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Magnetic and thermodynamic properties of the harmonic honeycomb iridates

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Spin and orbital quantum numbers play a key role in the physics of Mott insulators, but in most systems they are connected only indirectly—via the Pauli exclusion principle and the Coulomb interaction. Iridium-based oxides (iridates) introduce strong spin–orbit coupling directly, such that these numbers become entwined together and the Mott physics attains a strong orbital character. In the layered honeycomb iridates this is thought to generate highly spin–anisotropic magnetic interactions, coupling the spin to a given spatial direction of exchange and leading to strongly frustrated magnetism. Here we report a new iridate structure that has the same local connectivity as the layered honeycomb and exhibits striking evidence for highly spin–anisotropic exchange. The basic structural units of this material suggest that a new family of three-dimensional structures could exist, the 'harmonic honeycomb' iridates, of which the present compound is the first example.