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Spin-orbit entangled local moments in the iridate material Na_2IrO_3 are subject to strong exchange frustration, driving the system towards a spin-liquid phase with emergent fractional excitations.

Philipp Gegenwart and Simon Trebst

The subtle interplay of spin-orbit coupling and electronic correlations in ‘heavy’ transition-metal compounds with $4d$ and $5d$ elements has brought about a recent flurry of discoveries of novel types of quantum matter. A dominant spin-orbit coupling induces non-trivial topological features in the band structure, giving rise to topologically insulating behaviour, for example. However, novel physics can also arise in the correlated Mott regime. One example is the formation of local moments that are not conventional magnetic degrees of freedom, but spin-orbit entangled moments^{1,2}.

Interest in such spin-orbit entangled Mott insulators has been sparked by the theoretical proposal that for some lattice geometries the microscopic exchange between these effective moments might realize bond-directional interactions that couple different components of a moment along different spatial directions³. Bond-directional interactions of this form are the essential building blocks for

unconventional magnetism in the celebrated Kitaev model⁴. Writing in *Nature Physics*, Sae Hwan Chun and colleagues⁵ report direct experimental evidence of such Kitaev exchange interactions in Na_2IrO_3 , by means of X-ray scattering and spectroscopy techniques.

Na_2IrO_3 is a layered iridate, in which the Ir^{4+} ions carrying the spin-orbit entangled $j = 1/2$ moment form quasi-two-dimensional honeycomb layers (Fig. 1a). Upon cooling, characteristic zigzag correlations between neighbouring $j = 1/2$ moments are observed. These short-ranged and fluctuating zigzag arrangements of local moments are found to propagate along the three principal directions of the honeycomb lattice. Most remarkably, however, Chun *et al.*⁵ report that each fluctuation pattern along one of the three spatial directions is associated with one distinct spin quantization axis. This observation manifests a pronounced entanglement between the spin component and real-space direction. It is a rare showcase of spin-orbit coupling, not

realized in ordinary magnetic materials, and it constitutes direct evidence of Kitaev-type bond-directional exchange interactions dominating over other types of magnetic exchange.

Bond-directional interactions induce strong exchange frustration between the effective moments, as these cannot simultaneously align with all neighbours preferring distinct quantization axes (Fig. 1b). Exchange frustration in such a scenario can prohibit even a ferromagnetically coupled system from undergoing a finite-temperature ordering transition. This results in a highly correlated, but strongly fluctuating, low-temperature regime — a quantum spin liquid. For the tri-coordinated honeycomb lattice the bond-directional exchanges directly realize the Kitaev model⁴ — a quintessential spin model that harbours gapped and gapless quantum spin liquids with emergent fractional excitations obeying non-Abelian statistics. The Kitaev model stands out because of these unusually rich spin-liquid ground states, but it is also unique because it is one of the very few examples of an interacting spin model that can be rigorously solved. It has thereby become the gold standard for spin-liquid physics and associated topological phenomena in a microscopic model.

To probe the unconventional magnetism resulting from Kitaev-type exchange interactions, Chun *et al.*⁵ used an advanced experimental instrumentation that allows the study of magnetic fluctuations from diffuse scattering with unprecedented resolution and sensitivity to spin orientation. The incident photon energy of synchrotron radiation has been fine-tuned to the iridium L_3 absorption edge. During the photon-in–photon-out process, a substantial resonant enhancement of the scattering cross-section makes it possible to study the inelastic excitations over the entire Brillouin zone. Furthermore, it allows the scattering from fluctuating zigzag clusters to be resolved — a signal that is orders of magnitude weaker than elastic scattering from static long-range

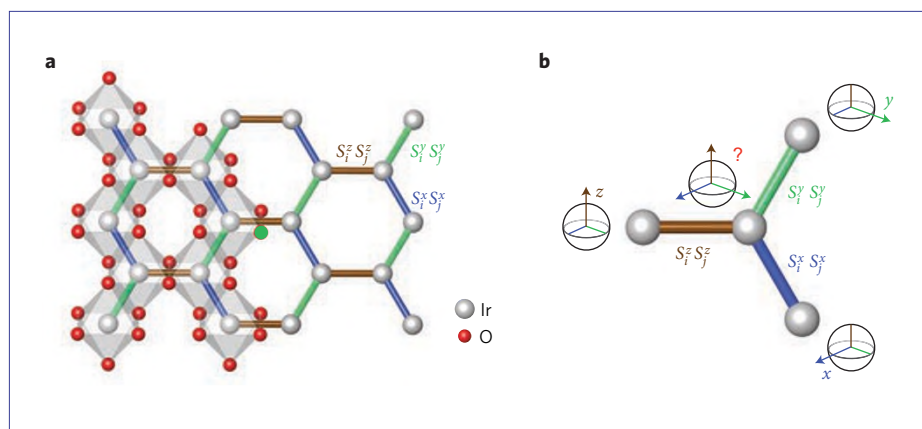


Figure 1 | Spin-orbit coupling in Na_2IrO_3 . **a**, The spin-orbit entangled $j = 1/2$ moments form layered honeycomb structures with bond-directional interactions that couple different components of the moments, S , for neighbouring sites, i and j , along the three principal lattice directions: x (blue), y (green) and z (brown). The bond-directional exchanges thereby implement a Kitaev model. **b**, Even a ferromagnetic bond-directional interaction yields a high-level of exchange frustration between the magnetic moments, whose orientation along one of the principal directions in spin space (illustrated by the spheres) is indicated by the arrows. The frustration effectively prohibits a finite-temperature ordering transition.

ordering. To selectively probe scattering from the different spin components, Chun and colleagues⁵ have carefully probed the polarization angle dependence with the spin-component scattering intensity largely exceeding that from spin-averaged correlations, and have thereby found a clear fingerprint of the Kitaev-type bond-directional interactions.

At very low temperatures, Na_2IrO_3 displays a static long-range ordering where one of the three zigzag ordering vectors is selected. However, the size of the ordered moment is reduced compared with the bare moment size. Indeed, diffuse scattering at temperatures below

the Néel temperature proves that zigzag fluctuations are persisting even at the very lowest temperatures, which indicates that these fluctuations are quantum rather than thermally driven.

An interesting open question for future experimental and theoretical consideration is related to the group's observation of magnetic fluctuations in an unusually wide energy range up to 100 meV. These experimental findings might suggest that zigzag correlations with intrinsic energy scales of about 5 meV emerge from Kitaev-type interactions, possibly much larger than anticipated. Or there might be other spin-orbit physics at

play in these Mott insulators that is yet to be explored.

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