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2D proximate quantum spin liquid state in atomic-thin α -RuCl₃

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Abstract

Two-dimensional (2D) atomic crystals have made major inroads into condensed-matter physics and give rise to fascinating phenomena due to quantum confinement. Here we report the first Raman scattering study on phonon-magnetic scattering coupling, proximate quantum spin liquid ground state and collective fractionalized excitations in exfoliated α -RuCl₃ atomic layers. Our results uncover that 2D α -RuCl₃ could harbour the unusual magnetic continuum, serving as a hallmark of the 2D proximate quantum spin liquid state and frustrated magnetic interactions. More importantly, our work demonstrates that the unusual magnetic scattering, as compared with bulk, is more obvious in 2D α -RuCl₃, indicating that the frustrated magnetic interactions are enhanced strongly. Such unusual enhancement of frustrated magnetic interactions may be responsible for the gigantic phonon-magnetic scattering coupling of 2D α -RuCl₃ and play a key role in stabilizing the 2D proximate quantum spin liquid state. Our work establishes a firm basis for exploring and understanding the 2D proximate quantum spin liquid and fractionalized excitations based on the atomically thin α -RuCl₃.

1. Introduction

Quantum spin liquid (QSL) is an exotic topologically ordered state of matter and a long sought goal in condensed-matter physics, where the atomic magnetic moments are long-range entangled but evade spontaneous breaking of translational or spin-rotational symmetry even at absolute zero temperature [1-7]. Such fascinating topological state was proposed by Anderson in 1973 and found to play a salient role in both the high-transition-temperature superconductors and topologically protected quantum bits [7-12]. Up to now, the QSL state has sparked significant attention and been demonstrated in a cornucopia of bulk single-crystal, such as $\text{ZnCu}_3(\text{OD})_6\text{Cl}_2$ [4], YbMgGaO_4 [3], A_2IrO_3 ($\text{A} = \text{Na}, \text{Li}$) [13] and $\alpha\text{-RuCl}_3$ [5, 6, 14-17]. However, the topological QSL state, to the best of our knowledge, has not been discovered in any 2D atomic crystals. Understanding the QSL state and fractionalized excitations in 2D limit is essential to the Kitaev model which is exactly solvable in 2D system [18] and would set a foundation for unraveling the mechanism of high-temperature superconductivity and numerous promising applications of quantum computation [7, 11].

According to the Mermin theorem [19, 20], reduced dimensionality would lead to the strong thermal fluctuations and may destroy the stabilities of 2D QSL, as in 2D ferromagnetism and antiferromagnetism [21-24]. On the other hand, being akin to the significantly enhanced electron-phonon coupling and charge-density-wave (CDW) order in monolayer limit [25-27], the frustrated magnetic interactions may be enhanced strongly in 2D atomic crystals and play a fundamental role in favouring the stronger QSL state. Due to the mutual competition between the above two factors, how is the 2D QSL? In spite of the significantly enhanced thermal fluctuations in low-dimensional systems, the breakthroughs of magnetism [22-24, 28], CDW [25] and Ising superconductivity [29-31] in 2D van der Waals crystals show us the possibility of 2D QSL.

$\alpha\text{-RuCl}_3$, a spin-orbit entangled $j_{\text{eff}} = 1/2$ Mott insulator on honeycomb lattice, is the celebrated candidate for proximate Kitaev QSL and exotic fractionalized excitations due to the strong spin-orbit coupling [5, 6, 17]. With the same lattice structure as 2D magnet CrI_3 [22], $\alpha\text{-RuCl}_3$ crystal is a layered material in which individual atomic layers with edge-sharing RuCl_6 octahedral (Fig. 1a) are stacked together by van der Waals interactions [6]. Such weak interlayer coupling makes it easy to extract 2D samples [32-34] and provides a perfect platform for investigating the 2D proximate QSL. In addition, recent theory shows that the proximate Kitaev QSL in $\alpha\text{-RuCl}_3$ is

stable against the interlayer coupling [35], suggesting that 2D α -RuCl₃ can possess the topological proximate QSL state. In fact, fractionalized Kitaev excitations and enhanced magnetic fluctuations have been uncovered recently in a 10 nm thick α -RuCl₃ nanoflake and atomically thin α -RuCl₃, respectively [32, 33]. These inspire us to explore the 2D proximate QSL state and fractionalized excitations in α -RuCl₃ atomic layers.

In this paper, we report the first experimental observation of the unusual magnetic continuum and 2D proximate QSL state in atomically thin α -RuCl₃ samples. As compared with bulk, the exotic magnetic scattering is more obvious and the frustrated magnetic interactions are enhanced strongly in 2D α -RuCl₃. Such unusual enhancement of frustrated magnetic interactions gives rise to the gigantic phonon-magnetic continuum coupling of 2D α -RuCl₃.

2. Results

Using a modified mechanical exfoliation method we developed previously [36], we obtained monolayer, bilayer and few-layer α -RuCl₃ samples on 300 nm SiO₂/Si substrates from the bulk crystals. Figure 1b shows the optical image of a typical exfoliated α -RuCl₃ sample with the number of layers overlaid. Atomically thin α -RuCl₃ flakes were first identified by their interference contrast through the optical microscope and then confirmed by atomic force microscopy (AFM). The height image obtained from a tapping mode AFM scanning of the square region in Fig. 1b (dotted black lines) is presented in Fig. 1c. Judging from the morphology of the freshly exfoliated samples at the nanometer scale, there is no evidence of structural irregularity or bubbles on the surfaces. This indicates the high quality of our samples and means that atomically thin α -RuCl₃ samples are relatively stable under atmospheric conditions. The height profiles taken along the blue and red lines in Fig. 1c are depicted in Fig. 1d. It is noted that the step height of an individual layer on α -RuCl₃ flake is ~ 0.6 nm, in fair agreement with the interlayer spacing of 5.73 Å [34]. While the height of monolayer (bilayer) α -RuCl₃ on SiO₂/Si substrate is about 0.83 nm (1.31 nm), larger than the theoretical thickness. The deviation implies that there are some absorbents at the interface between the α -RuCl₃ and SiO₂/Si substrate [33, 37].

Figure 2a presents the representative Raman spectra at room temperature for monolayer and few-layer α -RuCl₃ samples excited by 2.33 eV radiation in vacuum environments. The laser power on the sample during Raman measurement was kept below 100 μ W in order to avoid sample

damage and excessive heating. The silicon Raman mode at 520.7 cm^{-1} was used for calibration prior to measurements and as an internal frequency reference. Although different layers belong to distinct symmetries, for example, monolayer and bulk harbour D_{3d} and C_{2h} space group, respectively, we denote all phonons with the irreducible representations of the monolayer for simplicity [5, 15]. Based on the factor group analysis, the total irreducible representations for Raman active modes at the Γ point are given as follows [5]:

$$\Gamma = 4E_g + 2A_{1g} \quad (1)$$

In close analogy to bulk, five strong and sharp phonon modes are resolved at 117, 164, 270, 296 and 312 cm^{-1} for monolayer and few-layer samples. The mode at 312 cm^{-1} belongs to A_{1g} symmetry. The remaining four modes are assigned E_g symmetry. The absence of another A_{1g} mode is due to its weak intensity at room temperature [5].

As indicated by the dashed vertical lines in Fig. 2a, it can be seen that the energies of all phonons are independent on the number of layers. This is in marked contrast to transition metal dichalcogenides [27, 37, 38] and indicates that the van der Waals interlayer interactions in $\alpha\text{-RuCl}_3$ are extremely weak and have tiny effects on phonon energy. The weak interlayer coupling is also demonstrated by low-frequency Raman measurements, exhibiting neither interlayer shear mode nor breathing mode (Fig. S3, Supplemental Material). The extremely weak interlayer interactions lead to large thermal hysteresis and stacking faults causing a wealth of magnetic transition temperatures [15, 34, 39, 40]. In contrast to the phonon frequency, Raman intensity depends on the number of layers strongly. With increasing thickness, the phonon intensity increases first and then decreases, originating from the multilayer interference occurring for both the incident light and the emitted Raman radiation [27, 41, 42] (Fig. 2a and Fig. S4, S5 in Supplemental Material). For monolayer, the intensity is about half that of bulk. This goes against the recent results that the Raman intensity shows a nearly three order-of-magnitude decrease in monolayer limit [33]. The much higher intensity for our 2D $\alpha\text{-RuCl}_3$ samples signifies the high quality. We believe that the high quality of our 2D $\alpha\text{-RuCl}_3$ samples plays a key role in observing the topological 2D proximate QSL state, as will be discussed below.

Moreover, it can be seen clearly that the two lowest energy phonons show asymmetric Fano line shape, stemmed from the coupling between the discrete optical phonons and the magnetic scattering [5, 15, 43]. Here, we only focus on the optical mode 164 cm^{-1} with stronger intensity,

due to that the layer dependencies of phonon-magnetic scattering coupling for the two lowest energy phonons are the same [32]. For a quantitative analysis, the intensity of 164 cm^{-1} mode has been normalized and is fitted to a Fano profile [5, 15, 44]

$$I(\omega) = I_0 \frac{(q + \varepsilon)^2}{1 + \varepsilon^2} \quad (2)$$

where I_0 is the integrated intensity, $1/|q|$ is the asymmetry parameter, and a reduced energy ε is defined by the phonon frequency ω_0 and width Γ

$$\varepsilon = \frac{\omega - \omega_0}{\Gamma} \quad (3)$$

Figure 2b presents the normalized Raman spectra around 164 cm^{-1} for monolayer, bilayer, trilayer and bulk. It can be seen unequivocally that the 164 cm^{-1} optical mode shows Fano asymmetry and can be fitted well by the equation 2 (solid lines). The resulting layer-dependent Fano asymmetry ($1/|q|$) and linewidth (Γ) are summarized in Fig. 2c. Both Fano asymmetry parameter and linewidth increase exponentially with a descent of the number of layers. Fano asymmetry parameter $1/|q|$ shifts from 0.096 in the bulk to 0.21 in the monolayer with an increase of 130%, demonstrating the strongly enhanced phonon-magnetic scattering coupling in 2D limit. On the other hand, the full width at half maximum of high energy phonons, such as 270 cm^{-1} (Fig. S6, Supplemental Material), is thickness independent, indicating that the layer-dependent Γ for modes 164 cm^{-1} is indeed stemmed from phonon-magnetic scattering coupling, rather than extrinsic factors.

The sharp enhancement of phonon-magnetic scattering coupling in 2D $\alpha\text{-RuCl}_3$ may indicate the strongly enhanced frustrated magnetic interactions and thus play a crucial role in stabilizing and realizing the 2D proximate QSL. To confirm the topological proximate QSL state in 2D $\alpha\text{-RuCl}_3$, we performed the Raman measurements at 10 K to inspect the exotic magnetic continuum at low-energy, which serves as a hallmark for the proximate QSL state [4, 5, 13-15]. Figure 3a presents the Raman spectra for 2L, 3L, 7L and bulk, excited by 1.96 eV at 10 K. Strikingly, we can see a prominent difference between the Raman spectra at 10 K and room temperature. In addition to the five strong and sharp phonon modes, we can observe a magnetic continuum at 10 K for not only the bulk but also the 2D $\alpha\text{-RuCl}_3$ samples. The detail of the unusual magnetic scattering, guided by the shaded blue regions, is displayed in Fig. 3b and can extend up to roughly 200 cm^{-1} . The magnetic continuum in 2D $\alpha\text{-RuCl}_3$ could serve as the

hallmark for 2D proximate QSL state [5, 14, 15]. In close analogy to the breakthroughs in 2D magnetism and superconductivity [22, 23, 30, 31], our results demonstrate, for the first time, the topological 2D proximate QSL state in atomically thin α -RuCl₃. Moreover, it can be seen clearly that the magnetic continuum becomes more obvious and extends up to higher energy with a descent of layer thickness. Such enhancement of magnetic scattering in atomically thin α -RuCl₃ can be understood as a result of strongly enhanced frustrated magnetic interactions in 2D limit.

Having demystified the magnetic scattering and topological proximate QSL state in 2D α -RuCl₃, we focus on the temperature-driven evolution of magnetic continuum to inspect the layer dependent temperature that spin liquid correlations emerge. Figure 4a shows the Raman intensity at low energy for 2L, 3L, 7L and bulk. It can be known unequivocally that the low energy intensity, as compared with bulk, is enhanced strongly in 2D α -RuCl₃, indicating that the frustrated magnetic interactions become much stronger with a descent of the layer thickness. Figure 4b presents the evolution of spectral weight (SW) with temperature for distinct layers. The SW is defined as follows [5]:

$$SW = \int_{\omega_0}^{\omega_I} I(\omega) d\omega \quad (4)$$

where the cutoff ω_0 (ω_I) is 5 meV (12.5 meV). With decreasing temperature, the SW drops first, stemming from the thermal Bose factor [5]. When temperature is below 100 K, the SW gains the intensity again. The change for the SW provides a hallmark of the in-plane spin correlations and proximate QSL state [5, 15]. Moreover, it is prone to know that the temperature corresponding to the change of SW is about 100 K for both the bulk and 2D α -RuCl₃ (indicated by the dashed vertical lines in Fig. 4b). This indicates that the temperature at which the proximate QSL state appears is thickness independent and may be due to the mutual offset between thermal fluctuations and enhanced frustrated magnetic interactions. In addition, two new Raman peaks (as indicated by * in Fig. 3b and Fig. 4c) appear around 117 and 164 cm⁻¹ for 2D α -RuCl₃ samples below 100 K. The origin for these two new Raman peaks is not clear. Since the temperature corresponding to the emergence of these new modes and change of SW shows good consistency, one possibility is that these two new Raman peaks stem from the strongly enhanced frustrated magnetic interactions. Further in-depth quantitative studies and quantum scattering theories are needed to elaborate the microscopic mechanism.

As discussed already, atomically thin materials, as compared with bulk, possess strong thermal fluctuations and thus greater reactivity [19, 45, 46]. To confirm the stability of α -RuCl₃ in 2D limit, we measured the AFM image and Raman spectra of atomically thin α -RuCl₃ after a two months aging in atmosphere. The height image and 3D topography, obtained from the same square region with Fig. 1c, are presented in Fig. 5a and 5b, respectively. In stark contrast to freshly exfoliated α -RuCl₃ without structural irregularity or bubbles on the surfaces (Fig. 1c), a lot of structural irregularities appear on the surface of the monolayer and few-layer α -RuCl₃ samples after a two months aging (Fig. 5a and 5b), indicating that α -RuCl₃ samples have been worse. Figure 5c presents the representative normalized Raman spectra of monolayer and few-layer α -RuCl₃ samples after a two months aging (black lines). Strikingly, 1L – 4L α -RuCl₃ samples after a two months aging, as compared with freshly exfoliated samples (red lines), exhibit different Raman features, especially for the two lowest energy phonons. First, the scattering intensity below 120 cm⁻¹ shows a steplike increase in 1L – 4L α -RuCl₃ samples after a two months aging. Second, we cannot observe the 117 cm⁻¹ mode. This may stem from that the 117 cm⁻¹ mode disappears or is largely masked by the strong increase of the background. Third, the optical mode 164 cm⁻¹ becomes broadened and red-shifts to 152 cm⁻¹. Fourth, the intensity ratio between the lower energy mode around 160 cm⁻¹ and higher energy phonons about 300 cm⁻¹ decreases. As compare with recent result that shows the Raman spectra of atomically thin α -RuCl₃ for the first time [33], both our AFM image and Raman spectra of monolayer and bilayer α -RuCl₃ samples after a two months aging are akin to that in Ref. 33, indicating that the atomically thin α -RuCl₃ in Ref. 33 should have been worse. In other words, our results are the first experimental observation of the intrinsic Raman spectra and phonon-magnetic scattering coupling in monolayer and bilayer α -RuCl₃. Thus, it can be known that atomically thin α -RuCl₃ is not very stable in atmosphere. Our results provide a simple, nondestructive and fast characterization to determine whether or not the atomically thin α -RuCl₃ samples have been worse. We believe that such quality characterization would play a guiding role in future research.

3. Conclusion

In conclusion, monolayer, bilayer and few-layer α -RuCl₃ have been obtained by mechanical exfoliation method and characterized systematically using AFM and Raman. We demonstrate, for

the first time, the strongly enhanced frustrated magnetic interactions, unusual magnetic continuum and 2D proximate QSL state in atomically thin α -RuCl₃. The stability for the topological 2D proximate QSL can be understood as a result of significantly enhanced frustrated magnetic interactions. Moreover, the temperature corresponding to the change in SW and emergence of proximate QSL state is thickness independent, stemming from the mutual offset between thermal fluctuations and enhanced frustrated magnetic interactions. Our results set a foundation for a cornucopia of peculiar physical phenomena associated with the celebrated Kitaev topological 2D proximate QSL, gauge fields and fractional excitations and will motivate continuing major searches for realizations the quantum computation based on atomically thin α -RuCl₃.

Experimental Section

Monolayer, bilayer and few-layer α -RuCl₃ samples were mechanically exfoliated by scotch tape from a bulk crystal synthesized by vacuum sublimation of commercial RuCl₃ powder. Similar to the established processes for cleaving graphene, we used SiO₂/Si as the substrate and ordinary adhesive tape as the transfer medium. Prior to exfoliate atomically thin RuCl₃, the SiO₂/Si substrate was ultrasonically cleaned in acetone, 2-propanol, and deionized (DI) water, and then subjected to oxygen plasma to remove ambient adsorbates on its surface. Following the plasma cleaning step, the tape with RuCl₃ flakes was brought in contact with the substrate.

Raman spectra were acquired in vacuum environments using a micro-Raman spectrometer (Horiba LabRAM HR Evolution) in a confocal backscattering configuration (confocal pinhole of 200 μ m). Light from a 532-nm (room temperature) or 633-nm (low temperature) laser was focused down to a 2 μ m spot. The laser power on the sample during Raman measurement was kept below 100 μ W in order to avoid sample damage and excessive heating. The backscattered signal was collected by an Olympus 100 $\times$ objective (N.A. = 0.9) and dispersed by a 600-g/mm grating with Raman spectral resolution about 1.8 cm^{-1} . The low energy cut-off is 60 cm^{-1} and 5 cm^{-1} for 532-nm and 633-nm, respectively. The silicon Raman mode at 520.7 cm^{-1} was used for calibration prior to measurements and as an internal frequency reference.

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Figure 1. Microscopy and characterization of atomically thin α - RuCl_3 . (a) Top view of stick-and-ball hexagonal lattice structure of monolayer α - RuCl_3 . (b) White-light microscope images of a representative exfoliated sample with a $\times 50$ magnification. (c) Atomic force micrograph taken from the area indicated by dotted square in (b). (d) Height profiles taken along the red and blue lines in (c).

Figure 2. Raman characterizations using 532 nm laser line. (a) Raman spectra with various layer thicknesses. Traces are vertically offset for clarity. (b) Data and Fano fit (solid line) to the mode 164 cm^{-1} for monolayer, bilayer, trilayer and bulk. Inset is the schematic representation of eigenvector. (c) Fano asymmetry $1/|q|$ (blue) and linewidth Γ (magenta) versus the number of layers.

Figure 3. 2D magnetic continuum. (a) Raman spectra at 10 K for 2L, 3L, 7L and bulk, excited by 1.96 eV radiation. (b) Low-energy detail with corresponding to (a). The shaded blue region is a guide to the eye and indicates the magnetic scattering. The black dashed vertical lines indicate the new Raman peaks appeared in 2D α -RuCl₃.

Figure 4. (a) Raman intensity at low energy as a function of layer thickness. (b) Low-energy spectral weight versus temperature for 2L, 3L, 7L and bulk. (c) Temperature dependent Raman spectra for bilayer. Traces are vertically offset for clarity.

Figure 5. (a) Atomic force micrograph taken from the same area with Figure 1c after a two months aging in atmosphere. (b) 3D topography of (a). (c) Normalized Raman spectra with various layer thicknesses for freshly exfoliated α -RuCl₃ (red lines) and samples after a two months aging in atmosphere (black lines). Traces are vertically offset for clarity.