# A New Spin-Anisotropic Iridate

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## Introduction

The physics of Mott insulators underlies diverse phenomena ranging from high temperature superconductivity to exotic magnetism. Although both the electron spin and the structure of the local orbitals play a key role in this physics, in most systems these are connected only indirectly via the Pauli exclusion principle and the Coulomb interaction. Iridium-based oxides (iridates) open a further dimension to this problem by introducing strong spin-orbit interactions, such that the Mott physics has a strong orbital character. In the layered honeycomb iridates this is thought to generate highly spin-anisotropic interactions, coupling the spin orientation to a given spatial direction of exchange and leading to strongly frustrated magnetism. The potential for new physics emerging from such interactions has driven much scientic excitement, most recently in the search for a new quantum spin liquid, first discussed by Kitaev [1]. Here we report a new iridate structure that has the same local connectivity as the layered honeycomb, but in a three-dimensional framework. The temperature dependence of the magnetic susceptibility exhibits a striking reordering of the magnetic anisotropy, giving evidence for highly spin-anisotropic exchange interactions. This compound thus provides a unique and exciting glimpse into the physics of a new class of strongly spin-orbit coupled Mott insulators.

## Experimental

We have synthesized single crystals of a polytype of Li2IrO3 in which we reveal the effect of the spin-anisotropy of exchange across the Ir-O2-Ir bond from the temperature dependence of the anisotropic magnetic susceptibility, using a combination of magnetic susceptibility and torque magnetometry techniques at high fields.

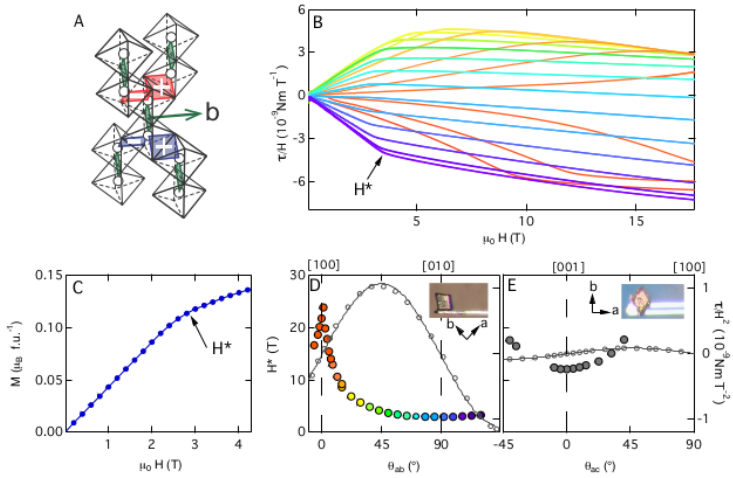


Figure 1: Low temperature magnetic properties of the Hh1i-Li2IrO3. (A) The Ir-O2-Ir planes defining three orthogonal directions of the spin-exchange, one parallel to ^b and the other two parallel to ^a -^c, labelled + and - (^a is the unit vector along a). This connects to the rotation used to describe the Kitaev Hamiltonian in SI III. (B) Torque signal divided by the applied magnetic eld H illustrating a linear low-field dependence and a kink at H*, which is strongly angle dependent (colors correspond to angles shown in (D)). (C) Magnetization vs magnetic field applied along the b-axis at a temperature of 15 K. (D) & (E) The angle dependence ab=ac of the kink field H$^{\perp}$ of the ordered state (full circles, left axes) with respect to the crystallographic axes a; b and c. H* is correlated to the magnetization anisotropy ij (open circles, right axes) indicating a common moment at H* in all field orientations.

## Conclusions

The observed ten fold increase in b cannot be driven by the g-factor of the local iridium environment, whose geometric constraints are temperature independent. The temperature dependence of b must therefore arise from spin-anisotropic exchange. We note that all the c-axis bonds have the Ir-O2-Ir plane normal to the b-axis, whether they preserve or rotate between the two honeycomb orientations (see green shading Fig. 1A indicate the Ir-O2-Ir planes). This is the only Ir-O2-Ir plane that is normal to a crystallographic axis, providing evidence for spin-anisotropic exchange across these links. This may arise from the mechanism suggested by JK in the context of the Kitaev model [2].

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## References

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[2] J. Chaloupka, G. Jackeli, and G. Khaliullin, Phys. Rev. Lett. 105, 027204 (2010).