Fragmentation in spin ice from magnetic charge injection

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The complexity embedded in condensed matter fertilizes the discovery of new states of matter, enriched by ingredients like frustration. Illustrating examples in magnetic systems are Kitaev spin liquids, skyrmions phases, or spin ices. These unconventional ground states support exotic excitations, for example the magnetic charges in spin ices, also called monopoles. Here, we propose a mechanism to inject monopoles in a spin ice at equilibrium through a staggered magnetic field. We show theoretically, and demonstrate experimentally in the Ho\textsubscript{2}Ir\textsubscript{2}O\textsubscript{7} pyrochlore iridate, that it results in the stabilization of a monopole crystal, which exhibits magnetic fragmentation. In this new state of matter, the magnetic moment fragments into an ordered part and a persistently fluctuating one. Compared to conventional spin ices, the different nature of the excitations in this fragmented state opens the way to tunable field-induced and dynamical behaviors.
The spin ice state is the archetype of an exotic magnetic state that can be produced in the presence of geometric frustration and results from the inability of the system to find a unique ground state that minimizes its energy. It emerges in pyrochlore lattices of vertex-sharing tetrahedra when the magnetic moments are subjected to an effective ferromagnetic interaction and are constrained along the local (111) directions joining the corners to the center of each tetrahedron (diagonals of the cubic crystal). It is, classically, a macroscopically degenerate ground state, in which the spins locally obey the ice rule, with two spins pointing in and two spins pointing out of each tetrahedron (212O). Spin ice is the realization of a Coulomb phase. Indeed, the ice rule is a local constraint that can be mapped on an emergent divergence free field, which is actually the local magnetization. This results in the famous pinch-point pattern in the magnetic diffuse scattering. The spin ice elementary excitations are obtained by flipping one spin at the center of a pair of tetrahedra, resulting in 3 spins in–1 spin out (311O) and 1 spin in–3 spins out (113O) configurations. These excitations, called magnetic monopoles, can be described as two effective magnetic charges \( q = \pm 1 \) (for the 311O, 113O configurations respectively) that can be deconfined.

In the presence of dipolar interactions, and provided the charge density is large enough, the Coulomb-like interaction between monopoles can stabilize a monopole crystal (staggered 311O and 113O configurations) in an underlying spin-disordered manifold. The magnetic charge crystallization results in the fragmentation of the magnetization, a remarkable state where a single degree of freedom, the Ising magnetic moment, fragments thermodynamically into two parts, each part sustaining a different phase: an ordered antiferromagnetic phase, also interpreted as a crystal of magnetic charges (divergence full), and a disordered Coulomb phase (divergence free) with predominant ferromagnetic correlations.

A magnetic charge order, on top of degenerate spin configurations, was actually predicted earlier in spin models of dipolar ice on a kagome lattice (corner-sharing triangles) before being observed in a two-dimensional artificial spin ice, where the nanomagnets featuring the spins lie on a kagome lattice, and in a kagome oxide compound. This peculiar state was finally shown to be associated with the fragmentation of the magnetization in an artificial kagome system. The local ice rule in the kagome case actually corresponds to 2 in–1 out or 1 in–2 out configurations, so that alternating charges \( q = \pm 1 \) are present on each triangle. The charge crystal can therefore easily be stabilized in the kagome ice at variance with the pyrochlore spin ice where magnetic charges are a priori not present in the ground state. The fragmented state has nevertheless also been evidenced in a pyrochlore oxide compound with more complex signatures due to quantum effects.

Here, we show that such fragmented state can be simply created in pyrochlore spin ices by injecting magnetic charges at equilibrium through a staggered magnetic field. By neutron scattering measurements, we provide evidence that this state is realized in the pyrochlore iridate compound \( \text{Ho}_2\text{Ir}_2\text{O}_7 \). In addition, signatures of unconventional excitations, equivalent to magnetic charges diffusing in a periodic potential, are observed, while plateau physics is identified in the magnetization as a function of an external magnetic field.

**Results**

**Model.** A very appealing proposal is indeed to generate fragmentation of the magnetization by injecting magnetic charges in a controlled way, that is, with a tunable parameter such as a magnetic field, without starting from this required state of

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**Fig. 1** Staggered field in a pyrochlore lattice and magnetic fragmentation. a Pyrochlore lattice submitted to (111) staggered field represented by the arrows on each site, with in and out tetrahedra when this field points either inward or outward of the tetrahedra. b Structure of the pyrochlore iridate \( \text{Ho}_2\text{Ir}_2\text{O}_7 \) with the \( \text{Ho}^{3+} \) ions (red) and the \( \text{Ir}^{4+} \) ions (blue), both occupying a pyrochlore lattice. Each rare-earth is surrounded by a hexagon of six Ir nearest neighbors. When the iridium lattice orders magnetically in the AIAO phase, as shown on the blue lattice, a local magnetic field, perpendicular to the hexagon plane, hence aligned along the local (111) directions, is felt by the central rare-earth ion, as represented in a, c Phase diagram as a function of reduced temperature \( T/J_{\text{eff}} \) and local field \( h_{\text{loc}}/J_{\text{eff}} \), defined through the charge order parameter \( M_q = \left( \sqrt{2} \sum_{n=1}^{N} \delta_q \alpha_n \right) \) where \( N \) is the number of tetrahedra and \( \delta_q = +1 \) (−1) for in (out) tetrahedra carrying a charge \( q_e \). As \( h_{\text{loc}}/J_{\text{eff}} \) increases, the ground state changes from the spin ice (\( M_q = 0 \), blue), to the AIAO state (\( M_q = 2 \), yellow), going through the charge crystal intermediate phase which fragments (\( M_q = 1 \), orange). The fragmentation can be described by the splitting of the pseudo-spin \( \alpha = \pm 1 \): the divergence free fragment on a tetrahedron can then be written as a state with four spins such that \( \sigma_{q=1} = (\pm 1, \pm 1, \mp 1, \mp 1) \), while the divergence full fragment that carries the magnetic charge corresponds to an AIAO configuration with half of the moment so that \( \sigma_{q=1} = (\pm 1, \pm 1, \mp 1, \mp 1/2) \). All the spin and charge configurations are shown in the left panel of c at \( T = 0 \). d Magnetic scattering function \( S(Q) \), with \( Q \) the scattering vector, calculated in the fragmented phase. It shows a diffuse pattern with pinch points (evidenced by arrows) together with the magnetic Bragg peaks (highlighted by circles) characteristic of the AIAO ordered state, but whose intensity (proportional to the magnetic moment squared) is a quarter of that expected when the whole magnetic moment is ordered.
monopole crystal, which imposes strong constraints on the Hamiltonian. However, in a pyrochlore spin ice, any external magnetic field does not act homogeneously on the magnetic moments of the unit cell, since the magnetic moments inside a tetrahedron point in different (111) directions. When the field is applied along one of the (111) directions, along which the pyrochlore lattice can be viewed as a stacking of triangular and kagome planes, only a partial fragmentation occurs (within the kagome planes). This leads to the so-called kagome ice phase in which a charge crystal is stabilized in the kagome planes through a 2 in–1 out, 1 in–2 out rule, similar to artificial spin ices. However, no magnetic charges are actually present in the tetrahedral units.

We propose below an approach to produce the fragmentation process in the whole pyrochlore lattice, even in the absence of dipolar interactions, by considering a magnetic field exerted along the local directions of the magnetic moments. As depicted in Fig. 1a, if the sign of this field alternates, pointing inward for a given tetrahedron (called in) and outward for the neighboring tetrahedra (called out), this staggered field competes with the spin ice state. This results, over a large field range, in a fragmented ground state supporting unconventional excitations.

Consider the nearest-neighbor spin ice Hamiltonian in the presence of a local magnetic field:

\[ H = -J \sum_{\langle ij \rangle} S_i \cdot S_j - g_\mu_B H B \sum_i H^{\text{loc}}_i \cdot S_i \]

\[ = J_{\text{eff}} \sum_i \sigma_i \sigma_j - h_{\text{loc}} \sum_i \sigma_i \]

(1)

Where, \( S_i \) is the \( i \)th magnetic moment pointing along its local trigonal direction and interacting with its nearest neighbors via a ferromagnetic interaction \( J \). \( \sigma_i = \pm 1 \) is the corresponding Ising pseudo-spin which is equal to \( +1 \) (–1) when the moment points inward (outward) the tetrahedron. \( J_{\text{eff}} = J / 3 S^2 > 0 \) is the effective nearest-neighbor interaction. \( H^{\text{loc}}_i \) is the staggered magnetic field described above, aligned along the (111) direction of the \( i \)th site, and \( h_{\text{loc}} = g_\mu_B H B \).

Minimization of Eq. (1) on a single tetrahedron, together with Monte Carlo simulations on a pyrochlore lattice (Fig. 1c and Supplementary Fig. 7), gives a succession of ground states with a different charge order parameter, depending on \( h_{\text{loc}} / J_{\text{eff}} \). At low fields, the ground state is the 2I2O spin ice state (\( |q_lq_l| = 0 \)). In the opposite limit of large fields, the stabilized state is the all in–all out (AIAO) antiferromagnetic order (charge crystal with \( |q_l| = 2 \)), in which all the spins of a given tetrahedron point either inward or outward, following the staggered field.

More exotic physics appear in the intermediate regime \( 2 < h_{\text{loc}} / J_{\text{eff}} < 6 \) (orange region in the phase diagram of Fig. 1c): the competition between \( J_{\text{eff}} \) and \( h_{\text{loc}} \) selects 3110 and 1310 configurations on the in and out tetrahedra, respectively. It thus stabilizes a charge crystal, with \( |q_l| = 1 \), and leads to the fragmentation process, characterized by a staggered magnetization at half of the magnetic moment. Another important signature is the coexistence of diffuse scattering with a pinch-point pattern and AIAO Bragg peaks in the computed scattering function (Fig. 1d). Here, the fragmented state does not result from the presence of the long-range monopole Coulomb energy but from the injection at equilibrium of magnetic charges into a spin ice state by a staggered magnetic field.

**Experimental evidence for fragmentation.** A prominent system to realize this model is the pyrochlore iridate family, of formula \( \text{R}_2\text{Ir}_2\text{O}_7 \) in which \( \text{R} \) is a rare-earth element. In these compounds, the iridium and rare-earth ions lie on two interpenetrated pyrochlore lattices (Fig. 1b). It was shown in pyrochlore iridites where \( R = \text{Nd}, \text{Eu}, \text{and Tb}, \) that the iridium sublattice orders in an AIAO magnetic arrangement simultaneously with a metal ininsulator transition \((T_{\text{MI}} = 30–140 \text{ K})\). This arrangement generates a molecular field oriented along the local (111) directions on the rare-earth sublattice, and thus plays exactly the same role as our staggered magnetic field \( h_{\text{loc}} \) in Eq. (1). The fragmentation process described above can thus be realized in the pyrochlore iridates, provided the rare-earth ions have a local easy axis anisotropy along the (111) directions and interact through a ferromagnetic interaction. This is the case of Ho and Dy ions in pyrochlore compounds with nonmagnetic ions instead of Ir\(^{4+}\), and where spin ice physics is observed at Kelvin temperatures. In the present article, we focus on the Ho pyrochlore iridate.

Our measurements were performed on a high-quality polycrystalline sample of Ho\(_2\)Ir\(_2\)O\(_7\). As previously reported, the magnetization exhibits a zero field cooled-field cooled (ZFC-FC) bifurcation at \( T_{\text{MI}} = 140 \text{ K} \) pointing out the magnetic ordering of the Ir sublattice (Supplementary Fig. 1). Below this temperature, the iridium sublattice starts to produce a molecular field on the holmium sites, which competes as the rise of magnetic Bragg peaks below about 80 K in the neutron diffraction pattern. Due to the very weak moment of the iridium, these Bragg peaks are mainly characteristic of an AIAO magnetic ordering of the holmium sublattice (Fig. 2a). This is the ordering expected in our model when the temperature and the staggered magnetic field are large with respect to the effective interaction. From the temperature evolution of the refined Ho\(^{3+}\) ordered magnetic moment, strongly increasing when decreasing the temperature, we could extract the value of the molecular field \( E^{\text{loc}} \) estimated to 0.725 ± 0.05 T below 40 K (Fig. 2b). This was achieved following the procedure of ref. 20 and using the crystal electric field parameters of the Ho\(^{3+}\) ion deduced from inelastic neutron scattering (INS) experiments (Supplementary Notes 1 and 2 and Supplementary Figs. 2–4).

When decreasing further the temperature, the Ho–Ho magnetic interactions start to become relevant. The ordered moment finally saturates below 2 K, ending with a value of 5 ± 0.06 \( \mu_B \) at 50 M.K. This value is only half the magnetic moment of the Ho\(^{3+}\) ions in their ground state doublet (Supplementary Note 2). A diffuse scattering signal is actually present and accounts for the missing moment, as seen in Fig. 2c. It has broad maxima at the scattering vectors \( Q \sim 0.7, 1.6, \) and 2.7 Å\(^{-1}\), evidencing additional spin correlations characteristic of the spin ice diffuse scattering, resulting from the powder average of the pinch-point pattern observed on single crystals. This coexistence of a spin ice diffuse scattering with an AIAO ordering, whose ordered magnetic moment is half of the total moment, is remarkably well reproduced in the calculations (Fig. 2d) and is the signature that a fragmentation process occurs in Ho\(_2\)Ir\(_2\)O\(_7\) below 2 K. Finally, the calculated temperature dependence of the ordered moment using Eq. (1) gives \( J_{\text{eff}} = 1.4 ± 0.1 \text{ K} \) and \( h_{\text{loc}} / J_{\text{eff}} = 4.5 ± 0.25 \) (Fig. 2b), which places Ho\(_2\)Ir\(_2\)O\(_7\) deep into the fragmentation regime according to Fig. 1c. It corresponds to a local field \( \mu_B H^{\text{loc}} = 0.94 ± 0.02 \text{ T} \), in the same range as the molecular field estimated from the high temperature behavior.

We have thus demonstrated that the thermodynamical fragmentation of the magnetic moment under the influence of a local staggered field predicted in our model appropriately describes the unexpected properties of Ho\(_2\)Ir\(_2\)O\(_7\). In the following, we probe the macroscopic magnetic properties of Ho\(_2\)Ir\(_2\)O\(_7\) to explore the excitations emerging from this fragmented state.

**Dynamics in the fragmented state.** In the fragmented regime, typically below 2 K, the alternative (AC) susceptibility displays a frequency dependence, which can be described by a thermally
The rise of magnetic Bragg peaks. The 1.5 K Rietveld refinement using a diffractograms recorded at 200 K (red dots) and 1.5 K (green dots) showing the rise of magnetic Bragg peaks. The 1.5 K Rietveld refinement (black line) using $a \mathbf{k} = \mathbf{0}$ propagation vector yields an AIAO magnetic order for the holmium sublattice. The difference between the 1.5 and 200 K data (blue line, shifted for clarity) enhances the magnetic Bragg peaks.

b Temperature evolution of the refined Ho$^{3+}$ magnetic moment, in a semi-logarithmic scale, measured with two diffractometers (D1B and G4.1). The red dashed curve is the calculated ordered magnetic moment induced by the temperature dependent molecular field created by the Ir magnetic ordering. Below 20 K, the calculated and experimental curves depart from each other due to the presence of Ho-Ho magnetic interactions. The red full curve is obtained from Monte Carlo calculations (Eq. (1)) with $J_{\text{eff}} = 1.4$ K and $h_{\text{loc}}/J_{\text{eff}} = 4.5$, the colored area representing the calculated values compatible with the experimental error bars (within symbol size). These calculations allow to account for the saturation of the ordered moment below 2 K due to fragmentation. The departure above 40 K between the calculated and experimental curves is attributed to the decrease of the Ir molecular fragmentation. The departure above 40 K between the calculated and experimental curves is attributed to the decrease of the Ir molecular fragmentation.

c Evidence for diffuse scattering at 1.5 K, from the difference between the 1.5 and 200 K diffractograms (the negative intensity is due to an imperfect subtraction of a strong nuclear peak caused by the lattice parameter variation). d Powder average magnetic scattering function $S(Q)$ from Monte Carlo calculations at $T/J_{\text{eff}} = 0.05$ with $h_{\text{loc}}/J_{\text{eff}} = 4.5$.

activated process above an energy barrier $E \approx 4.8$ K (Fig. 3a, Supplementary Note 3 and Supplementary Figs. 5 and 6). It is associated with a freezing in the ZFC-FC magnetization below 1 K, where a strong irreversibility is observed. Similar slow dynamics are observed in spin ice materials 26, 27, where they are induced by the diffusion of monopoles, with a relaxation timescale inversely proportional to the monopole density 17, 18, 29. In the present case, the dynamics can also be understood by the diffusion of the fractional excitations, of energy $2J_{\text{eff}}$ and charge $q = \pm 1$, emerging from the fragmented Coulomb phase. But a marked difference with the spin ice is that these excitations are diffusing in a periodic potential $V_{q}^{\nu}$ that depends on the local field. It is created by the underlying charge crystal fragment through a local interaction between the diffusive charge and the ordered charges. The energy of a single excitation is then given by

$$E_{q}^{\nu} = 2J_{\text{eff}} + V_{q}^{\nu},$$

with $V_{q}^{\nu} = (4J_{\text{eff}} - h_{\text{loc}})q_{\nu}^{2}q_{\nu}$, and where $q_{\nu}^{2} = -1, +1$ is the ordered charge on a tetrahedron with $\alpha = \text{out}$, in. For $h_{\text{loc}}/J_{\text{eff}} = 4.5$, corresponding to the Ho$_{2}$Ir$_{2}$O$_{7}$ case, the periodic potential is alternately repulsive (attractive) for a diffusing charge interacting with an ordered charge of opposite (same) sign (Fig. 3b). The relaxation timescale is then given by the highest energy barrier $E = h_{\text{loc}} - 2J_{\text{eff}} \approx 3.5$ K, rather close to the experimental value. Long-range dipolar interactions 17, as well as more complex diffusion mechanisms (Supplementary Note 4 and Supplementary Figs. 8 and 9), could significantly affect the
The introduction of a staggered magnetic field in the nearest-neighbor spin ice Hamiltonian on the pyrochlore lattice thus allows, through magnetic charge injection, to stabilize a fragmented state, and offers a rich playground to study the various facets of spin ice and monopoles physics in a renewed perspective. A step further would be to account for dipolar interactions in the Hamiltonian, which might affect the actual ground state and the dynamics at very low temperature.

We have shown that, in the pyrochlore iridate Ho$_2$Ir$_2$O$_7$, the ratio between the rare-earth interactions and the staggered field induced by the iridium moment places the system deeply in the fragmented phase. Experimentally, it would then be very appealing to explore the full fragmentation phase diagram and the associated exotic dynamics by varying the ratio between rare-earth interactions and the staggered local field. This can be done by changing the rare-earth element. Preliminary measurements on the Dy pyrochlore iridate suggest that fragmentation also occurs in this compound, and hopefully, this system will be located at a different $h_{loc}/J_{eff}$. This phase diagram mapping should also be achieved more systematically by applying an external isostatic pressure. Finally, the model we have developed is perfectly equivalent to the case of antiferromagnetically-coupled collinear Ising spins on a pyrochlore lattice submitted to a uniform external field, which can be easily varied. It thus opens the route to the tuning of fragmentation in new systems.

### Discussion

The introduction of a staggered magnetic field in the nearest-neighbor spin ice Hamiltonian on the pyrochlore lattice thus allows, through magnetic charge injection, to stabilize a fragmented state, and offers a rich playground to study the various facets of spin ice and monopoles physics in a renewed perspective. A step further would be to account for dipolar interactions in the Hamiltonian, which might affect the actual ground state and the dynamics at very low temperature.

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### Methods

#### Calculations

Thermodynamical quantities as well as structure factors have been calculated through Monte Carlo simulations of the spin model described by Eq. (1). A single spin-flip Metropolis update has been combined to a loop algorithm preventing the system from being trapped into a small region of the configuration space at low temperature. The simulations were performed on $16 \times 16 \times 16$ lattice sites with $L = 8\times 12$ (Figs. 1d and 4b) and 24 (Figs. 1c and 2d) with periodic boundary conditions. In total, $10^4$ hybrid Monte Carlo steps were used for thermalization, while the measurements were computed over $N$ steps, where $N$ is adapted to ensure stochastic decorrelation between measurements (typically, from $N = 10^3$ at high temperature to $10^5$ at low temperature). The powder magnetization of Fig. 4b, averaged over 800 random field directions, has been computed at sufficiently high temperature to preserve the efficiency of the used algorithm.

#### Synthesis

Polycrystalline samples of Ho$_2$Ir$_2$O$_7$, with $R =$ Ho$^{3+}$, $4f^{10}$, $S = 2$, $L = 6$, $J = 8$, $g_z = 5/4$, $g_l = 10$ were synthesized by a mineralization process. The starting reagents Ho$_2$O$_3$ and IrO$_2$ were mixed together with a small amount of KF flux and pressed into a pellet. After being placed in a Pt crucible, they were submitted to a heat treatment under air atmosphere: 200 °C/h$^{-1}$ until a plateau of 120 h at 1100 °C and a decrease to room temperature at 200 °C/h$^{-1}$. The pyrochlore iridates crystallize in the $F\overline{4}3m$ cubic space group, with the O occupying the 48f and 8b Wyckoff positions, the rare-earth and the Ir occupying the 16d and 16c positions respectively. The structure and quality of the samples were checked by X-ray diffraction, with no trace of parasitic phases. The lattice parameter and the $\alpha$ coordinate of the 48fO were found at room temperature equal to 10.170(2) Å and 0.343(2) Å.

#### Magnetization measurements

The temperature and field dependences of the magnetization of the Ho$_2$Ir$_2$O$_7$ powder sample down to 2 K were measured using Quantum Design VSM and MPM’s SQUID magnetometers and are shown in Supplementary Fig. 1. The magnetization vs temperature exhibits a ZFC-FC bifurcation at $T_{c}$ = 140 K pointing out the ordering of the Ir sublattice concomitant with the metal-insulator transition. The magnetic isothersms, linear at high temperature, are approaching a plateau of $m\approx 5$% at 2 K, which reflects the strong Ising anisotropy of Ho$^{3+}$.

Very low temperature magnetization measurements were performed on SQUID magnetometers equipped with a dilution refrigerator developed at the Institut Néel. The powder samples were crushed and mixed with Apiezon grease in a copper pouch to ensure a good thermal contact between the copper sample holder and the sample. AC susceptibility measurements were performed with an AC field of 0.1 or 0.23 mT. Demagnetization corrections were performed assuming an average demagnetization factor of 0.1 (cgs units) estimated from the shape of the copper pouch.

#### Neutron diffraction

The CRG-D1B diffractometer at the Institut Laue Langevin (ILL) (wavelength $\lambda = 2.52$ Å) was used in the 1.5–300 K temperature range and the G4.1 diffractometer at the LLB ($\lambda = 2.427$ Å) was used for the lower temperature range. It is equipped with a Cryoconcept-France HD dilution refrigerator (100 µW at 100 mK), and fitted with vanadium shields. The sample was put in a vanadium cell in 14 bars of He gas. Rietveld refinements were carried out with the FULLPROF program.

#### Inelastic neutron scattering

The magnetic excitations in Ho$_2$Ir$_2$O$_7$ were studied by INS on the same polycrystalline sample as the one used for neutron diffraction and magnetometry measurements. The experiments were carried out at the ILL on the IN6 and IN4 neutron spectrometers. On the IN6 cold neutron time-of-flight spectrometer, the incoming neutron wavelength (energy) was 5.1 $\AA$ (3.145 meV). The energy resolution for elastic scattering was $\Delta E \approx 70$ µeV. On the IN4 thermal neutron time-of-flight spectrometer, the incoming neutron wavelengths (energies) used were 2.44 $\AA$ (13.74 meV), 1.3 $\AA$ (48.4 meV), and 0.9 $\AA$ (101 meV). The energy resolution for elastic scattering in the respective settings was $\delta E \approx 3.75$ and 9.5 meV. Results are shown and discussed in Supplementary Notes 1–2.

#### Data availability

All relevant data are available from the authors. Neutron scattering data performed at the ILL are available at the doi: 10.5291/ILL-DATA.5-31-2406 for neutron diffraction data and doi: 10.5291/ILL-DATA.4-01-1478 for INS data.
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