Lattice-TunedMagnetism of Ru⁴⁺(4d⁴) Ions in Single-Crystals of the Layered Honeycomb Ruthenates: Li₂RuO₃and Na₂RuO₃

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We synthesize and study single crystals of the layered honeycomb latticeMott

insulatorsNa₂RuO₃ and Li₂RuO₃ with magnetic Ru⁴⁺(4d⁴) ions. The newly found

Na₂RuO₃features a nearly ideal honeycomb lattice and ordersantiferromagnetically at 30

K. Single-crystalsof Li₂RuO₃adopta honeycomb lattice witheitherC2/m or more distorted

P2₁/m below 300 K, depending on detailed synthesis conditions. We find that Li₂RuO₃ in

both structureshostsa well-defined magnetic state, in contrastto the singlet ground state

found in polycrystalline Li₂RuO₃. A phase diagramgenerated based on our results

uncoversa new, direct correlation between the magnetic ground state and basal-plane

distortions inthehoneycomb ruthenates.

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IntroductionIt has been of great interest to study interacting electrons on the honeycomb lattice in various contexts both experimentally (e.g. graphene) and theoretically (e.g the Kitaev model). Studies of honeycomb materials have intensified in recent years [1-19]in part because strong spin-orbit coupling (SOC) along with other competing interactions and geometric frustration in the honeycomb iridates Na₂IrO₃ and Li₂IrO₃favorsa highly anisotropic Kitaev interaction[20] that stabilize exotic ground states such as topological spin-liquids[1].It is now experimentally established that Na₂IrO₃ exhibits a peculiar zigzag magnetic order at T_N=18 K[5, 14, 15], and Li₂IrO₃ also orders at T_N=15 K but with a different ground stateyet to be defined [3, 17, 21, 22, 23]. Indeed, for $(Na_{1-x}Li_x)_2IrO_3$ with $0 \le x \le 0.90$, the measured phase diagram demonstrates a dramatic suppression of T_N at intermediate x suggesting that the magnetic order in Na₂IrO₃ and Li₂IrO₃ is different;however, no spin liquid has been observed thus far[17]. Our pursuit of an understanding of the honeycomb iridates has led us to their ruthenate counterparts, Na₂RuO₃ and Li₂RuO₃. These materials feature Ru⁴⁺(4d⁴) ions and a weaker or "intermediate strength" SOC (~ 0.16 eV, compared to ~ 0.4 eV for Ir ions) [24]. The different d-shell filling and contrasting hierarchy of energy scales between the ruthenates and iridates provide a unique opportunity for a deeper understanding of the fundamental problem of interacting electrons on the honeycomb lattices. The magnetism of Ru⁴⁺ ions as well as other heavy "d⁴ ions" (such as Rh⁵⁺(4d⁴), Re³⁺(5d⁴), Os⁴⁺(5d⁴) and Ir5+(5d4))is interesting in their own right, as emphasized recently[25]. Materials with heavyd⁴ ions tend to adopt a low-spin state because larger crystal fields often overpower the Hund's rule coupling. On the other hand, SOC with the intermediate strength may still be strong enough to impose a competing, singlet ground state or an angular momentum J=0 state. Novel magnetic states may thus emerge when the singlet-triplet splitting (0.05-0.20 eV) becomes comparable to exchange interactions (0.05-0.10 eV) and/or non-cubic crystal fields[25-27]. This is evidenced in a recent study of materials containing 5d⁴ ions [28].

Up until now, no physical and structural properties of Na₂RuO₃ have been investigated but a few experimental and theoretical studies of polycrystalline Li₂RuO₃ have been reported in recent years [29-32]. In essence, polycrystalline Li₂RuO₃ undergoes a structural phase transition near T_D=540 K that features a change of space group from C2/m (No. 12) at high temperatures to P2₁/m (No. 11) at low temperatures. The low-temperature phase adopts a strongly distorted honeycomb lattice, which prompts a simultaneous dimerization that results in a singlet ground state [29]. The observation of dimerized zigzag chains has recently stimulated more investigations of Li₂RuO₃[30-32], in which the dimerization is attributed to orbital ordering [29], creation of valence bond crystal [30] andJahn-Teller distortions [31], respectively. It is noted that all reported experimental results were culled from polycrystalline Li₂RuO₃[29, 31, 32].

Here we report structural, magnetic, and thermal properties of single-crystal Li₂RuO₃ and Na₂RuO₃. The newly found Na₂RuO₃ with space group C2/mfeatures a nearly ideal honeycomb lattice and orders antiferromagnetically below 30 K. It may serve as a reference for almost perfect honeycomb symmetry. On the other hand, single-crystal Li₂RuO₃ adopts a less ideal honeycomb lattice with either C2/m or more distorted P2₁/m below 300 K but both phases exhibit a well-defined, though different, magnetic state, which sharply contrasts with the singlet ground state due to dimerization observed in polycrystalline Li₂RuO₃[28]. This work produces a phase diagramthat uncovers a direct

correlation between the ground state and basal-planedistortions or lattice-tuned magnetismin all honeycomb ruthenates studied.(Both Li₂RuO₃ and Na₂RuO₃ are highly insulating; their transport properties are not included in this paper.)

Crystal Structures Single-crystals of Li₂RuO₃ and Na₂RuO₃ were synthesized using the self-flux method, which is described elsewhere [17]. For synthesis of single-crystal Li₂RuO₃the mixed chemicals were first heated up to 1250 °C and then cooled to 900 °C at 2 °C/hour and finally room temperature at 50 °C/hour. In contrast, the polycrystalline Li₂RuO₃ was synthesized at much lower temperature of 950 °C.Thedifferent synthesis conditionsmay have important implications for the ground state of Li₂RuO₃.For more experimental details, see Supplemental Material [33]. Crystal structures on which the ground state so sensitively hinges require a close examination. Table 1 includes the lattice parameters of single-crystals Li₂RuO₃ and Na₂RuO₃ as well as those of polycrystalline Li₂RuO₃ and iridate counterparts for contrast and comparison. For the sake of discussion, single-crystal Li₂RuO₃ with C2/m and P2₁/m are labeled as Li₂RuO₃ (C) and Li₂RuO₃ (P), respectively. A major distinction between Li₂RuO₃ (C) and Li₂RuO₃ (P) is the number of unequal Ru-Ru bond distances, which measures distortions that in turn dictate the ground state. Li₂RuO₃ (C)features two bond distances, or a long and short one, L₁ and L_s, respectively, whereas Li₂RuO₃ (P)has three bond distances, i.e., L_L L_s and a medium bond distance, L_m. The basal-plane distortion is characterized by the bond difference ratio defined as $(L_1-L_s)/L_s$, which is shown in **Table 1,Figs 1a** and **1b**. In general,honeycomb lattices with C2/mtend to have a largera-axis lattice parameter and smaller ratio $b/a(\sim\sqrt{3})$ than those with P2₁/m, thus less distorted. Figs.1c and 1ddemonstrate the lattice parameters of single-crystal and polycrystalline samples as a function of temperature. As

seen,no structural transition is discerned in the single crystals studied forthe temperature range measured. In short, the structural differences between the polycrystalline Li_2RuO_3 and Li_2RuO_3 (C) or Li_2RuO_3 (P) are distinguished by the different space groups or by the difference in $(L_l-L_s)/L_s$. It is clear that Li_2RuO_3 (P) is more distorted than Li_2RuO_3 (C) but much less distorted than the polycrystalline sample despite the same space group shared by both (Table 1).

Table 1. Structural comparison between the honeycomb lattices at 100 K

Compound	Space Group	a (Å)	b (Å)	b/a	$(\mathbf{L_{l}}\mathbf{-L_{s}})/\mathbf{L_{s}}$
Li ₂ RuO ₃ (Powder)*	$P2_1/m$	4.9210(2)	8.7829(2)	1.785	18.6%
$Li_2RuO_3(P)$	$P2_1/m$	4.963(3)	8.766(6)	1.766	10.1 %
$Li_2RuO_3(C)$	C2/m	5.021(4)	8.755(6)	1.744	2.1 %
Na ₂ RuO ₃	C2/m	5.346(1)	9.255(2)	1.731	0.17 %
$(Li_{0.9}Na_{0.1})_2IrO_3$	C2/m	5.186(1)	8.964(2)	1.728	0.6 %
Na_2IrO_3	C2/m	5.319(1)	9.215(2)	1.732	0.14 %

^{*} Taken at 300 K

Physical Properties Na₂RuO₃ exhibits a sharp antiferromagnetic (AFM) transition at T_N =30 K, as shown in **Fig. 2a**. The magnetic anisotropy leads to a stronger out-of-plane magnetic susceptibility χ_{\perp} than in-plane magnetic susceptibility χ_{\parallel} . The linearity illustrated in $1/\Delta\chi_{\parallel}$ (right scale in **Fig. 2a**) indicates that the data fit well with the Curie-Weiss law for 100 < T < 350 K, and yield the Curie-Weiss temperature $\theta_{CW} = -137$ K and effective moment $\mu_{eff} = 2.45$ μ_B/Ru (**Table 2**). The frustration parameter defined as FP = $|\theta_{CW}|/T_N$ is estimated to be 4.6. This value suggests a presence of modest frustration, comparable to that for its iridate counterpart.

The magnetic ordering is confirmed by the specific heat C(T) (**Fig. 2b**). However, an additional peak at $T_{N2} = 26$ K that is absent in $\chi(T)$ is also seen in C(T). This behavior, which is reproducible, is remarkably similar to that observed in Na_2IrO_3 where an additional, weaker anomaly in C(T) is discerned at $T^* = 21$ K that is followed by the

zigzag order at T_N = 18 K [15, 17]. This two-step transition is discussed in context of the Kitaev-Heisenberg model on the hexagonal lattice [34]. A similar argument could be applied to Na₂RuO₃ although the origin of this magnetic behavior needs to be further investigated. The C(T) data also indicate that the entropy removal due to the two-step magnetic transition is small, less than 10% of Rln3 expected for an S=1 magnet. This implies that the magnetic ordering may not be fully developed perhaps in part because of the tendency of SOC to impose a singlet state. Application of magnetic field up to 14 T causes no visible changes in both C(T,H) and χ (T,H).

Table 2. Physical parameters of the single-crystal honeycomb lattices

Compound	$T_{N}(K)$	$\theta_{\text{CW}}(\mathbf{K})$	FP	$\mu_{\rm eff}$ ($\mu_{\rm B}/{ m Ru}$ or Ir)
Li ₂ RuO ₃ (P)	~ 5	-58	11.6	1.46
$Li_2RuO_3(C)$	9	-112	12.4	2.77
Na ₂ RuO ₃	30	-137	4.6	2.45
$(Li_{0.9}Na_{0.1})_2IrO_3$	7	-18	2.6	1.95
Na ₂ IrO ₃	18	-119	6.6	1.76

FP stands for frustration parameter

The magnetic properties of both single-crystal Li₂RuO₃(C) and Li₂RuO₃(P) are examined for 1.7< T < 900 K. Neither shows the singlet ground state observed in the polycrystalline Li₂RuO₃. Instead, Li₂RuO₃(C) displays paramagnetic behavior at T > 20 K with the magnetic susceptibility χ following the Curie-Weiss law for 20 K < T \leq 750 K (**Fig.3a**). Data fits to the Curie-Weiss law yield an effective moment μ_{eff} = 2.77 μ_{B} /Ru, consistent with that expected for an S=1 system, and a Curie-Weiss temperature θ_{CW} =-112 K. A signature for a long-range order near T_N=9K is evident in both χ (T) and C(T)(**Fig. 3b**).A largefrustration parameter, FP = $|\theta_{CW}|/T_N$ = 12.4 suggests the presence of significant frustration (**Table 2**). Indeed, the two unequal Ru-Ru bondsmay favor a formation of zigzag chains along the a-axis (see schematic in the inset of **Fig. 4**) as the

inter-chain interaction is weakdue to the long Ru-Ru bond L_l . Therefore, nomagnetic orderingoccursuntil below $T_N=9$ K when three-dimensional correlations are established.

For more distorted Li₂RuO₃ (P), a magnetically ordered state also takes placebut at a lower temperature, T_N =4 K (**Fig. 3c** and **3d**). Remarkably, the magnetic anisotropy is much stronger, and the magnitude of χ_{\perp} is significantly larger than that in Li₂RuO₃ (C), implying the importance of SOC. However, the temperature dependence of χ at high temperatures is much weaker than that for Li₂RuO₃ (C). The results suggest that Li₂RuO₃ (P) is "half-way" to dimerizationas the lattice is more similar to that of the polycrystalline sample; the magnetic state eventually prevails below T_N =4 K becauseLi₂RuO₃(P) is after all not as distorted as the polycrystallineLi₂RuO₃.

Computational Results OurLDA (local density approximation) calculations using the LMTO (linearized muffit-tin orbitals) method [35] andWannier function projection method [36] show that the crystal-field splitting in the Ru t_{2g} shell does not exceed 70 meV, indicating that the comparable SOC may play a significant role. However, the off-diagonal matrix elements of the Hamiltonian, hopping parameters, are even larger, ~ 200 meV, which is strong enough to form the quasi-molecular orbitals (QMOs) similar to those in Na₂IrO₃ where QMOsinvolve six Iratoms arranged in a hexagon and each Ir atom belongs to three differentQMOs, which dominate the formation of electronic structure[13]. The results of the optimization of the crystal structure performed in the GGA (generalized gradient approximation) calculations using the pseudopotential method [37] indicate that the nearly ideal honeycomb Na₂RuO₃ indeed corresponds to a minimum of the total energy for an AFM state. In addition, our GGA+U calculations show that a relatively small on-site Coulomb repulsion U ~ 1.5 eV is sufficient to

suppress the dimerization observed in polycrystalline Li₂RuO₃. The band structure of single-crystal Li₂RuO₃ strongly differs from that of both Na₂RuO₃ and Na₂IrO₃ on the LDA level (see SM Fig. 2[33]); and consequently, there is no sign of the QMOs.According to a recent study [30], when one of QMOs (of E2u symmetry) is half-filled, the correspondinginstability may induce the Jahn-Teller distortions (JTDs) that in turn lead to the dimerization. In less distorted single-crystal Li₂RuO₃, no sign of the JTDs is seen sincethe formation of the zigzag chains effectively removes the orbital degeneracy or JTDs. Therefore the zigzag chains constitute an alternative state to the dimerization when the JTDsare absent. However, both the zigzag chains and dimerized latticecost certain elastic energy that tends to stabilize uniform structure, and the prevailing state sensitively depends on details of the band structure and bulk modulus of the system (seeSM[33] for details).

Indeed, all relevant energies vigorously compete and critically bias their mutual competition to stabilize ground states. This explains that there exist nearly degenerate states in these materials, and the prevailing ground state critically depends on details of the structure, as illustrated in **Fig. 4**. The magnetic ordering systematically decreases with increasing (L₁-L_s)/L_sand eventually vanishes at a critical value where the dimerization emerges, leading to the singlet ground state observed in polycrystallineLi₂RuO₃. All results strongly indicate a direct correlation between the ground state and basal-plan distortions. The newly found Na₂RuO₃ provides a reference for almost perfect honeycomb symmetry.

The absence of the dimerization in single-crystal Li₂RuO₃ cannot be due to either impurity or quality of the single crystals. In fact, the singlet ground state is unusually

resilient to heavy impurity doping and is even enhanced by 5% Na doping (see Fig. 3 in SM [33]) and survives up to 50% Ir substitution for Ru in the polycrystalline samples [31]. It is likely that the difference between the two forms of Li₂RuO₃ arises from different synthesis conditions, as discussed above, which might cause different degrees of site-disorder in the honeycomb network due to the similar ionic radius of Li and Ru, and/or slightly different stoichiometry (e.g. oxygen content) (see SM [33]). Hence, this work does not rule out the possibility that single-crystal Li₂RuO₃ having the same structural distortions and singlet ground state as polycrystalline Li₂RuO₃ may eventually form under certain synthesis conditions.

Thework also offers the following general observations. Both Li₂RuO₃ and Li₂IrO₃ are more structurally distorted and behave with more complexities than their Na counterparts.SOC is expected to impose a J=0 state for Ru⁴⁺(4d⁴) ions (anda J_{eff}=1/2 state for Ir⁴⁺(5d⁵) ions) butthe observed magnetic states in the honeycomb ruthenates as in many other ruthenates [25] indicate that SOC is not sufficient to induce a J=0 state. It is intriguing thatall honeycomb ruthenates and iridatesmagnetically order in a similar temperature range (see SM-Fig. 4 [33])despite the different role of SOC in them.

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Captions

Fig. 1. Diffraction images in the (*h0l*) plane of the single crystal Li₂RuO₃with space group (**a**) P2₁/m and (**b**) C2/m. **Insets**: The corresponding honeycomb lattice and Ru-Ru bond distances. The temperature dependence of (**c**)the *a*-axis and (**d**)the ratio *b/a* from our single crystal P2₁/m phase (blue), C2/m phase (purple), powder samples (red star), and powder data from Ref.28 (black circles). Notethat the sharp diffraction pattern clearly indicates the high quality of the single-crystal Li₂RuO₃.

Fig. 2. Single-crystal Na_2RuO_3 :(a) The temperature dependence of the magnetic susceptibility for the basal plane $\chi_{\square}(T)$ and out-of-plane $\chi_{\perp}(T)$ for single-crystal Na_2RuO_3 ; Right scale: $1/\Delta\chi_{\square}$ where $\Delta\chi = \chi - \chi_{\square}$ and χ_{\square} is the temperature-independent contribution to χ . (b) The temperature dependence of the specific heat C(T) and $\chi_{\perp}(T)$ (right scale).

Fig. 3. Single-crystal Li_2RuO_3 (C): The temperature dependence of (**a**)the magnetic susceptibility $\chi_{\square}(T)$ and $\chi_{\perp}(T)$ and $1/\Delta\chi_{\perp}$ (right scale) for 1.7 < T < 850 K and(**b**) the specific heat C(T) and $\chi_{\square}(T)$ and $\chi_{\perp}(T)$ (right scale) at low T. Single-crystal Li_2RuO_3 (P): The temperature dependence of (**c**) $\chi_{\square}(T)$ and $\chi_{\perp}(T)$ and $1/\Delta\chi_{\perp}$ (Inset) and (**d**) $\chi_{\square}(T)$ and $\chi_{\perp}(T)$ and $\chi_{\perp}(T)$

Fig. 4. The Neèl temperature T_N as a function of the bond distance ratio $(L_l-L_s)/L_s$ for all honeycomb ruthenates. Inset: a schematic of the honeycomb lattice featuring L_l

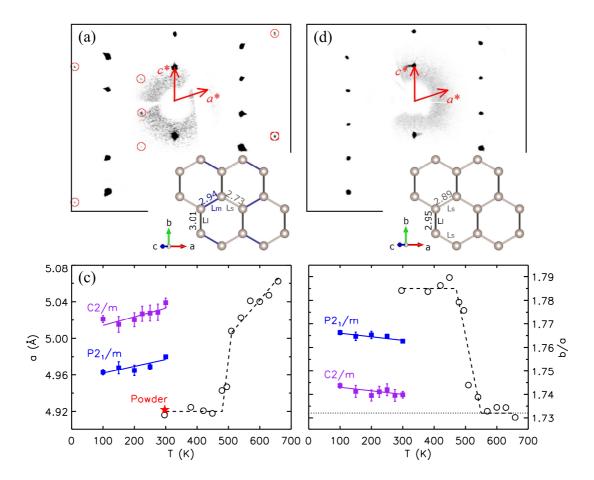


Fig. 1

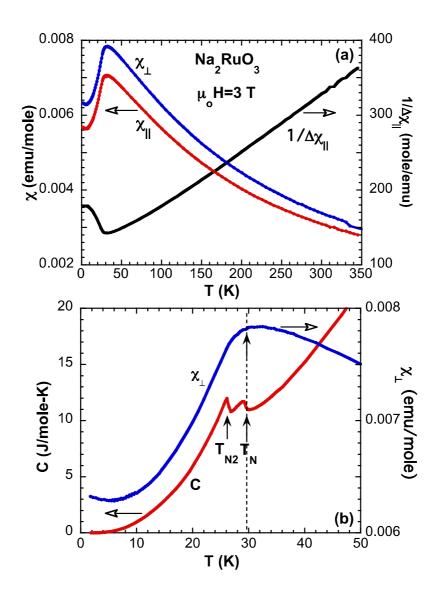


Fig. 2

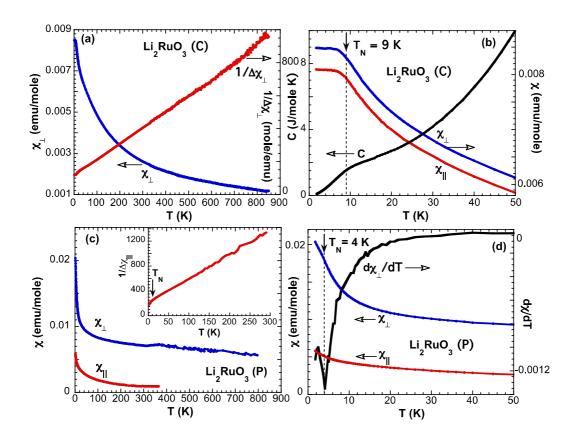


Fig.3

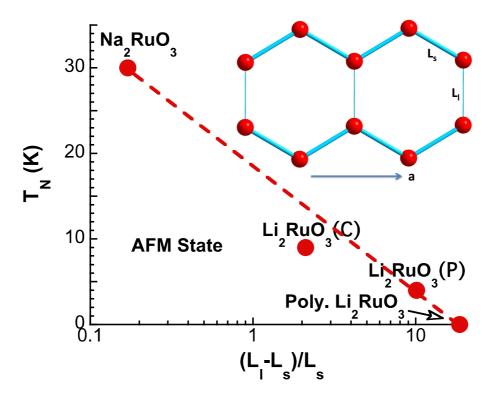


Fig. 4