the $B_0$ dependence of the magnon frequencies using the Heisenberg spin Hamiltonian and fitting the magnetic resonance spectroscopy data using LSWT\(^{32}\), with the fitting details shown in Supplementary Fig. 22 (Supplementary Section 13). The triaxial anisotropy in spin-exchange interaction and interlayer-exchange interaction in CrSBr results in two non-degenerate magnon modes. The LSWT analysis gives the in-phase mode at frequency $\nu_{IP} = 2S_{z}(A_x + A_yf_{int})$ and the out-of-phase mode at frequency $\nu_{OP} = 2S_{z} (A_x + A_yf_{int})$, where $S$ is the total spin quantum number. The $A_x$, $A_y$ and $f_{int}$ values from LSWT fits (solid curves in Fig. 2b) are 14 µeV, 58 µeV and 6 µeV, respectively. The fits give magnon frequencies at $B_0 = 0.2$ T (external field used in Fig. 1) of $\nu_{IP} = 24.4$ GHz and $\nu_{OP} = 33.8$ GHz, in good agreement with those measured from exciton sensing. Similar agreement with experimental results is also achieved in analysis based on the classical LL equations\(^{31}\), with the spin configuration shown in Supplementary Fig. 23 (Supplementary Section 14).

Data availability
The data that support the plots within this paper are available from the corresponding author upon reasonable request. Source data are provided with this paper.

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Author contributions X.Z. and Y.J.B. conceived this work. Y.J.B. carried out all optical measurements at fixed magnetic fields with assistance from J.W., Y.B. and M.D. Bulk crystals were synthesized and characterized by D.G.C. and M.E.Z. under the supervision of A.D.K. The magnetic-field-dependent optical measurements were performed by G.M.D. and J.C. under the supervision of X.X. The theoretical analysis was performed by Y.J.B. and A.S. X.Z. supervised the project. The manuscript was prepared by Y.J.B. and X.Z. in consultation with all other authors. All authors read and commented on the manuscript.

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Additional information

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