**Ferromagnetic Kitaev interaction and the origin of large magnetic anisotropy in α-RuCl₃**

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α-RuCl₃ is drawing much attention as a promising candidate for the Kitaev quantum spin liquid⁴⁵. However, despite intensive research efforts, controversy remains about the form of the basic interactions governing the physics of this material. Even the sign of the Kitaev interaction (the bond-dependent anisotropic interaction responsible for Kitaev physics) is still under debate, with conflicting results from theoretical and experimental studies⁸⁹⁻¹⁰⁻¹⁵. The significance of the symmetric off-diagonal exchange interaction (referred to as the J' term) is another contentious question⁶⁻¹⁰⁻¹⁵⁻¹⁸. Here, we present resonant elastic X-ray scattering data that provide unambiguous experimental constraints to the two leading terms in the magnetic interaction Hamiltonian. We show that the Kitaev interaction (K) is ferromagnetic, and that the J' term is antiferromagnetic and comparable in size to the Kitaev interaction. Our findings also provide a natural explanation for the large anisotropy of the magnetic susceptibility in α-RuCl₃, as arising from the large J' term. We therefore provide a crucial foundation for understanding the interactions underpinning the exotic magnetic behaviours observed in α-RuCl₃.

The magnetic behaviour of the honeycomb material α-RuCl₃ has been the topic of much recent work, following the discovery in this material of an unusual continuum of magnetic excitations not well explained by spin-wave theory⁵⁻¹⁶. The structural environment and electronic state of the ruthenium atoms in α-RuCl₃ are such that the Kitaev magnetic interaction⁹ is expected to be substantial⁵⁻¹⁹. For this reason, these remarkable findings have been attributed to fractionalized excitations analogous to those found in the spin liquid ground state of the Kitaev model⁵⁻¹⁶. Recent discovery of quantization of the thermal Hall signal in the intermediate magnetic field phase has further stimulated interest in this material⁸.

Understanding the salient features of magnetism in α-RuCl₃ requires a good knowledge of the magnetic interactions between the ruthenium magnetic moments. The magnetic Hamiltonian relevant to this material includes an isotropic Heisenberg (J) term as well as bond-dependent anisotropic Kitaev (K), and off-diagonal J' terms⁵⁻¹⁰⁻¹⁵⁻¹⁸. The general Hamiltonian for spins (S) at adjacent sites i and j takes the following form, where α, β and γ denote the spin components:

\[
\mathcal{H}^{(J')}_{ij} = JS_i \cdot S_j + K S_i^\alpha S_j^\alpha + \Gamma (S_i^\beta S_j^\delta + S_i^\delta S_j^\beta) + \Gamma' (S_i^\gamma S_j^\gamma).
\]  

Often included in this Hamiltonian are further-neighbour isotropic interaction terms (J₂, J₃, and so on) and additional off-diagonal term J' due to non-zero trigonal crystal fields⁴⁻¹⁰⁻¹⁵. Early ab initio calculations⁴ and fits to inelastic neutron scattering measurements⁵⁻¹⁶ suggested an antiferromagnetic Kitaev interaction (K > 0). However, later calculations using the updated monoclinic crystal structure⁴ instead suggested that the Kitaev term is ferromagnetic (K < 0)⁴⁻¹⁵⁻¹⁶⁻¹⁸. Further studies have shown that many aspects of the behaviour of α-RuCl₃ can be modelled using Hamiltonians featuring ferromagnetic K interactions supplemented by J and J' terms as in equation (1). Magnetization⁴⁻¹⁵⁻¹⁶⁻¹⁸⁻¹⁹⁻²⁰⁻²¹⁻²²⁻²³⁻²⁴⁻²⁵⁻²⁶⁻²⁷⁻⁻²⁸⁻²⁹⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻⁻˓
Fig. 1 | Characterization of magnetic scattering. a, The $C2/m$ crystal structure and ordered moment directions of $\alpha$-RuCl$_3$ proposed in ref. $^3$. Crystal structure drawings were produced using VESTA$^4$. b, Temperature dependence of magnetic diffraction intensity at (0, $-1$, 1.43), showing an ordering temperature of 12 K. At each temperature, the magnetic peak was measured by simultaneously scanning the sample and detector angles. The red solid line is a guide to the eye. Inset: intensity dependence on $L$ (perpendicular to the honeycomb plane) in reciprocal lattice units (rlu) showing a broad peak at the $L = 1.5$ position. At each $L$ value, the magnetic peak was measured by scanning along the momentum-space $K$ direction. The integrated intensities for all scans were found by fitting the scans with a Gaussian peak shape. Error bars shown are the square root of covariance value from the fit. The vertical dashed line indicates the $L$ position used in the azimuthal dependence study. c, Energy dependence of the magnetic diffraction intensity at (0, $-1$, 1.43), showing resonance at the Ru L$_3$ resonant energy of 2,837.5 eV denoted with the vertical dashed line. Integrated intensities and error bars were calculated from combined scans of the sample and detector angles, as in b. d, Comparison of the magnetic signals obtained with the incident photon energy at the Ru L$_3$ edge (2,837.5 eV) and the Ru L$_2$ edge (2,970 eV). Scans were collected by simultaneously scanning sample and detector angles. Error bars shown are the square root of the number of photons detected. A constant background was subtracted, and the overall photon counts normalized to the monitor recording incident beam intensity. a.u., arbitrary units.

Fig. 2 | Azimuthal dependence of magnetic scattering intensity. a, Schematic diagram showing the geometry of the REXS experiment. $k_\text{in}$ and $k_\text{out}$ show the direction of incoming and outgoing photons, while $E_\text{in}$ and $E_\text{out}$ indicate their linear polarization direction. $q$ and $\Psi$ are the momentum transfer and the azimuthal angle, respectively. b, Azimuthal dependence of the magnetic diffraction signal at (0, $-1$, 1.43). The azimuthal dependence is fit best with a magnetic moment angle of $\Theta = +32^\circ$. The modelled intensities for $\Theta = +25^\circ$, $+45^\circ$ and $-35^\circ$ are shown for comparison. $\Psi = 0$ corresponds to the position with the in-plane direction (−2, 0, 0.65) pointing along the scattered beam. The magnetic peak was measured by scanning the sample angle. Integrated intensities were found by fitting the scans with a Gaussian peak shape. Error bars shown are the square root of the covariance value from the fit. a.u., arbitrary units.
on momentum, temperature, and incident photon energy. The momentum dependence of the magnetic signal showed an extended rod in the out-of-plane direction, with a broad peak at the position expected for ABAB-type layer stacking as shown in Fig. 1b. This stacking order has previously been reported in neutron diffraction measurements with an ordering temperature of 14 K, as opposed to the 7 K ordering temperature observed for the three-layer stacking. We measured an ordering temperature of 12 K for this sample, as shown in Fig. 1b. This apparent suppression (by 2 K) of the 14 K ordering temperature is most likely due to beam heating. We note that diffraction measurements at this X-ray energy will be highly surface-sensitive, since the beam penetrates the sample to a depth of only a few hundred nanometres, and the observed two-layer stacking may not reflect the bulk crystal structure. As the magnetic interactions are strongly two-dimensional, we do not anticipate that stacking has a large effect on the moment direction. Scans along the \( L \) direction in reciprocal space were also collected at several different azimuthal positions, to ensure that the azimuthal dependence does not depend on the \( L \) position selected. The position \( L = 1.43 \) was selected to maximize intensity while maintaining an accessible position for the diffractometer.

The azimuthal-dependent measurement was collected at an incident photon energy of 2,837.5 eV (corresponding to the Ru L\(_3\) edge), where the intensity is at a maximum. The dependence of the peak intensity on the incident photon energy was measured both to find the optimal energy for measurement, and to confirm the resonant nature of the magnetic peak. This energy dependence is plotted in Fig. 1c. Following the measurements at the \( L_\gamma \) edge, the magnetic peak intensity at the same reciprocal space position was also measured at the Ru \( L_\alpha \) edge (incident photon energy 2,970 eV). The integrated intensity was substantially weaker at the \( L_\alpha \) edge (comparison is shown in Fig. 1d), and we calculate a ratio of approximately 20 for the intensities at the two photon energies. This relatively large ratio is shown in Fig. 1d, and we calculate a ratio of approximately 20 for

\[
\frac{I(\text{Ru L}_\alpha)}{I(\text{Ru L}_\gamma)} = \frac{2.3}{S = 1/2}
\]

in order to have \( \Theta \sim 32^\circ \) the magnitude of \( \Gamma \) must be a large fraction of, or even exceed the magnitude of \( K \). We note that the moment direction has also been determined via REXS for another Kitaev material Na\(_2\)IrO\(_3\) (Ref. 30). This measurement found a moment direction of \( \Theta = 45^\circ \), which would suggest that the \( \Gamma \) term is much smaller in Na\(_2\)IrO\(_3\). The lithium analogue Li\(_2\)IrO\(_3\) has also been examined as a potential realization of the Kitaev model, however this material shows an incommensurate spiral magnetic ground state in contrast to the collinear zigzag ordering found in Na\(_2\)IrO\(_3\) and Cr\(_2\)Ru\(_2\)Cl\(_6\). The non-collinear ordering means that the ordered moment direction is not a well-defined quantity in Li\(_2\)IrO\(_3\), and the results of Ref. 30 cannot be applied.

Our REXS results provide a clue for solving one of the remaining questions regarding the magnetic properties of Cr\(_2\)Ru\(_2\)Cl\(_6\): its large magnetic anisotropy. As reported by many groups\(^{24,31,32}\), the in-plane magnetic susceptibility measured by applying a magnetic field along the direction in the \( a-b \) plane is much larger than the out-of-plane susceptibility. A conventional way to explain this would be resorting to the \( g \)-factor anisotropy. However, experimental data suggest that \( g \)-factor anisotropy cannot be very large, certainly not large enough to account for the anisotropic susceptibility\(^{25,31}\). Another route to obtain a large magnetic anisotropy is via a large \( \Gamma \) term as suggested in Refs. 32, 39. Physically, the effect of the \( \Gamma \) interaction is to force the moments towards the \( a-b \) plane, which accentuates magnetic anisotropy.

We demonstrate that a large \( \Gamma \) is sufficient to explain the observed magnetic anisotropy by comparing the experimental data with theoretical calculation results. The low-field magnetization data for fields applied in-plane and out-of-plane are plotted in Fig. 3, which shows that the susceptibility (slope) anisotropy is about \( \chi_{ab} / \chi_{c} \sim 8 \). These data are fit with the classical \( JK\Gamma \) model (equation (1)), where the model parameters are chosen to be consistent with the magnetic moment direction determined by REXS. Either a small \( \Gamma \) or \( J_1 \) term was added to ensure the zigzag ground state of the model (details about the calculation are provided in the Supplementary Information). The data can be fitted for several...
parameter choices with ferromagnetic $K$ and antiferromagnetic $\Gamma$ of similar magnitude, demonstrating that the magnetization data can be explained without resorting to g-factor anisotropy. Finally, we note that the parameter regime deduced from our REXS measurement also allows for a qualitative description of the unusual continuum of magnetic excitations observed in this material. In ref. 59 it was shown that $K < 0$ and $\Gamma > 0$ with a ratio of $|\Gamma / K| \sim 1$ can explain the star-shaped continuum intensity centred around the Brillouin zone centre observed in inelastic neutron scattering.

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Methods
REXS measurements were carried out at the beamline P09 at PETRA III at DESY (see ref. 41 for details) at the Ru L₂ and L₃ edges (2,838 keV and 2,967 keV, respectively). Most of the measurements, including momentum, temperature and azimuthal dependence, were collected at the L₃ edge. The magnetic intensity was also measured at the L₂ edge to determine the branching ratio. The monochromator was detuned to minimize the presence of higher harmonics in the beam, and the measurements were made with a sodium iodide scintillation detector. An all-in-vacuum path was used to minimize X-ray absorption by air. α-RuCl₃ single crystals were grown by vacuum sublimation in sealed quartz tubes using commercial RuCl₃ powder. The single crystal used for this measurement was a flat plate with largest dimension ~400 μm. This crystal was selected from the same batch as crystals used for our previous neutron scattering measurements. Crystals from this batch were also characterized by bulk heat capacity and magnetization measurements, and showed ordering at 8 K with a small feature at 14 K indicating the presence of a minor phase with this higher ordering temperature. The crystal used for the REXS measurement was not large enough to permit bulk measurement, however it is likely composed of a combination of phases with the two ordering temperatures.

The orientation of the crystal used for this measurement was checked at room temperature with the crystal in the known monoclinic structural phase, by checking structural Bragg peaks using higher-energy (third-harmonic) photons. The crystal was also checked to ensure that it did not possess a twin rotated by 180°, which would affect the result of the azimuthal measurement. This was done by searching for structural peaks at the positions expected for the rotated structure. No intensity was found at the peak positions expected for the rotated crystal structure. The azimuthal dependence data were corrected for beam absorption (as described in ref. 43) and the beam footprint on the sample. Beam footprint on the sample was calculated from the angle between the sample surface and the incoming beam, and depended only on the ratio of the beam height and the smallest dimension of the sample. This ratio of beam height to sample size and the magnetic moment angle Θ were the only parameters refined in the fit of the azimuthal dependence data. More detailed information about the fitting procedure can be found in the Supplementary Information.

The magnetization measurements were carried out using a commercial superconducting quantum interference device (SQUID) magnetometer at 2 K. Measurements were carried out by applying magnetic fields in two directions, one along the crystallographic b-direction within the honeycomb layer, and the other along the direction perpendicular to this plane (c*-direction).

Data availability
All data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

Code availability
The computer code used to generate results that are reported in the paper is available from the authors on reasonable request.

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Author contributions
J.A.S. and Y.-J.K. conceived the experiments. J.A.S., P.J.B. and S.F. performed the experiments and J.A.S. analysed the data. J.A.S. synthesized and characterized the sample. S.K. provided the magnetic susceptibility data. L.E.C. and Y.B.K. performed theoretical calculations. J.A.S. and Y.-J.K. wrote the paper with contributions from all co-authors.

Competing interests
The authors declare no competing interests.

Additional information
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