Raman studies of two-compound spin-liquid candidate $(Na_{1-x}Li_x)_2IrO_3$

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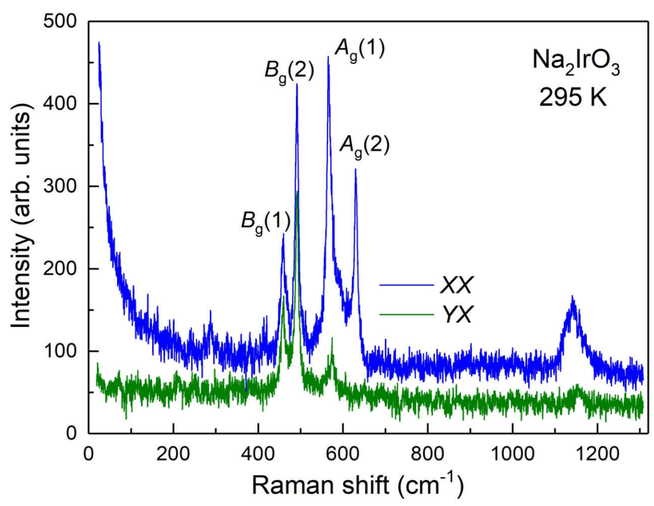
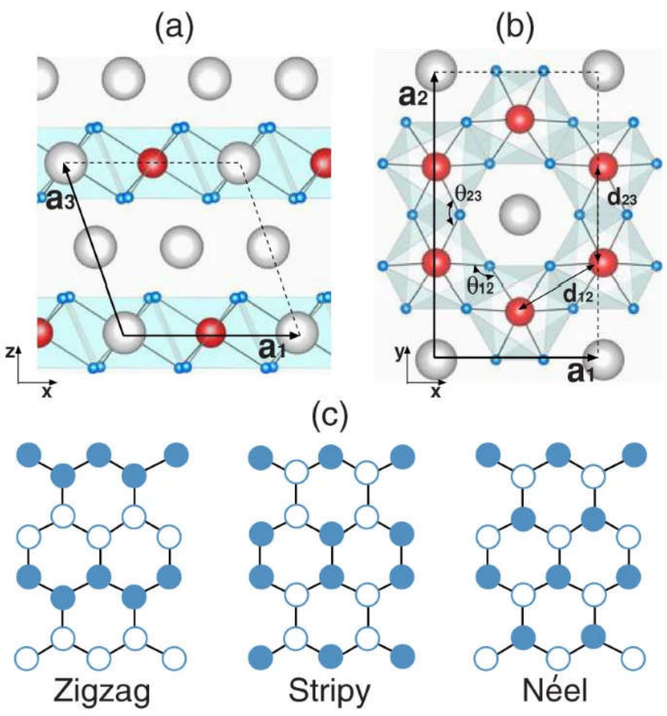
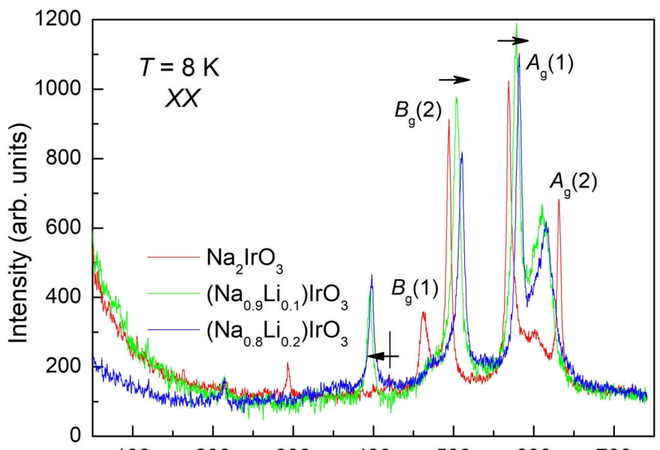
The purposeful search for real crystalline compounds in which the Kitaev spin-liquid state can be realized and their study has recently been one of the urgent tasks of solid-state physics and magnetism. The prospect of the possible implementation of these materials in the field of quantum computers and quantum communications makes these efforts relevant. The present work was aimed at obtaining important information about the structure of the ground and low-energy excited states of the magnetic subsystem in the crystals of the two-dimensional alkali metal iridate family and the manifestation of spin-liquid properties in this structure, in particular, magnetic excitations with fractional torque and flux visions. Raman light scattering in a Mott insulator with spin-orbit interaction in a cellular lattice, the system $(Na_{1-x}Li_x)_2IrO_3$ (x = 0 - 0.2), was carried out.



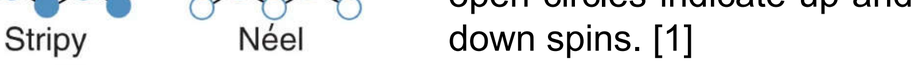
Raman spectra of Na_2IrO_3 measured in two polarization geometries at room temperature.
$A_g(1)$ - Stretching vibrations of IrO6;
$A_g(2)$ - Bending vibrations of IrO6 + Out-of-phase vibrations of Ir atom along the z-axis + Out-of-phase vibrations of Na atoms along the z-axis



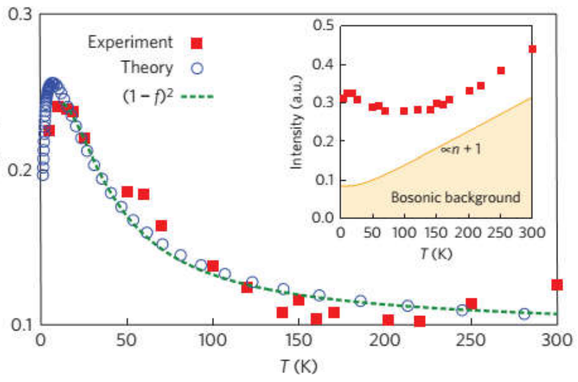
Crystal structure of Na_2IrO_3. (a) projection on the *xz* plane and (b) projection on the *xy* plane. $\mathbf{a}_1$, $\mathbf{a}_2$ and $\mathbf{a}_3$ d enote unit vectors of the unit cell. The large, medium and small circles represent Na, Ir and O atoms, respectively. Three different AFM structures with the zigzag, stripy and Néel spin orders are schematically shown in (c), where the solid and open circles indicate up and down spins. [1]



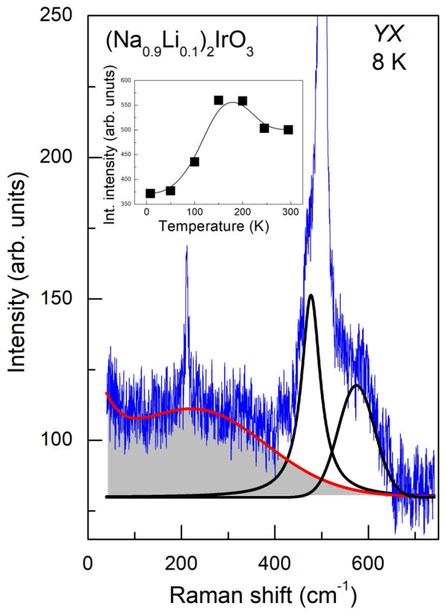
Raman spectra of single crystals of $(Na_{1-x}Li_x)_2IrO_3$ (x = 0, 0.1, 0.2) were measured in the XX polarization geometry at a temperature of 8 K. The arrows show the shift of selected phonon lines in the spectra upon doping Na_2IrO_3 with lithium.



Schematic pictures for the Kitaev model and Raman processes. **a**, Honeycomb lattice structure. Blue, green and red bonds represent Ising-like interactions between *x*-, *y*- and *z*-components of the *S*=1/2 spins, respectively. Incoming and outgoing photons, whose frequencies are ω_i and ω_f, are also depicted. **b**,**c**, Feynman diagrams of the Raman scattering processes. [2]



Comparison between the numerical results and the experimental data for α-$RuCl_3$. [2]



YX раманівський спектр монокристала $(Na_{0.9}Li_{0.1})_2IrO_3$ при температурі 8 К. Магнітний континуум зафарбований сірим. На вставці наведена температурна залежність інтенсивності континуума.

New data on the temperature behavior, as well as the doping dependence of the phonon line parameters (intrinsic energy, intensity and width) and their spectral shape were obtained. A quasi-critical behavior of the phonon system with the replacement of sodium by lithium in the $(Na_{1-x}Li_x)_2IrO_3$ system was revealed, which is associated with the displacement of oxygen in the IrO_6 octahedra, which in turn leads to a significant variation of spin correlations. The detected anomalies in the phonon spectra correlate with the features of magnetic excitations, which indicates a strong coupling of the lattice and magnetic subsystems in $(Na_{1-x}Li_x)_2IrO_3$. The obtained results are important for understanding the influence of the lattice topology on the spin and, as a consequence, the formation of the spin-liquid state in materials - candidates for Kitaev physics.

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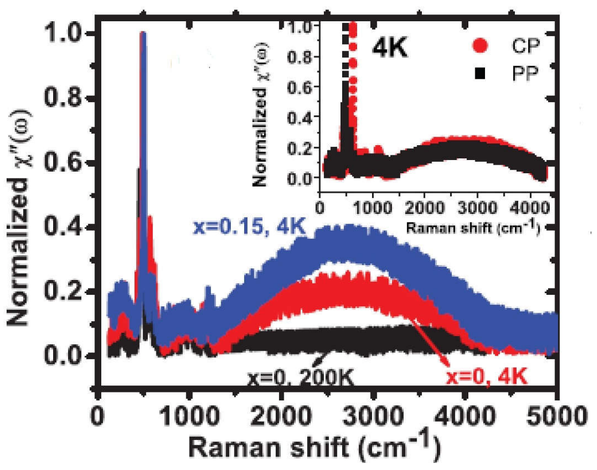

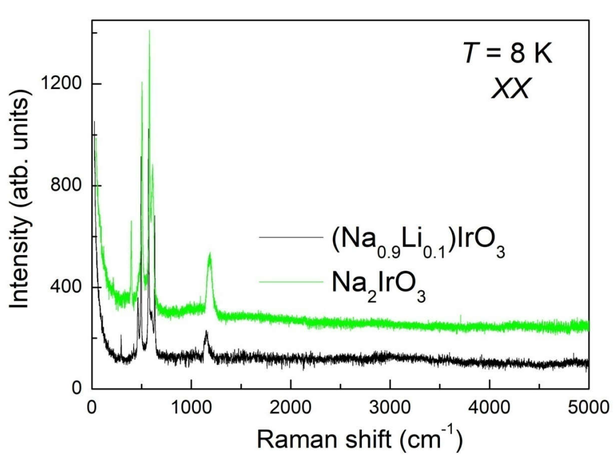

(left) Raman spectra of single crystals of $(Na_{1-x}Li_x)_2IrO_3$ at 4 K (for x = 0, 0.15) and 200 K (for x = 0) [3]. (right) Raman spectra of samples with x = 0 and 0.1 at 8 K obtained in our studies.

References
[1] Kim, H.J., Lee, J.H. & Cho, J.H. Antiferromagnetic Slater Insulator Phase of Na_2IrO_3. *Sci Rep* **4**, 5253 (2014).
[2] Nasu, J., Knolle, J., Kovrizhin, D. *et al.* Fermionic response from fractionalization in an insulating two-dimensional magnet. *Nature Phys* **12**, 912–915 (2016).
[3] Gupta, S.N., Sriluckshmy, P.V. *et al.* Raman signatures of strong Kitaev exchange correlations in $(Na_{1-x}Li_x)_2IrO_3$: Experiments and theory *EPL*, **114**, 47004 (2016).