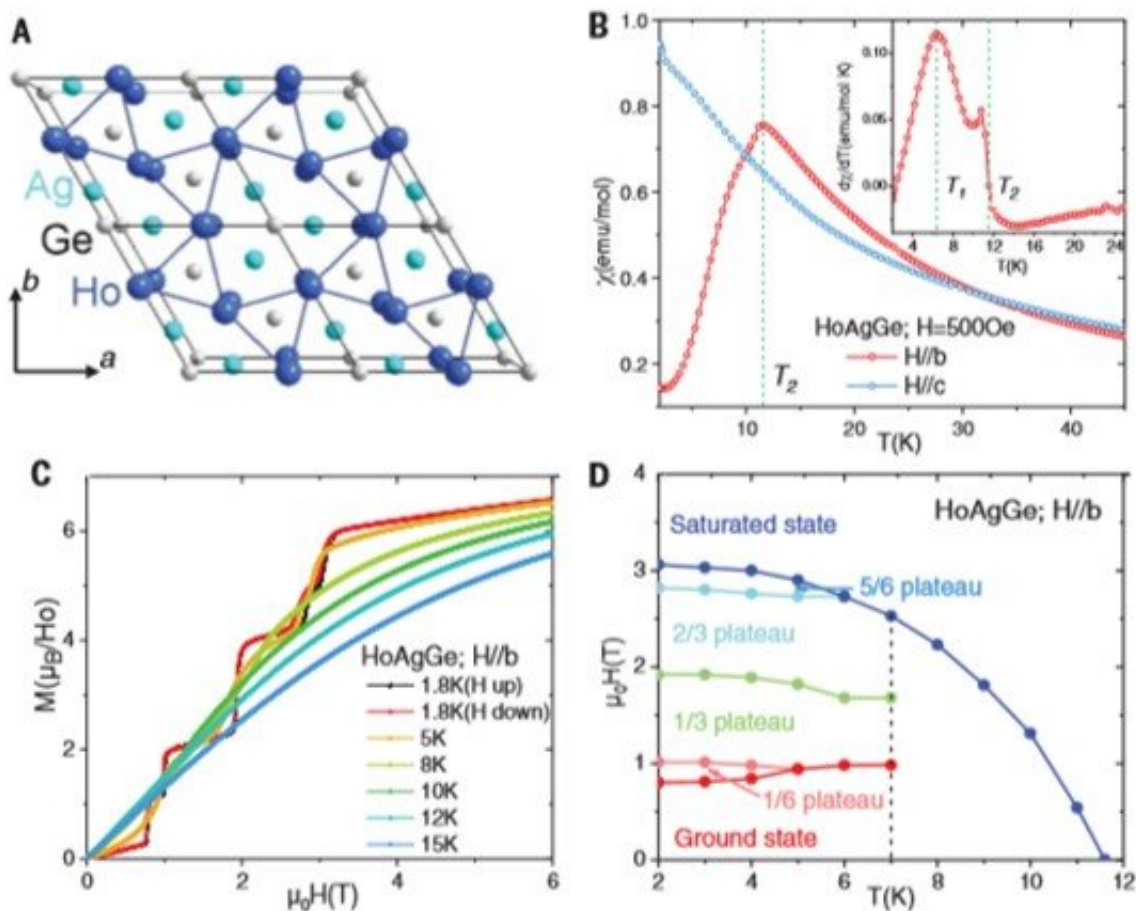


# Realizing kagome spin ice in a frustrated intermetallic compound

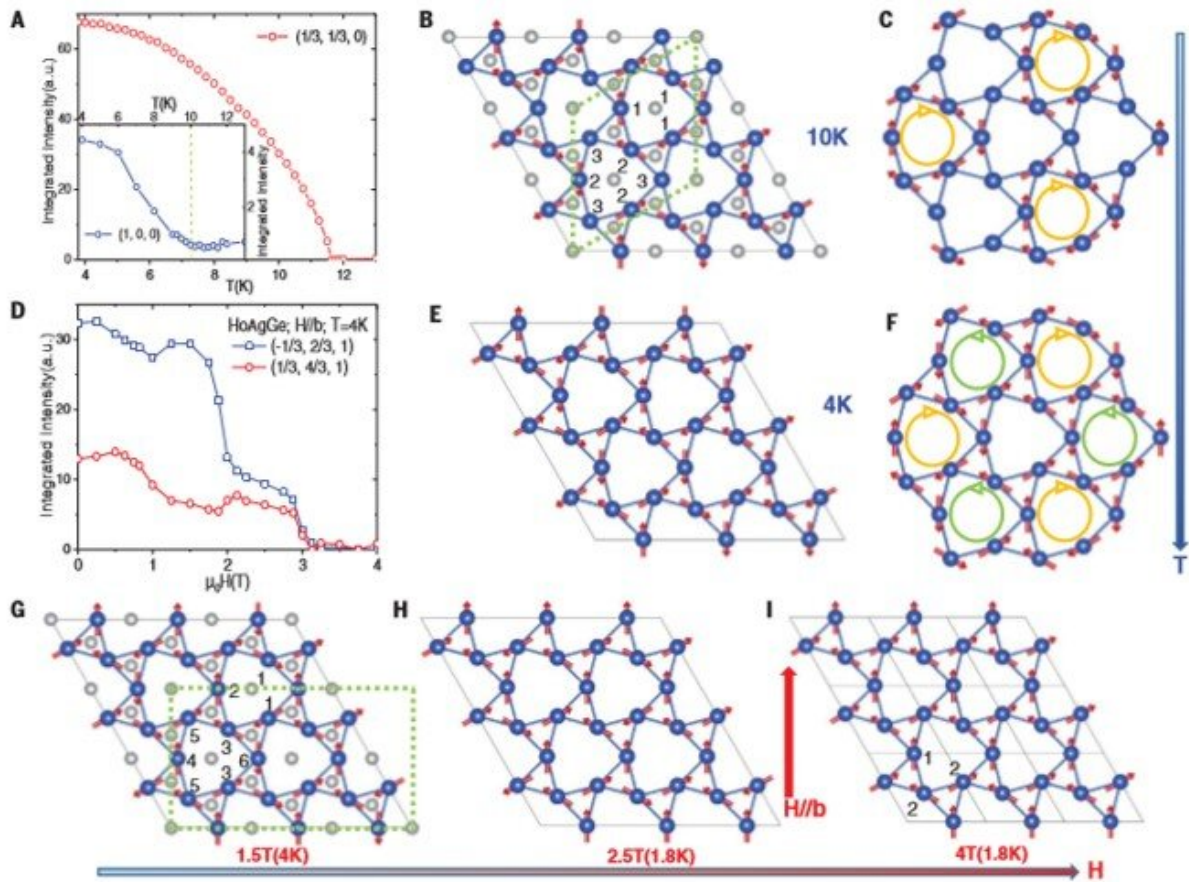
March 23 2020, by Thamarasee Jeewandara



Crystal structure and magnetic properties of HoAgGe. (A) c-Axis projection of the HoAgGe crystal structure, with the definition of  $a$  and  $b$  directions. (B) Low-temperature susceptibility  $\chi(T)$  of HoAgGe for both  $H//b$  and  $H//c$  under 500 Oe, with  $d\chi(T)/dT$  in the inset. (C) Isothermal in-plane ( $H//b$ ) magnetization for HoAgGe at various temperatures. (D) Dependence of the metamagnetic transitions on temperature, with the dotted line indicating  $T_1$  Credit: Science, doi:10.1126/science.aaw1666

Exotic phases of matter known as [spin ices](#) are defined by frustrated spins that obey local "ice rules"—similar to [electric dipoles](#) in water ice. Physicists can define ice rules in two-dimensions for in-plane [Ising](#)-like spins arranged on a [kagome lattice](#). The ice rules can lead to diverse orders and excitations. In a new report on *Science*, Kan Zhao and a team in experimental physics, crystallography, and materials and engineering in Germany, the U.S. and the Czech Republic used experimental and theoretical approaches including magnetometry, thermodynamics, neutron scattering and Monte Carlo simulations to establish the [HoAgGe](#) crystal as a crystalline system to realize the exotic kagome spin ice state. The setup featured a variety of partially and fully ordered states as well as field-induced phases at low temperatures consistent with the kagome experimental requisites.

Formation of exotic phases of matter can cause frustrations in spin systems. For example, local constraints in a molecule can lead to a macroscopic number of degenerate ground states or to an [extensive ground state in entropy](#). In two-dimensional setups, ice rules require [elaborate arrangements of spins](#) on triangular shaped kagome lattices. Consequently, the kagome spin ices showed multi-stage ordering behavior under changing temperature. Physicists had thus far only experimentally realized kagome spin ices in artificial spin ice systems formed by nanorods of ferromagnets organized into [honeycomb networks](#). In this work, Zhao et al. used multiple experimental and theoretical approaches to demonstrate the intermetallic compound HoAgGe as a naturally existing kagome spin ice with a fully ordered ground state.

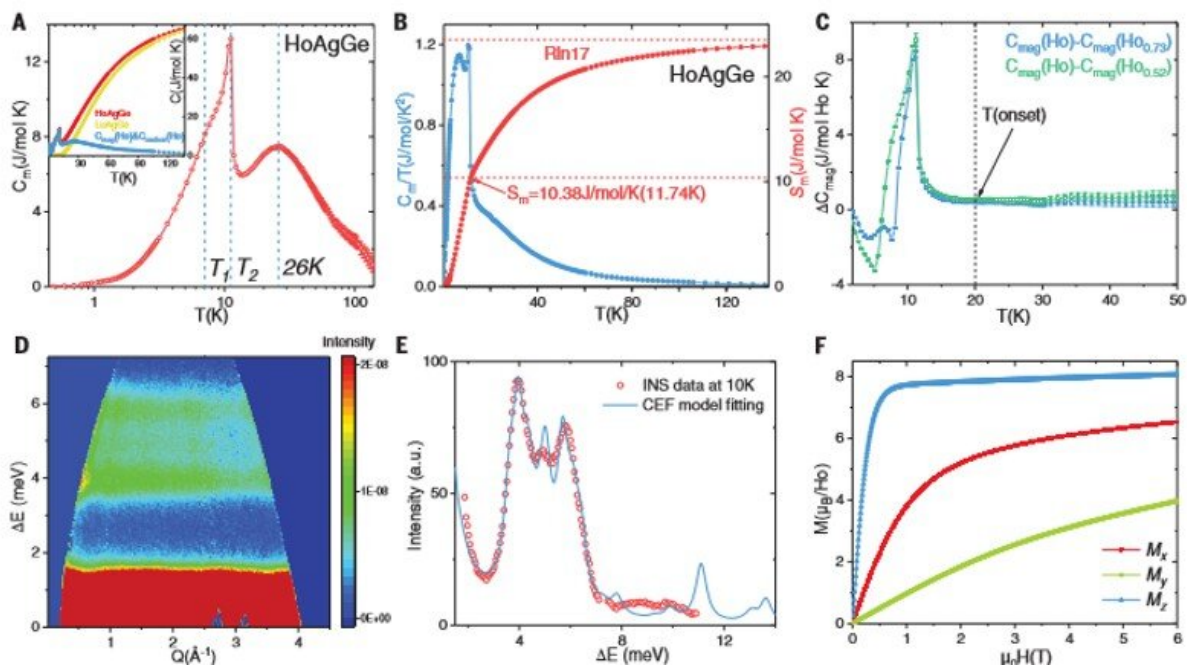


Magnetic structures of HoAgGe versus temperature and field with  $H/b$ . (A) Integrated intensity of the magnetic peak  $(1/3, 1/3, 0)$  from 13 K down to 3.8 K according to the neutron diffraction, with the integrated intensity of nuclear site  $(1, 0, 0)$  as an inset. (B) Refined magnetic structures of HoAgGe at 10 K. The magnetic unit cell is indicated by the green rhombus, with the three inequivalent Ho sites Ho1, Ho2, and Ho3 labelled by 1, 2, and 3, respectively, for simplicity. (C) Counter-clockwise hexagons of spins in the partially ordered structure of HoAgGe at 10 K, with  $1/3$  spins not participating in the long-range order. (D) Integrated intensity of magnetic peak  $(-1/3, 2/3, 1)$  and  $(1/3, 4/3, 1)$  versus field at 4 K. (E) Refined magnetic structure of HoAgGe at 4 K. (F) Clockwise and counter-clockwise hexagons of spins in the magnetic structure of HoAgGe at 4 K, which is exactly the expected  $\sqrt{3} \times \sqrt{3}$  ground state of kagome spin ice. (G) Refined magnetic structure of HoAgGe at  $H = 1.5$  T and  $T = 4$  K. The refinement was done in the  $3 \times \sqrt{3}$  light-green rectangle. The six inequivalent Ho sites are labelled by numbers 1 to 6 for simplicity. (H) Refined magnetic

structure of HoAgGe at  $H = 2.5$  T and  $T = 1.8$  K. (I) Refined magnetic structure of HoAgGe at  $H = 4$  T and  $T = 1.8$  K, with the two inequivalent Ho sites labelled by 1 and 2. The field direction is marked by the red arrow for (G) to (I). Credit: Science, doi:10.1126/science.aaw1666

The team then conducted structure and [magnetometry](#) measurements of HoAgGe. Although [neutron diffraction](#) measurements [conducted in the past](#) suggested noncollinear magnetic structures of HoAgGe—these experiments were based on powder samples that were insufficient to fully determine the magnetic structure in the presence of frustration. Zhao et al. combined neutron diffraction with thermodynamic measurements in single-crystalline HoAgGe to show its exotic temperature and magnetic-field dependent magnetic structures—consistent with the kagome ice rule. To fully determine magnetic structures from neutron diffraction based on nontrivial spin structures of HoAgGe, Zhao et al. performed single-crystal neutron diffraction experiments, down to 1.8 K. Below a high-temperature transition at 11.6 K, the team observed a magnetic peak.

When they refined the neutron data at 4 K, the team observed a more detailed [magnetic structure](#) where the fully ordered ground state indicated alternating clockwise and counter-clockwise hexagonal spins. The resulting  $\sqrt{3} \times \sqrt{3}$  ground state precisely represented the classical kagome spin ice, as [theoretically predicted](#). According to the [kagome ice rule](#), the dominating nearest-neighbor ferromagnetic coupling must occur between co-planar spins with site-dependent Ising-like uniaxial [anisotropy](#). In the present work, Zhao et al. calculated and confirmed Ising-like anisotropy of the crystalline electric field (CEF) for the HoAgGe crystals.



Magnetic specific heat and INS results of HoAgGe. (A) Magnetic contribution to the specific heat  $C_m$  of HoAgGe with the dotted lines indicating  $T_1$ ,  $T_2$ , and a broad peak at 26 K. Note that the error bars below 30 K are smaller than the symbol sizes. (Inset) Specific heat of HoAgGe, LuAgGe, and their difference. The latter is defined as the sum of the magnetic and the nuclear contributions to the specific heat of HoAgGe. (B)  $C_m/T$  data and the corresponding magnetic entropy  $S_m$ , which approaches the theoretical value of  $R\ln 17$  above 100 K. (C) Difference between the magnetic specific heat of HoAgGe and that of  $\text{Lu}_{1-x}\text{Ho}_x\text{AgGe}$  ( $x = 0.52$  and  $0.73$ ) after normalization (see text). (D) INS spectra of HoAgGe at 10 K with incident neutron wavelength 3 Å. (E) Constant  $Q$  cuts (1.4

To further confirm the authenticity of HoAgGe as a kagome spin ice, the research team investigated if established ice rules were applicable even outside the fully ordered ground state. Using [neutron diffraction](#) under magnetic fields they showed that HoAgGe satisfied these requisites and observed an increasing magnetic field with sudden changes during [metamagnetic transitions](#). For further information, Zhao et al. refined the magnetic structures obtained from [neutron scattering](#) and noted magnetic transitions to result from the competition between

the external magnetic field and weaker couplings that do not affect the ice rule.

After establishing that the kagome ice rule applied to HoAgGe crystals at low temperature, the team examined thermodynamic behaviors of kagome spin ice by isolating the magnetic contribution to specific heat by deducing contributions from the nuclei, lattice vibrations and itinerant electrons of the crystal. To determine the extent to which Ho ionic spins of the HoAgGe crystal could be viewed approximately as Ising, Zhao et al. next discussed the crystalline electric field (CEF) effects. To directly understand CEF splitting, they conducted inelastic neutron scattering (INS) experiments of HoAgGe crystals using the advanced [time-of-flight spectrometer](#). The results indicated four low-energy CEF modes showing Ising-type anisotropy.

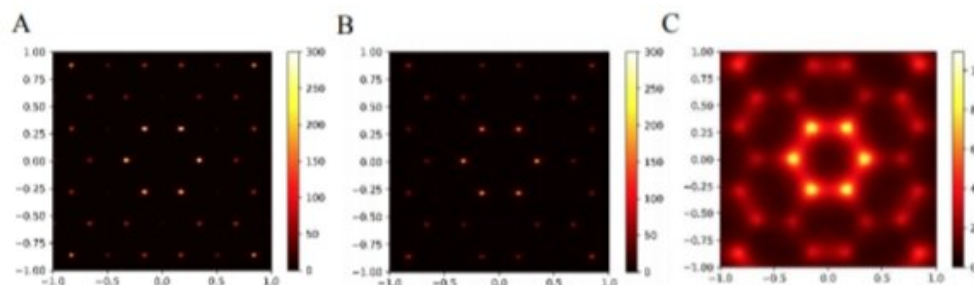
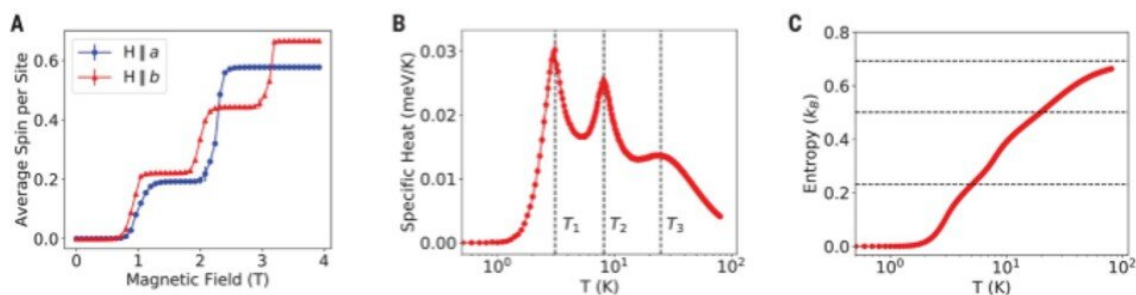


Fig. S19: Magnetic structure factor from Monte Carlo simulations in an  $18 \times 18$  unit cell at (A)  $T=1$  K, (B)  $T=5$  K, and (C)  $T=15$  K, respectively. The horizontal and the vertical axes are respectively  $(H, H, 0)$  and  $(-K, K, 0)$ , similar as in Fig. S5.

Magnetic structure factor from Monte Carlo simulation in an  $18 \times 18$  unit cell at (A)  $T=1$  K, (B)  $T=5$  K and (C)  $T=15$  K. The horizontal and vertical axes were respectively  $(H, H, 0)$  and  $(-K, K, 0)$ . Credit: Science, doi:10.1126/science.aaw1666

Based on the experimental evidence, they proposed a classical spin model containing Ising-like in-plane spins on a 2-D distorted [kagome lattice](#). Using Monte Carlo simulations of the classical spin model on an  $18 \times 18$  lattice, they reproduced the ground state and partially ordered state to capture the classical

spin model and the main characteristics of the HoAgGe magnetism at low temperatures. The model developed in the study differed from both dipolar and short-range kagome ice cases relative to exchange couplings and long-range dipolar interactions, with further investigations requiring a separate study.



Monte Carlo simulations of the 2D classical spin model for HoAgGe. (A) M(H) curves at 1 K for H along the a and b axes. (B) Temperature dependence of the specific heat per spin. (C) Magnetic entropy per spin calculated from the specific heat. The three horizontal dashed lines correspond to  $\ln 2 \approx 0.693$  (paramagnetic Ising), 0.501 (short-range ice order), and  $1/3 \ln 2 \approx 0.231$  (toroidal order), respectively. An  $18 \times 18$  cell was used for the calculation. Credit: Science, doi:10.1126/science.aaw1666

In this way, the Monte Carlo simulations of the classical spin model only partially agreed with the experiments. The discrepancy may have resulted from multiple, low-lying CEF levels of the Ho<sup>3+</sup> ions. In HoAgGe, the metallicity simultaneously suppressed CEF splitting of Ho<sup>3+</sup> ions to enhance exchange coupling between them, making the two energy scales comparable to low-lying CEF levels. The resulting semi-classical model can still be mapped to an Ising model, thereby explaining the validity of the experiment. Compared to other [pyrochlore](#) spin ices, the metallic nature of HoAgGe made it a high-temperature [kagome](#) ice and may also lead to further exotic phenomena, including interactions between electric currents and magnetic monopoles as well as [metallic magnetoelectric effects](#).

**More information:** Kan Zhao et al. Realization of the kagome spin ice state in

a frustrated intermetallic compound, *Science* (2020). [DOI: 10.1126/science.aaw1666](https://doi.org/10.1126/science.aaw1666)

Leon Balents. Spin liquids in frustrated magnets, *Nature* (2010). [DOI: 10.1038/nature08917](https://doi.org/10.1038/nature08917)

Gia-Wei Chern et al. Magnetic charge and ordering in kagome spin ice, *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* (2012). [DOI: 10.1098/rsta.2011.0388](https://doi.org/10.1098/rsta.2011.0388)

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