Magnetization dependent tunneling conductance of ferromagnetic barriers

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Recent experiments on van der Waals antiferromagnets such as CrI3, CrCl3 and MnPS3 have shown that using atomically thin layers as tunnel barriers and measuring the temperature (T) and magnetic field (H) dependence of the conductance allows their magnetic phase diagram to be mapped. In contrast, barriers made of CrBr3—the sole van der Waals ferromagnet investigated in this way—were found to exhibit small and featureless magnetoconductance, seemingly carrying little information about magnetism. Here we show that—despite these early results—the conductance of CrBr3 tunnel barriers does provide detailed information about the magnetic state of atomically thin CrBr3 crystals for T both above and below the Curie temperature (TC ≃ 32 K). Our analysis establishes that the tunneling conductance depends on H and T exclusively through the magnetization M(H,T), over the entire temperature range investigated (2-50 K). The phenomenon is reproduced in detail by the spin-dependent Fowler-Nordheim model for tunneling, and is a direct manifestation of the spin splitting of the CrBr3 conduction band. These findings demonstrate that the investigation of magnetism by tunneling conductance measurements is not limited to antiferromagnets, but can also be applied to ferromagnetic materials.

Probing magnetism in atomically thin van der Waals crystals is challenging because most experimental methods commonly employed to study bulk compounds are not sufficiently sensitive to detect any magnetic signal from such a small amount of material1-6. Recently, it has been shown that magnetic phase boundaries—and even the complete magnetic phase diagram—of insulating atomically thin magnets can be detected by using them as tunnel barriers, and measuring their temperature-dependent magnetoconductance7-15. The sensitivity of the tunneling magnetoconductance to magnetism originates from the dependence of the tunneling probability on the magnetic state16. As H and T are varied across a magnetic transition, the alignment of the spins in the barrier changes sharply, and so does the tunneling prob-
tunneling formula (i.e., \( \log(I/V^2) \propto d/V \))\(^{27,28} \). As shown in Fig. 1d, application of an external magnetic field up to 3 T at \( T = 2 \) K causes only minor (<6%) and featureless variations in the conductance \( G = I/V \). This is consistent with previous reports\(^{13,14} \) and expected for a ferromagnetic semiconductor in which –at low temperature– the spins are spontaneously fully polarized already in the absence of an applied magnetic field.

Despite the negligible low-temperature magnetoconductance, Fig. 1e shows that the conductance measured at zero applied magnetic field \( G(H = 0, T) \) increases by a factor of three as \( T \) is lowered from the Curie temperature \( T_C = 32 \) K down to 2 K. As the conductance is virtually temperature independent above \( T_C \) (for \( 32 < T < 50 \) K), we infer directly from the experimental data that the conductance increase is due to \( \text{CrBr}_3 \) entering the ferromagnetic state. This observation implies that magnetism does influence the electrical conductance of the tunnel barriers and that the effect is sizable: a threefold increase in conductance is comparable to the magnetoresistance of most common magnetic tunnel junctions (i.e., tunneling spin valve devices\(^{29} \)) and of \( \text{CrCl}_3 \) antiferromagnetic tunnel barriers\(^{10–14} \).

This observation motivates us to look more in detail at the temperature-dependent magnetoconductance of \( \text{CrBr}_3 \) barriers. \( \delta G(H, T) \equiv [G(H, T) - G(0, T)]/G(0, T) \). The full dependence of \( \delta G(H, T) \) on \( T \) and \( H \) is shown in Fig. 2a for both the 7- and 8-layer \( \text{CrBr}_3 \) devices, with the two of them exhibiting identical behavior. In both devices, \( \delta G(H, T) \) is positive and peaks at \( T = T_C \), irrespective of the applied magnetic field \( H \). The positive magnetoconductance can be understood, since at high \( T \) the application of a magnetic field does lead to a better alignment of the spins in \( \text{CrBr}_3 \). Similarly to what happens in \( \text{CrI}_3 \) and \( \text{CrCl}_3 \) tunnel barriers, a better spin alignment enhances the tunneling probability, causing the conductance to increase. We note that when \( T \) approaches \( T_C \) from above –coming from the paramagnetic state of \( \text{CrBr}_3 \)– the magnetic field required to increase the conductance systematically decreases, indicating that the spin susceptibility \( \chi \) is enhanced. This trend is reminiscent of the behavior expected from the critical fluctuations in the neighborhood of the ferromagnetic transition\(^{30} \).

Fig. 1. Tunneling conductance of \( \text{CrBr}_3 \) multilayers. a: Schematics of the crystal structure of \( \text{CrBr}_3 \): the orange balls represent the Cr atoms with the associated spins pointing perpendicularly to the layers; the light blue balls represent the Br atoms. b: Temperature dependence of the magnetization measured on bulk \( \text{CrBr}_3 \) crystals with a magnetic field of 1 mT applied perpendicular to the layers. The inset shows the plot of \( dM/dT \), with a sharp minimum close to 32 K, corresponding to the Curie temperature of \( \text{CrBr}_3 \). c: Tunneling current as a function of applied voltage measured on a 7 (blue curve) and 8 (orange curve) layer \( \text{CrBr}_3 \) device at \( T = 2 \) K (curves of the same color in panels d and e represent data measured on the same devices). The up left insert is a cartoon representation of our hBN-encapsulated graphene/\( \text{CrBr}_3 \)/graphene tunnel junction devices. The down right insert shows that –for sufficiently large applied bias \( V \)– \( \log(I/V^2) \) is linearly proportional to \( d/V \) (\( d \) is the thickness of \( \text{CrBr}_3 \)), as expected in the Fowler-Nordheim tunneling regime. d: Magnetoconductance \( \delta G(H, T) \equiv [G(H, T) - G(0, T)]/G(0, T) \) measured at \( T = 2 \) K, on the 7 and 8 layer devices. e: Temperature dependence of the zero-field tunneling conductance of the 7 and 8 layer devices, exhibiting an increase of approximately 300 %, as \( T \) is lowered from \( T_C \) to 2 K. Data in panels d and e have been acquired with an applied bias voltage \( V = 1 \) V and \( V = 1.4 \) V for the 7 and 8 layer devices, respectively.
The idea that the magnetoconductance for \( T > T_C \) probes the fluctuations of the spins in the critical regime of the paramagnetic state can be tested quantitatively if we recall that in a mean-field description of this regime, the linear spin polarizability \( \chi \propto 1/(T - T_C) \) as \( T \) approaches \( T_C \).[30] We can then check whether the conductance depends on the magnetic field induced spin polarization, or equivalently on the magnetization (whose mean-field expression is given by the Curie-Weiss law, \( M \propto \mu_0 H/(T - T_C) \)), by simply plotting \( \delta G(H,T) \) as a function of \( \mu_0 H/(T - T_C) \) for any \( T > T_C \) (see Fig. 2b).

For sufficiently small \( \mu_0 H/(T - T_C) \) all curves indeed collapse on top of each other (Fig. 2c), irrespective of the temperature at which they are measured, confirming that in the linear regime the field-induced increase of the conductance is determined by the net spin polarization (i.e., by the field-induced magnetization).

The relation between magnetoconductance and magnetization can be tested beyond the linear regime, by using the magnetization \( M(H,T) \) measured on bulk crystals (Fig. 3b) to re-plot the magnetoconductance of our tunnel barriers \( \delta G(H,T) \) (Fig. 3a) as a function of \( M \). The result is shown in Fig. 3c, with curves of different colors representing magnetoconductance measurements done at different temperatures. When plotted as a function of \( M \) all curves collapse on top of each other throughout the entire range of \( H \) and \( T > T_C \) investigated. We can therefore conclude directly from the data that the magnetoconductance \( \delta G \) depends on \( H \) and \( T \) only through the magnetization \( M(H,T) \) even outside the linear regime. That is: for \( T > T_C \),

\[
\delta G(H,T) = \delta G(M(H,T))
\]

To extend our analysis from the paramagnetic state to \( T < T_C \), when the CrBr\(_3\) barriers are ferromagnetic, we look at the temperature dependence of the conductance measured at zero applied field. To this end, we consider the quantity \( \Delta G(H,T) \equiv [G(0,T) - G(0,T_C)]/G(0,T_C) \), i.e., the relative increase in conductance observed as \( T \) is lowered below the Curie temperature. If the conductance is a function of magnetization, this function should be the same underlying the behavior of \( \delta G(H,T) \) for \( T > T_C \), because the temperature dependence of \( G(0,T) \) originates exclusively from the temperature dependence of the spontaneous magnetization \( M(H=0,T) \), which in the ferromagnetic state increases from zero at \( T = T_C \), to its saturation value for \( T < T_C \). Consistently with this idea, the data in Fig. 1c shows an increase in conductance upon lowering \( T \) between 34 and 48 K, in 2 K steps. The result is shown in Fig. 3c, with curves of the same color correspond to measurements done at the same temperature.

Fig. 2. Temperature evolution of the tunneling magnetoconductance. a: Color plot of tunneling magnetoconductance for the 7 (left) and 8 layer (right) device as a function of applied magnetic field \( \mu_0 H \) and temperature \( T \). At fixed applied field, the magnetoconductance is maximum close to \( T_c = 32 \) K. b: Magnetoconductance of 7 layer device for \( T > T_c \), as \( T \) is varied from 34 to 48 K, in 2 K steps. c: when plotted as a function of \( \mu_0 H/(T - T_C) \), the magnetoconductance curves shown in panel b collapse on top of each other at small field, indicating that in the linear regime the magnetoconductance \( \delta G(H,T) \) depends on \( H \) and \( T \) only through the magnetization \( M(H,T) \). In panels b and c, curves of the same color correspond to measurements done at the same temperature.
Fig. 3. Magnetization dependence of tunneling magnetoconductance. a: Tunneling magnetoconductance measured for $T > T_c$, as $T$ is varied from 34 K to 48 K in 2 K steps (curves of the same color in panels b and c represent measurements taken at the same temperature). b: Magnetic field dependence of the magnetization measured on bulk CrBr$_3$ crystals. c: Magnetization dependence of tunneling magneto-conductance. Colored lines represent the magnetoconductance measured at different $T > T_c$, plotted as a function of the bulk magnetization measured at the same temperature. The orange open circles represent the relative change in conductance due to the increase in the spontaneous magnetization of CrBr$_3$, measured for different $T < T_c$, obtained from the data shown in panels d and e. All data collapse on top of each other, indicating that the conductance is a function of the magnetization, i.e. it depends on $H$ and $T$ exclusively through $M(H,T)$, throughout the entire $T$ range investigated (i.e., from well below to well above $T_C$). The black line is a fit based on the expression obtained from the spin-dependent Fowler-Nordheim tunneling model, under the assumption that the splitting between the spin up and down bands is proportional to the magnetization (see main text for details). d: Temperature dependence of the relative conductance increase as $T$ is lowered below $T_C$, in the ferromagnetic state of CrBr$_3$. e: Spontaneous magnetization of CrBr$_3$ calculated by XXZ model with anisotropic exchange interactions that –as shown in Ref. 22– accurately reproduces the measured magnetization of atomically thin CrBr$_3$ crystals.

is described by the same function from well above $T_C$ to the lowest temperature reached in our measurements (2 K). This conclusion is extremely robust, because it is drawn directly from the analysis of the experimental data, without any theoretical assumption (the 8-layer device exhibits an identical behavior, as discussed in Supplementary Note 3 and shown in Supplementary Fig. 5).

These experimental results have a straightforward interpretation within the context of Fowler-Nordheim (FN) tunneling transport commonly used to interpret the conductance of van der Waals magnetic barriers. In FN tunnelling, the applied bias tilts the conduction band across the CrBr$_3$ layer, effectively reducing the thickness of the tunnel barrier, so that eventually the tunneling probability for electrons becomes sizable and a finite current is observed$^{27,28}$. The $I – V$ characteristics in the FN tunneling regime satisfy the relation:

$$I \propto \frac{V^2}{\phi_B} e^{-\frac{8\pi e}{\hbar v} \sqrt{\frac{\phi_B}{m^*}} \phi_B^{3/2}}$$

(1)

where $m^*$ is the effective mass describing the motion of electrons in CrBr$_3$ in the direction perpendicular to the planes, $\phi_B$ is the barrier height determined by the distance between the Fermi level in the contacts and the conduction band edge in CrBr$_3$, $h$ is Planck’s constant and $e$ the (modulus of the) electron charge. For a ferromagnet, an analogous relation is expected to hold separately for electrons with spin up and down, which experience different barrier heights $\phi_\uparrow$ and $\phi_\downarrow$, due to the spin-splitting of the conduction band present for $T < T_C$ (see Fig. 4a). The total conductance is then given by the sum of the contributions given by electrons with spin up and spin down:

$$G = G^\uparrow + G^\downarrow = AV_\uparrow e^{-\frac{8\pi e}{\hbar v} \sqrt{\frac{\phi_B}{m^*}} \phi_B^{3/2}} + AV_\downarrow e^{-\frac{8\pi e}{\hbar v} \sqrt{\frac{\phi_B}{m^*}} \phi_B^{3/2}},$$

(2)

where $A$ is a constant determined by the barrier dimensions.

We use this expression to analyze the experimental data by assuming that the spin splitting of the conduction band in the ferromagnetic state is linearly proportional to the magnetization, resulting in barrier heights for spin up and down given by $\phi_{\uparrow,\downarrow} = \phi_{B0} \pm \gamma M$. We calculate the magnetoconductance $\frac{|G(M)-G(M=0)|}{G(M=0)}$.
using the value of $\sqrt{2m^*\phi_{B0}}$ extracted from the measured $I-V$ curves, and treating $\gamma$ as the sole fitting parameter. The result of this procedure—represented by the black curve in Fig. 3c—reproduces the experimental data perfectly. Interestingly, a conceptually similar approach has been followed in earlier beautiful work on the tunneling conductance of EuO barriers in the ferromagnetic state\textsuperscript{32}. That work, however, focused exclusively on the case of $T < T_C$ and zero applied magnetic field, by analyzing the temperature dependence of the conductance in terms of the measured temperature dependence of the magnetization. Our results show that the approach has a much broader validity: it can be applied both below and above $T_C$, it remains valid in the presence of a magnetic field, and—as CrBr\textsubscript{3} and EuO are very different materials—it describes very different classes of ferromagnetic insulators. If used in conjunction with a model predicting the magnetic field and temperature dependence of the magnetization, this approach allows the full magnetococonductance to be calculated. This is illustrated by the color plot in Fig. 4d that—despite having been obtained with the simplest possible Weiss model of Ising ferromagnetism—reproduces all the qualitative features observed in the experiments (compare Fig. 4d with Fig. 2a), and even exhibits a nearly quantitative agreement. Alternatively, it is also possible to extract the temperature and magnetic field dependence of the magnetization from the measured magnetococonductance, as discussed in Supplementary Note 2 and shown in Supplementary Fig. 4.

The excellent agreement between the calculated and the measured magnetococonductance (see Fig. 3c) suggests the possibility to extract the spin splitting energy quantitatively, from the value of the fitting parameter $\gamma$. This is however not straightforward, because Eq. (2) depends on the product $\sqrt{m^*(\phi_0)^3/2}$, and the effective mass $m^*$ is not known. Fig. 4b shows the spin splitting energy as a function of magnetization obtained by taking the value of $\gamma$ used to fit the $\delta G = \delta G(M)$ curves in Fig. 3c, and assuming the effective mass $m^*$ to be either the free electron mass $m_0$ or $10m_0$, respectively. A very large value chosen to mimic the flatness of the CrBr\textsubscript{3} bands in the direction perpendicular to the layers. We find that at saturation the energy splitting separating the spin-up and spin-down bands is approximately 110 meV if $m^* = m_0$ and 50 MeV if $m^* = 10m_0$, indicating that for realistic values of the effective mass the spin-splitting energy is between 50 and 110 MeV. We emphasize, however, that care is certainly needed in interpreting the meaning of this quantity microscopically, because—as applied to CrBr\textsubscript{3} barriers—the Fowler-Nordheim model is a phenomenological approach that does not take into account the complexity of the microscopic electronic structure of the material. In particular, it does not take into account that the conduction band consists of two distinct, nearly degenerate electronic bands originating from the $e_{1g}$ and $t_{2g}$ orbitals of the Cr atoms.

Irrespective of these details, the key result presented here is that the measured tunneling magnetococonductance of CrBr\textsubscript{3} is entirely determined by its magnetization, which is why magnetococonductance measurements can be used to investigate the magnetic properties of the material. We envision, for instance, that magnetococonductance measurements will allow detailed investigations of the critical behavior of the magnetic susceptibility in the paramagnetic state for $T$ very close to $T_C$ and provide a
new, experimentally simple way to determine critical exponents. This is possible because the required data analysis only relies on the fact that at small $M$ the magnetococonductance is a quadratic function of the magnetization (see Supplementary Note 2 and Supplementary Fig. 3). Another interesting possibility is to analyze magnetoconductance measurements over a broad range of temperatures and magnetic fields, to discriminate between microscopic theoretical models that predict a different functional dependence for $M(H,T)$ (see Supplementary Note 2 and Supplementary Fig. 4). These are just two examples that illustrate the most important aspect of our results, namely that measurements of the tunneling conductance are not limited to the investigation of antiferromagnetic barriers, but can also provide detailed information about the magnetic properties of ferromagnetic insulators.

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Methods

Bulk crystal growth and characterizations

Crystals of CrBr$_3$ were grown by the Chemical Vapour Transport method as reported earlier. Pure Chromium (99.95% CERAC) and TeBr$_4$ (99.9% Alfa Aesar) were mixed with a molar ratio 1 : 0.75 to a total mass of 0.5 g, and put in quartz tube with an internal diameter of 10 mm and a length of 13 cm. The preparation of the quartz reactor was done inside a glove box under pure Ar atmosphere. The tube was evacuated down to $10^{-4}$ mbar and sealed under vacuum, then put in a horizontal tubular furnace in a temperature gradient of about 10°C/cm, with the hot end at 700°C and the cold end at 580°C. After 7 days at this temperature, the furnace was switched off, and the tube cooled to room temperature. CrBr$_3$ was found to crystallise at the cold end of the tube. Shiny, thin platelet-like, dark green-blackish single crystals of typical lateral size of 2-5 mm were extracted. Bulk crystals of 2.1 mg were used for the magnetic characterization in a MPMS3 SQUID magnetometer (Quantum Design). The magnetic moment of the crystals was measured with magnetic field parallel to the crystallographic c-axis.

Tunneling junction fabrication and transport measurements

CrBr$_3$ multilayers were mechanically exfoliated from the crystals discussed in the section of crystal growth. Tunnel junctions of multilayer graphene/CrBr$_3$/multilayer graphene were assembled using a pick-and-lift technique with stamps of PDMS/PC. To avoid degradation of thin CrBr$_3$ multilayers, the exfoliation of CrBr$_3$ and the heterostructure stacking process were done in a glove box filled with Nitrogen gas, and the whole tunneling junction was encapsulated with hBN before being taken out. Conventional electron beam lithography, reactive-ion etching, electron-beam evaporation (10 nm/50 nm Cr/Ar) and lift-off process were used to make edge contacts to the multilayer graphene. The thickness of the layers was determined by atomic force microscope measurements performed outside the glove box, on the encapsulated devices. Transport measurements were performed in a cryostat from Oxford Instruments, using home-made low-noise electronics.

Data availability

All relevant data are available from the corresponding authors upon reasonable and well-motivated request.

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Author contributions

Z.W. and A.F.M. conceived the work. D.D. and E.G. grew CrBr$_3$ crystals and performed bulk characterization. T.T. and K.W. provided high-quality boron nitride crystals. Z.W. fabricated samples and performed transport measurements with help of I.G. and N.U. Z.W., I.G., N.U., M.G. and A.F.M. analyzed and interpreted the magnetoconductance data. All authors contributed to writing the manuscript.

Competing interests

The authors declare no competing interests.