Interlayer electronic coupling on demand in a 2D magnetic semiconductor

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When monolayers of two-dimensional (2D) materials are stacked into van der Waals structures, interlayer electronic coupling can introduce entirely new properties, as exemplified by recent discoveries of moiré bands that host highly correlated electronic states and quantum dot-like interlayer exciton lattices. Here we show the magnetic control of interlayer electronic coupling, as manifested in tunable excitonic transitions, in an A-type antiferromagnetic 2D semiconductor CrSBr. Excitonic transitions in bilayers and above can be drastically changed when the magnetic order is switched from the layered antiferromagnetic ground state to a field-induced ferromagnetic state, an effect attributed to the spin-allowed interlayer hybridization of electron and hole orbitals in the latter, as revealed by Green’s function–Bethe–Salpeter equation (GW-BSE) calculations. Our work uncovers a magnetic approach to engineer electronic and excitonic effects in layered magnetic semiconductors.

One of the most exciting prospects of 2D materials is their stacking into natural or artificial van der Waals (vdW) structures, in which interlayer electronic coupling can result in new and emergent properties. For example, interlayer hybridization turns the momentum direct bandgaps in transition metal dichalcogenide (TMDC) monolayers to indirect bandgaps in bilayers or multilayers1–5, introduces moiré bands that host highly correlated phenomena in twisted graphene6–9 and TMDC10–12 bilayers and results in the formation of periodic arrays of potential traps for excitons in TMDC heterobilayers13–15. In these examples, interlayer hybridization is predeterminated and in limited cases can it be tuned, for example, with mechanically rotatable structures16, with hydrostatic pressure17 and with vertical electric fields18–20. Here we explore in situ tuning of interlayer electronic hybridization based on the control of layered magnetic order. A-type antiferromagnets are ideal materials for this purpose: they consist of vdW ferromagnetic (FM) monolayers that are coupled antiferromagnetically along the stacking direction. Such interlayer antiferromagnetic (AFM) order can be switched to FM with an external magnetic field, often accompanying a change of properties and symmetry. Tuning the spin structures of layered antiferromagnets has thus led to a number of emerging physical phenomena. Examples based on 2D CrI3 include giant tunnelling magneto-resistance via spin filtering effects18, very large second harmonic generation (SHG) from the AFM state due to magnetic state induced inversion symmetry breaking20 and tuning inelastic light scattering and spin waves via symmetry controls21. In the layered magnet MnBi2Te4, a topological quantum phase transition occurs when the intrinsic interlayer AFM state is fully polarized to the FM state with an external magnetic field22.

CrSBr: an A-type 2D AFM semiconductor

In this work, we report the unique magneto-electronic coupling effects that emerge in CrSBr, a 2D material that combines a direct electronic bandgap with layered A-type AFM order22. We examine this coupling by probing optical transitions associated with Wannier excitons that are sensitive to interlayer electronic coupling in CrSBr. The use of Wannier excitons to probe electronic-magnetic coupling in CrSBr contrasts with CrI3, where the dominant optical transitions are from localized and parity forbidden d–d orbitals centred on the Cr atom23. The lattice of CrSBr consists of vdW layers made of two buckled planes of CrS terminated by Br atoms (Fig. 1a). These layers stack along the c axis to produce an orthorhombic structure with Pmmm (D2h) space group. The mechanical exfoliation of CrSBr single crystals produces elongated flakes, a manifestation of the anisotropic structure of the material (Fig. 1b). The long and short lateral edges of the crystals correspond to the crystallographic a and b axes determined by single crystal X-ray diffraction (XRD) (Methods). Below the bulk Néel temperature (TN ≈ 132 K), each CrSBr vdW layer orders ferromagnetically and couples antiferromagnetically to adjacent layers. CrSBr possesses biaxial magnetic anisotropy, with easy and intermediate magnetic axes along the crystallographic b and a axes, respectively, and a hard axis along c (ref. 23). A recent SHG study confirmed that this magnetic structure persists to the FM monolayer and AFM bilayer24. Unlike other 2D magnets, the Néel temperature in CrSBr is found to increase with decreasing layer number, from TN = 132 K in the bulk to TN = 150 K in the bilayer24.

Electronic structure calculations of monolayer CrSBr in its FM ground state within the GW approximation reveal a semiconducting bandgap of roughly 1.8 eV and highly anisotropic band dispersion; see the GW band structure of monolayer in Fig. 1c and density functional theory (DFT) band structures of one, two and three layers in Supplementary Fig. 1. The valence band maximum (VBM) is at the Γ point, and two nearly degenerate conduction band minima (CBM) appear at the Γ and X points. Despite having a different quasiparticle bandgap and dispersion, our GW calculations that include
the self-energy corrections are in agreement with previous DFT calculations that also predict anisotropic and spin-polarized bands.25–27 Away from Γ, there is considerable anisotropy of the conduction bands, with dispersion along Γ−Y and almost flat bands along Γ−X. This feature is consistent with the calculated optical matrix element that is dipole-allowed along the b axis, but forbidden along the a axis, for interband transitions from the VBM to the CBM at the Γ point (further discussions of the optical selection rules can be found in Supplementary Fig. 1d). Similar to those in TMDGs,35 the dominant optical transitions come from Wannier excitons in CrSBr (below), with the calculated exciton binding energy of roughly 0.5 eV for a free-standing monolayer. For comparison, the exciton binding energy estimated from a comparison of scanning tunneling spectroscopy and photoluminescence (PL) spectrum is roughly 0.25 eV for a bulk crystal.37 The larger exciton binding energy of the monolayer than that of the bulk is expected from the reduced dielectric screening in the former.

The anisotropic optical transition is captured in polarization-resolved optical spectroscopy on exfoliated CrSBr flakes. Figure 1d shows the linear polarization-resolved differential reflectance (ΔR/R) spectra of bilayer CrSBr. The lowest-energy excitonic transition at 1.34 eV is visible only with light polarized along the b axis, with no detectable exciton peak for polarization along the a axis. The same polarization dependence is seen for the higher energy exciton peak at 1.75 eV, which can be attributed to the transition to the CBM from the second valence band located at roughly 0.4 eV below the VBM (Fig. 1c). The behaviour in the reflectance spectrum of the lowest-energy excitonic transition is mirrored in the PL spectra. The bright PL emission at 1.34 eV is observed with near-unity linear polarization (red curves in Fig. 1e) and this optical anisotropy is present for all thicknesses of CrSBr flakes, regardless of magnetic order (below or above the Néel temperature, Supplementary Fig. 2a–j). A comparison in Fig. 1e of the polarization-resolved PL spectra (red) and differential reflectance spectra (blue) underlines the excellent agreement between the position of the excitonic peaks, which is consistent with a direct bandgap transition. Note that the excitonic transition shows an asymmetric shape; a possible reason could be the involvement of phonon-assisted transitions of momentum indirect excitons due to the nearly flat conduction band in the Γ−X direction.

**Evidence for magneto-electronic coupling**

Evidence for electronic-magnetic order coupling is first found in the temperature dependence of the PL spectrum. For monolayer CrSBr (Supplementary Fig. 3a,e), the PL spectrum only shows a gradual blueshift of the emission with decreasing temperature as expected for a direct-gap semiconductor. For bilayer CrSBr (Fig. 1f), the blueshift is accompanied by a change in the peak shape across the magnetic phase transition, as evidenced by a kink at 150 ± 5 K, which closely matches the bilayer T_N determined by SHG35. The magneto-electronic coupling becomes even more evident for thicker flakes (Supplementary Fig. 3b–d,f–h) as we discuss below.

To gain a deeper understanding of the magneto-electronic coupling in CrSBr, we measured PL as a function of applied magnetic field (B) at 5 K and establish that the magneto-excitonic coupling originates from interlayer electronic interaction. For monolayer CrSBr, the PL spectrum shows no discernible evolution as B is swept along the easy (Fig. 2a) or hard axes (Supplementary Fig. 4a).

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**Fig. 1 | Structure and optical properties of CrSBr.** a, Crystal and magnetic structures of CrSBr. The top image shows a top view of a single layer (1L) and the bottom image shows a side view of a bilayer (2L) in which the AFM order is represented by the red arrows. b, Optical microscope image of exfoliated CrSBr. Scale bar is 5 μm. 3L, three layer and 4L, four layer. c, Calculated quasiparticle band energies using the GW method for monolayer CrSBr. The bands of majority- and minority-spin electrons are shown in red and black, respectively. d, Two-dimensional pseudo-colour (−ΔR/R) plot of differential reflectance of bilayer CrSBr as functions of light wavelength (x axis) and angle of in-plane polarization with respect to the b axis. White dashed lines represent polarization along a and b axes. e, Comparison of differential reflectance spectra (blue, right axis) and PL spectra (red, left axis) with polarization along the b (solid) and a (dashed) axis, respectively, from bilayer CrSBr. The inset shows PL intensity versus polarization angle. f, PL spectra from bilayer CrSBr, which coincides with a change in PL peak shape. Measurements in d and e are carried out at a sample temperature of T = 5 K.
In stark contrast to the monolayer results, the PL response of the bilayer varies dramatically with $B$. Figure 2b presents bilayer PL spectra as $B$ is swept along the easy axis. The PL redshifts and intensifies abruptly above a critical field $B_c = 0.134 \pm 0.003 \text{T}$ and is otherwise constant above and below this transition (Supplementary Fig. 4b). A comparison of monolayer and bilayer PL spectra at selected $B$ along the $b$ axis can be found in Supplementary Fig. 5. Field sweeps along the intermediate and hard axes result in a continuous evolution of the PL spectrum up to saturation fields $B_{sat}$ of roughly 0.9 and 1.6 T, respectively, beyond which the PL spectrum remains unchanged (Fig. 2c,d and Supplementary Fig. 4c,d). This spin-canting process is corroborated by reflectance magnetic circular dichroism (RMCD) measurements with $B$ along the $c$ axis (Fig. 2e). The $B_{sat}$ values for bilayer measured by RMCD and inferred from magneto-PL (Fig. 2c) are identical. Similar switching (along $b$) and canting (along $a$ or $c$) behaviours with external magnetic field are also observed in the differential reflectance spectra of the bilayer for both excitonic resonances (Supplementary Figs. 6 and 7).

The above results demonstrate that the dramatic change of excitonic properties arises from the tuning of interlayer magnetic order. When $B$ is along the easy axis, the abrupt switch in the PL spectra of the bilayer at $B_c$ comes from a spin flip transition (that is, transition from AFM to FM order), resulting in a sudden transformation of the electronic structure and excitonic transitions. When $B$ is along the intermediate or hard axes, spin canting produces continuous changes to the interlayer magnetic order, concurrent with the continuous evolution of the electronic structure and the PL spectra. As we show below, a GW-BSE calculation reproduces the redshift of the exciton from AFM to FM, and a perturbation picture predicts a quadratic energy shift in $B$ below $B_{sat}$ as confirmed in Fig. 1f for spin canting when $B$ is applied to the intermediate axis.

Theory reveals the origin of magneto-electronic coupling

We use first-principles GW-BSE calculations (details in Methods) to obtain the quasiparticle band structures of bilayer CrSBr in the AFM (Fig. 3a) and FM (Fig. 3b) states. In the AFM bilayer, the product symmetry of time reversal and spatial inversion makes the band structure degenerate in spins. In each Bloch band near the doubly degenerate CBM and VBM, the spin-up or down electrons are localized at the top and bottom layers, respectively, since their interlayer hybridization is suppressed by the interlayer AFM order. In the FM bilayer, by contrast, the electrons in the two layers can resonantly couple with each other, leading to band splitting of the CBM and VBM and a band gap reduction of roughly 0.1 eV relative to the AFM bilayer.

The GW-BSE calculations unveil the nature of the optical transitions. The dominant optical transitions arise from the lowest-energy exciton in CrSBr bilayers, with the calculated exciton binding energies of roughly 0.46 and 0.37 eV for free-standing AFM and FM bilayers, respectively. In the AFM bilayer, the bright excitons in the top and bottom layers are virtually decoupled due to the anti-aligned spins between layers. The lowest-energy bright excitons are twofold degenerate (with energy difference <0.5 meV), and account for the optical transitions at roughly 1.34 eV in the experimental spectra. A comparison between the experimentally measured transition energy and the calculated exciton excitation energy (around 1.23 eV) shows a roughly 0.1 eV difference, which arises from the error of GW-BSE calculations (details in Methods). The calculated exciton wavefunction (top view in Fig. 3c) extends over several unit cells, and is about two to three times more delocalized along the $b$ axis than that along the $a$ axis, revealing an anisotropic Wannier character. The side view of the exciton wavefunction in Fig. 3d confirms that the electron is localized in the same layer as the hole in the AFM bilayer. In the FM bilayer, by contrast, the
Magnetic order-dependent band structure and excitonic transitions. a,b, Band structures of the AFM (a) and FM (b) CrSBr bilayers. Blue and red dots are Kohn–Sham band energies calculated using the DFT-Perdew–Burke–Ernzerhof method and quasiparticle band energies calculated using the GW method, respectively. In the AFM bilayer, bands are degenerate in spin. In the FM case, the bands of majority spins are shown (those of minority spins have much larger gaps). c–e, Real-space wavefunction of the lowest-energy exciton, shown as $|\psi_n|_2$ for an electron with the hole fixed near a Cr atom (green circle) in the bottom layer. The iso-surfaces represent $|\psi_n|_2^2$ set at 1% of its maximum. c, Top view in the AFM bilayer. Top view in the FM bilayer is very similar. d,e, The side view in the AFM bilayer showing that the electron is virtually localized in the same layer as the hole (d) and side view in the FM bilayer showing that the electron wavefunction is delocalized across both layers for a hole (green circle) fixed in the bottom layer (e). f, Calculated optical absorption spectra of linearly polarized light (polarization along the $b$ axis) of the AFM (blue) and FM (red) CrSBr bilayers. A Gaussian broadening of 0.005 eV is used to model the imaginary part of the dielectric function. For polarization along the $a$ axis, absorbance goes to zero. The two spectra are normalized to the peak absorbance in the AFM state. g, Experimentally measured differential reflectance spectra of the AFM (blue) and FM (red) CrSBr bilayers.

Bright excitons in the two layers are no longer decoupled states due to the interlayer hybridization of the electron and hole orbitals. For the lowest-energy exciton in the FM bilayer, when the hole is fixed in one layer, the associated electron shows amplitude in both layers (shown in Fig. 3c with a typical side view). We further quantify interlayer spatial distribution of the exciton wavefunction by first integrating the wavefunction module square in the top or bottom layer, with the hole fixed to a given position in the bottom layer (Fig. 3d,e), and then summed over all the possible hole positions in the bottom layer (see Supplementary Information for details). The sum of the exciton wavefunction module square in the top layer is <1% of that in the bottom layer in the AFM bilayer, and roughly 50% of that in the bottom layer in the FM bilayer. In other words, the probability density of finding the bound electron in the layer different from that of the fixed hole is >50 times larger in the FM state than the AFM state. The calculated optical spectra in the AFM and FM states are shown in Fig. 3f. By turning on the interlayer electronic coupling in the FM state, the most visible effect is a redshift of the optically bright exciton by roughly 10 meV from that in the AFM state. This calculated redshift is consistent with the experimental measurements, as shown by the differential reflectance spectra from bilayer CrSBr in the AFM (blue) and field-induced FM (red) states, respectively (Fig. 3g). The same redshift is seen in the magnetic field-dependent PL spectra in Fig. 2b–d.

The interlayer hybridization explains the stark difference between the discrete switching behaviour of the excitonic transitions when the magnetic field is along the easy axis (Fig. 2b) and the continuous evolution (Fig. 2c,d) when it is along the intermediate/hard axis. The wavefunctions near the VBM or CBM of each CrSBr layer can be approximated by the product of the spatial and the spinor parts, with the spatial part nearly independent of the spin orientation. The interlayer hopping integral $t_{ij}$ is therefore proportional to the inner product of the spinor wavefunctions of adjacent layers, $t_{ij} \propto \langle S_i | S_j \rangle = \cos(\theta/2)$, where $\theta$ is the angle between the magnetization vectors of the layers. For the spin flip transition, $\theta$ jumps from $\pi$ to zero at $B_{c1}$, while for the spin-canting behaviour, $M = M_{sat} \cos(\theta/2) \propto B$ up to $B_{c2}$. When $B$ is along the intermediate axis, lowest-order energy shift in the perturbation theory gives $\Delta E \propto t_{ij}^2 \propto B^2$, as is confirmed in Fig. 2f. Moreover, the interlayer hybridization across the vdW interface makes relaxation into the lowest-energy bright exciton more efficient and, thus, more competitive with non-radiative recombination. This explains the abrupt increase in PL intensity across the spin flip transition (Fig. 2b) and the gradual increase in PL intensity as the spins are progressively canted away from the easy axis (Fig. 2c,d).

Multi-step magneto-electronic switching in thicker layers

Our interpretation indicates that thicker layers should give rise to intermediate excitonic transitions due to transitional magnetic states between the AFM and the fully polarized FM orders. To identify these different magnetic states, we performed PL and differential reflectance measurements as a function of $B$ on three- and four-layer CrSBr flakes. We focus on four-layer data in Fig. 4 and the three-layer data are summarized in Supplementary Fig. 8. The temperature dependence of the PL spectrum of four-layer CrSBr (Fig. 4a) shows that the broad PL peak observed at high temperature resolves into two narrower branches below $T_{c1} = 139$ K. This splitting is consistent with our understanding: the excitons are strongly...
coupled between different layers in the high temperature paramagnetic phase due to interlayer hybridization, and become localized in individual layers in the AFM phase. We expect the layer-localized excitons to experience different environments, such as local dielectric screening depending on the layer in which they reside. Furthermore, the asymmetric dielectric environment caused by the substrate may also brighten otherwise forbidden optical transitions, for example, involving the second conduction band roughly 40 meV higher in energy (Supplementary Fig. 1a–d). These may provide an explanation for the PL peak splitting observed in the AFM phase for three-layer or thicker CrSBr flakes.

Measuring the PL and differential reflectance spectra as a function of $B$ at 5 K provides additional insights into the coupling between magnetic order and excitonic transitions. When $B$ is swept along the easy axis, the PL spectra of four-layer CrSBr change abruptly at certain critical fields, with no detectable changes between those fields (Fig. 4b). Unlike the single magnetic transition observed for the bilayer, there are two separate transitions at $B_{c1} = 0.17$ and $B_{c2} = 0.33$ T for four-layer CrSBr. These two transitions correspond to spin flip transitions of individual layers, as illustrated by the arrows in Fig. 4b and similarly observed before in few-layer CrI$_3$ (ref. 30). The PL response when $B$ is along the intermediate axis (Fig. 4c) mimics that of the bilayer (Supplementary Fig. 9 presents PL data when $B$ is along the hard axis). The PL peaks redshift with increasing $B$ and the higher energy peak vanishes when approaching $B_{c2}$ along the intermediate axis (Fig. 4d). Magneto-PL of 4L CrSBr along the easy axis, with possible magnetic configurations denoted by the red/blue arrows. Layer-flip transitions that of the bilayer (Supplementary Fig. 9 presents PL data when magnetic order-dependent excitons in four-layer CrSBr.

**Outlook**

The results presented here demonstrate an effective approach to tailor interlayer electronic coupling in vdW semiconductors by control of their magnetic order. The magnetization-dependent interlayer electronic coupling, as revealed in excitonic transitions, can be used as an ‘on/off’ or ‘rotary’ switch when an external magnetic field is applied to the easy axis of an AFM bilayer or multilayer, and as a ‘dimmer’ when the field is applied to the intermediate or hard axis. Moreover, the magneto-electronic coupling may enable simple optical means to probe or manipulate spin information, such as the launching or tracking of spin waves. Finally, the possibility of controlling interlayer twist angles in artificially stacked magnetic semiconductor bilayers or multilayers adds rich dimensions to the
burgeoning field of moiré physics, with the tantalizing prospect of controlling periodic arrays of interlayer hybridization and moiré bands by spin order and external magnetic fields.

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Methods

Sample preparation and characterization. We synthesized single crystal CrSBr using a modified procedure based on ref. 31, as detailed elsewhere63,64. The long axis of bulk needle crystals of CrSBr has been correlated to the a crystal axis by XRD experiments. Bulk crystals were exfoliated by first cleaving them on tape with a known, fixed crystal orientation, then exfoliating them mechanically on an Si/SiO2 substrate passivated by 1-dodecanol41. The orientation of the crystals on the tape transfers to the exfoliated crystals, allowing for the crystal orientation of exfoliated samples to be correlated to previous XRD characterization. Samples were handled, transported and characterized entirely in an oxygen and moisture-free environment (both less than 1 ppm).

Optical spectroscopy. Magnetic field dependence measurements were carried out in a custom Montana Instruments closed-cycle cryostat equipped with a three-axis vector magnet. Sample crystal axes were aligned visually to the vector magnet axes to within 2° along all directions. RMCD measurements were performed using non-resonant 633-nm laser light with quarter-wave modulation from a photoelastic modulator, focused to a spot size of roughly 2 μm on the sample by an aspheric singlet lens. Unless otherwise indicated, PL and reflectance measurements were carried out at a sample temperature of 5 ± 1 K. Excitation for magneto-PL measurements was from a 633-nm laser at a power of 10 μW using a beam spot size of roughly 1 μm. For magneto-reflectance spectroscopy, a thermal white light source was used. Spectra were measured using a CCD array following dispersal by a 500-nm spectrometer. Temperature-dependent PL experiments were performed with a 633-nm HeNe laser as excitation source and on-sample power of 200 μW. Emission was collected with a Princeton Instruments PyLoN-IR and SpectraPro HRS-300. In temperature- or magnetic field-dependent PL, both excitation and collection polarization axes are along the b direction, unless otherwise noted.

First-principles calculations. The mean-field starting point of the GW calculations uses DFT calculations within the spin-polarized generalized gradient approximation, performed using the Quantum ESPRESSO package38. We used norm-conserving pseudopotentials, with a plane-wave energy cutoff of 85 Ry. In the structural relaxation, we included dispersion corrections within the D2 formalism to account for the vdW interactions63. The structure was fully relaxed until the force on each atom was smaller than 0.01 eV Å−1. The calculated lattice constants along the a and b axes are 3.5 and 4.7 Å, respectively, in agreement with experimental results32. The calculated interlayer distance is 8.1 Å in bilayer. The scalar-relativistic and full-relativistic band structures show little differences near the VBM or CBM. The GW34 calculations were carried out using the BerkeleyGW package2 at the GwW level. The supercell in the monolayer and bilayer calculations uses out-of-plane lattice constants of 16 and 28 Å. A truncated Coulomb interaction is used along the out-of-plane direction to avoid interactions between the free-standing CrSBr layers and its periodic images. In the calculation of the electron self-energy, the dielectric matrix was constructed with a cutoff energy of 35 Ry. The dielectric matrix and the self-energy were calculated on an 8 × 6 × 1 k-grid. Ten subsampling points along the in-plane diagonal of the supercell are included in the calculation of the dielectric function. A static remainder approach is used, together with 1,700 bands in the bilayer calculation63. These parameters lead to a converged quasiparticle bandgap within 0.1 eV. The exciton energy levels and wavefunctions are calculated using the GW-BSE methods38. The exciton interaction kernel is calculated on a 32 × 24 × 1 k-grid in bilayer, which converges the exciton binding energy to within 0.1 eV, similar to a previous report on CrI3 (ref. 32).

Data availability

All relevant data are available in the main text, in the Supporting Information, or from the authors. These include all panels in Figs. 1–9 in the main text, Supplementary Figs. 1–9 in the Supporting Information and optimized atomic coordinates used in electronic structure calculations. There is no restriction on data availability.

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Author contributions

Y.-X.Z., X.X., K.L. and N.P.W. conceived this work. Bulk crystals were synthesized and characterized by A.H.D. and E.J.T. with supervision by X.R. and C.D. Sample preparation was carried out by K.L. and A.H.D., assisted by N.P.W. and J.C. Temperature-dependent measurements were performed by K.L. with supervision from X.-Y.Z. Field-dependent and polarization-dependent measurements were performed by N.P.W. and J.C. with supervision from X.X. The vector magnet was operated by J.F., K.X., S.S. and T.C. performed first-principles calculations that interpreted the results. The manuscript was prepared by N.P.W., K.L., J.C., K.X., T.C., X.X. and X.-Y.Z. in consultation with all other authors. All authors read and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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