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Title: Crystal Growth and Magnetic Property Study of layered honeycomb materials with Kitaev-like

interactions

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Keywords: honeycomb materials

quantum magnets thermodynamic electrical transport Crystal growth

Issue

1-Sep-2018

Date:

Publisher: IISERM

Abstract:

Kitaev-like bond-directional exchange interactions are novel and quite different from the ubiquitous Heisenberg interactions found in most magnets. These kind of interactions open up new possibilities in the exploration and designing of new quantum magnets which can host novel quantum ground states like spin-liquids. However, Kitaev-like in- teractions have remained only a theoretical construct until recently. Honeycomb iridates A 2 IrO 3 (A = Na, Li), offer potential realizations of such novel exchange coupling. In this thesis I have synthesized single crystals of A 2 TO 3 (A = Na, Li, K, and T = Ir, Ru) and studied their electrical transport, magnetic, and thermal properties. Our work provides several new results: (i) the first thermodynamic evidence of possible fractionalization of electrons in Na 2 IrO 3 because of proximity to the Kitaev spin-liquid state, (ii) evidence through magnetic impurity doping, of fragile magnetic order and importance of nearestneighbour interactions and spin-orbit coupling in deciding the magnetic ground state in Na 2 IrO 3, (iii) a novel method (reactive ion etching) of surface doping Na 2 IrO 3 and possibly other layered oxides has been discovered. The surface conductivity of Na 2 IrO 3 crystals could be increased by 11 orders of magnitude by varying etching times. The samples which turned metallic show transport anomalies consistent with charge density wave or structural instabilities, (iv) first crystal growth of Li 2 RuO 3 is reported where the Xtals crystallize in the P2 1 /m structure and show the expected high temperature magneto-structural transition, and (v) design and crystal growth of a new layered honey- comb lattice iridate K 2 IrO 3 with an interlayer separation between Ir honeycomb planes which is more than a factor of 2 larger than in Na 2 IrO 3. Magnetic measurements on crys- tals reveal localized effective spin S = 1/2 interacting strongly θ = -210 K but without magnetic order down to 1.8 K. Thus K 2 IrO 3 is a new Kitaev spin liquid candidate

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