

Computational exploration of a viable route to the Kitaev-quantum spin liquid phase in monolayer OsCl_3

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In this computational study, we explore a viable route to access the Kitaev-quantum spin liquid (QSL) state in a recently synthesized monolayer of a so-called spin-orbit assisted Mott insulator OsCl_3 . In addition to other magnetic ground states in different regions, a substantial region of small J_{H}/U in our Hubbard U -Hund's J_{H} quantum phase diagram, obtained by combining second-order perturbation and pseudo-fermion functional renormalization group calculations, hosts Kitaev-QSL phase. Only the first neighbor Kitaev interaction of a smaller magnitude, nearly independent of the choice of U , is obtained in this region. Experimental tuning of the single parameter J_{H} to access the QSL state can purportedly be achieved with epitaxial strain in advanced crystal growth techniques. Furthermore, negligibly small farther neighbor interactions appear as a distinct feature of monolayer OsCl_3 , suggesting this material to be a better candidate for the exploration of the possible Kitaev-QSL state than previously proposed materials.

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I. INTRODUCTION

The Kitaev honeycomb model with bond-dependent Ising-type exchange interactions on a honeycomb lattice naturally hosts the novel quantum spin liquid (QSL) state [1]. The realization of the Kitaev model in so-called spin-orbit coupling (SOC) assisted Mott insulators was proposed by Jackeli and Khaliullin [2]. This requires SOC active transition-metal ions to be in an edge-shared octahedral crystal field (CF) of anions and Sr_2IrO_4 was initially proposed as an example [2]. Since then, many materials like α - RuCl_3 [3–7] and cobaltates [8–17] and other iridates [18–25] are proposed as pertinent Kitaev-QSL candidates [26,27]. In these materials, the formation of $j_{\text{eff}} = 1/2$ Kramers doublets, which form the low-energy space in the materials, results from the interplay of octahedral CF (Δ^{CF}) and SOC.

Although the dominant magnetic interaction between these pseudospin $1/2$ states is proposed to be Kitaev (K) type [2], other *undesirable* interactions such as isotropic Heisenberg (J) and off-diagonal terms (Γ), as well as farther neighbor couplings, also appear, causing long-range magnetic order in the materials mentioned above and driving them away from a possible QSL ground state. For example, the appearance of third nearest neighbor (3NN) J_3 [28] and strong neutron

absorbing Ir ions along with rapidly decaying magnetic form factor with an increase in momentum transfer in inelastic neutron scattering experiments on iridates are some of the experimental challenges [21,27]. In another example, smaller SOC strength of Ru^{+3} ions as well as a significant J_3 in α - RuCl_3 are the hurdles in the quest for a QSL state in this material. Despite such inevitable challenges, attempts have been made to access the QSL state using external knobs. For example, magnetic interactions in α - RuCl_3 were manipulated by means of the external magnetic field to achieve the QSL state [29–31]. In another theoretical attempt by Liu *et al.* [11], $\Delta_{\text{tri}}^{\text{CF}}$ was proposed as a tunable parameter with pressure to realize a QSL state in cobaltates. Towards this goal, our computational study in this article aims to explore the Kitaev-QSL state in SOC-assisted Mott insulators by tuning electronic parameters (U and J_{H}) which can be manipulated through solid-state effects in experiments. Our study is also motivated in part by recent theoretical work that argues for the experimental tuning of U and J_{H} parameters in perovskites with epitaxial strain, based on advancements in experimental epitaxial growth techniques [32].

To realize our goal, we consider the OsCl_3 monolayer as a case study. Our material choice is based on the fact it satisfies all the necessary criteria mentioned earlier to host strong Kitaev interactions, viz. $\text{Os}^{+3} 5d^5$ ions in the edge-shared OsCl_6 octahedron possess large SOC interaction. In a recent experimental study by Kataoka *et al.* [33], magnetization and heat capacity measurements on $\text{Os}_{0.81}\text{Cl}_3$ revealed the absence of any long-range magnetic ordering of Os^{+3} spins arranged on the triangular lattice (with local honeycomb domain formation) up to 0.08 K temperature, which can be an indication of underlying strong magnetic frustration in this compound,

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which is also a building block for Kitaev physics. A previous computational attempt to study the topological properties of this compound proposed a ferromagnetic (FM) ground state for the monolayer OsCl_3 [34], but a scrupulous investigation of its magnetic properties is warranted in the light of the fact that the isoelectronic $\alpha\text{-RuCl}_3$ has a zigzag (ZZ) antiferromagnetic (AFM) ground state. From a computational modeling perspective, electronic parameters U and J_H are part of the Hubbard-Kanamori H_{int} Hamiltonian in Eq. (1). The full model Hamiltonian $\mathcal{H}$ in Eq. (1) has been quite successful and hence widely accepted for the estimation of magnetic interactions in these materials [7,16,28,35,36]. Thus one can explore the access to the Kitaev-QSL phase by tuning the U and J_H parameters and calculating the corresponding magnetic interactions which is the objective of this article.

We first used *ab initio* calculations of higher accuracy to show that the ZZ and FM configurations in this material are nearly degenerate, indicating highly competing magnetic interactions that may be a reason for the absence of long-range magnetic ordering in this compound. We then varied U and J_H/U in the second-order perturbation calculations to obtain magnetic interactions and input them into pseudofermion functional renormalization-group (pf-FRG) calculations, which allowed us to obtain a $U\text{-}J_H/U$ quantum phase diagram. At small values of J_H/U , we found the emergence of only K -type 1NN interaction, albeit of smaller magnitude, resulting in the Kitaev-QSL state occupying a substantial part of the phase diagram which is nearly independent of the choice of U . Experimental tuning of a single parameter J_H with epitaxial strain in an advanced crystal growth techniques state may be a viable route to access the QSL state. In addition to the QSL state, we also find FM, ZZ, and Néel states present in different parts of the phase diagram. Much more advantageous than previously suggested Kitaev-QSL material candidates like iridates, $\alpha\text{-RuCl}_3$, and cobaltates, we find that the second and third neighbor magnetic interactions are negligibly smaller in the monolayer OsCl_3 , suggesting this material to be a better candidate for the exploration of a possible Kitaev-QSL state.

II. METHODS

In this study, we have used a combination of different computational methods, details of which can be found in the Supplemental Material (SM) [37]. Details of *ab initio* calculations, phonon dispersion, and magnetic anisotropy calculation can be found in Sec. S1 of the SM. Methods used to estimate various electronic parameters such as hopping amplitudes, crystal field splitting, and SOC strength can be found in Sec. S2. The second-order perturbation method used to estimate magnetic interactions in monolayer OsCl_3 is discussed in Sec. S4 of the SM [37]. Details of the pseudofermion functional renormalization group (pf-FRG) calculations used to obtain the quantum phase diagram presented in this paper is discussed in Sec. S5.

III. RESULTS

A. Crystal structure and magnetism—*ab initio* calculations

We consider the $C2/m$ honeycomb crystal structure of the monolayer OsCl_3 , which is similar to $\alpha\text{-RuCl}_3$ [34,38]. Very

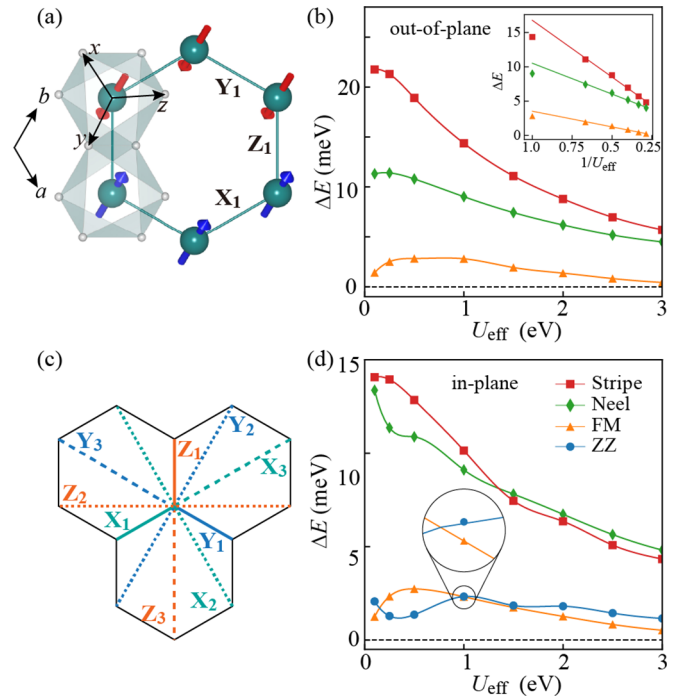


FIG. 1. (a) Top view of out-of-plane spin arrangement in ZZ magnetic state within monolayer OsCl_3 : spins are arranged out of plane on a hexagon towards the center of a Cl-Cl edge in an OsCl_6 octahedron. Large cyan and small gray balls represent Os and Cl atoms, respectively. Local axes x, y , and z along with crystallographic axes a and b are also shown. Variation in total energy differences (ΔE) for various magnetic configurations with U_{eff} on Os d states, using the out-of-plane ZZ state as the overall reference point for (a) out-of-plane spin alignments and (b) spins along the a axis. Clearly, the out-of-plane ZZ state has the lowest energy. The inset in (b) depicts the fitting of ΔE curves to a $1/U_{\text{eff}}$ function at large U_{eff} values. (c) Color-coded representation of X, Y, and Z bonds up to 3NN is shown.

recently, a triangular lattice with nanometer-sized honeycomb domains of Os atoms has been observed experimentally [33]. The structure is shown in Fig. 1(a) and Fig. S1 of the SM [37]. The full optimization of lattice constants within highly accurate *ab initio* calculations after imposition of both the ZZ and FM state brings nearly identical crystal structure in both cases. Our phonon calculations with this optimized crystal structure indicate that the honeycomb lattice of the OsCl_3 monolayer is at least dynamically stable. Details of *ab initio* calculations, optimized crystal structure, and phonon calculations can be found in Sec. S1 of the SM [37].

To understand the nature of magnetism and underlying frustration, highly accurate *ab initio* total energy calculations are performed, with tighter energy and force convergence criterion, after imposing various magnetic configurations like FM, ZZ, Néel, and Stripe. We varied U_{eff} on Os d states within the Dudarev [39] scheme in which U and J are on-site Coulomb and exchange parameters, with only the difference $U_{\text{eff}} = U - J$ being meaningful. Choosing range 0.1–3 eV for U_{eff} and the SOC effect at the self-consistent level, total energies are calculated for two spin orientations: out of plane [towards the center of one of the Cl-Cl edges in an OsCl_6

octahedron as depicted in Fig. 1(a)] and along a axis (in plane). We emphasize that while this particular out-of-plane spin orientation naturally arises during our self-consistent calculations, the orientation of spins along the a axis is preferred if the spins are allowed to rotate only in the ab plane (see Fig. S3 of the SM). The energy differences (ΔE) are calculated with the out-of-plane ZZ state as the overall reference point in both Figs. 1(b) and 1(d). The total energy plots for out-of-plane and in-plane spin orientations (spins are aligned along the a axis) are shown in Figs. 1(b) and 1(d), respectively. In Fig. 1(b), the ZZ state emerges as the ground state, with the closely competing FM state positioned higher in energy. Since both plots share the same reference point, i.e., the out-of-plane ZZ state, we can conclusively affirm that this magnetic configuration represents the overall lowest energy state. It is worth noting that a similar magnetic ground state was also proposed earlier for Na_2IrO_3 [40,41].

The energy difference between the ZZ and FM states nearly vanishes for the in-plane arrangement of spins along the a axis in Fig. 1(d), which is consistent with the previous study by Sheng *et al.* [34]. The FM and ZZ states on a honeycomb lattice differ by spin flip of one out of three nearest neighbors for any given lattice site. Hence their small energy difference, which has an entirely magnetic contribution, indicative of large magnetic frustration present in the system which might explain the absence of long-range magnetic ordering observed in experiment [33]. We find that the in-plane magnetic anisotropy for monolayer OsCl_3 is quite small (see Fig. S3 of the SM). Also, the large relative energies of other AFM configurations like Néel and stripe have a hint that the underlying exchange interactions in monolayer OsCl_3 may not be weak. In the most simplistic second-order perturbation theory, magnetic exchange interactions are $\propto t^2/U_{\text{eff}}$ at large U_{eff} values if one treats hopping t as a perturbation. To show the expected $1/U_{\text{eff}}$ behavior [42,43], we fit the ΔE curves in Fig. 1(b) to a $1/U_{\text{eff}}$ function at large U_{eff} values using linear regression. The fitting, shown in the inset of Fig. 1(b), yields excellent agreement with coefficient of determination (R^2) ~ 0.99 , confirming the expected $1/U_{\text{eff}}$ scaling of the magnetic energy differences. Next, we describe the electronic Hamiltonian considered in this study for exploration of the U - J_{H} quantum phase diagram in the quest of a QSL state.

B. Model Hamiltonian

The full Hamiltonian for d^5 systems is given as [7,16,28,35]

$$\mathcal{H} = H_{\text{hop}} + H_{\text{cf}} + H_{\text{soc}} + H_{\text{int}}, \quad (1)$$

where terms on the right-hand side are hopping, CF, SOC, and Hubbard-Kanamori Hamiltonians, respectively. For Mott insulators, in the limit $t \ll U$, one can separate the “on-site” term H_i^0 from $\mathcal{H}$. Such a limit is well justified in monolayer OsCl_3 . The largest value of t in Table S2 of the SM [37], obtained from a tight-binding (TB) model, is ~ 0.2 eV, while U is expected to be at least an order of magnitude larger [constrained random phase approximation (cRPA) estimates $U = 3.8$ eV]. H_i^0 , represented in basis $\psi_{i\sigma}^\dagger = [d_{z^2}^\dagger, d_{xz}^\dagger, d_{yz}^\dagger,$

$d_{x^2-y^2}^\dagger, d_{xy}^\dagger]_{i\sigma}$ for site i and spin σ , then reads as

$$\begin{aligned} H_i^0 &= H_{\text{cf}} + H_{\text{soc}} + H_{\text{int}} \\ &= \sum_{\sigma} \psi_{i\sigma}^\dagger \Delta_i^{\text{CF}} \psi_{i\sigma} + \sum \lambda \mathbf{L}_i \cdot \mathbf{s}_i \\ &\quad + \frac{U}{2} \sum_{\alpha} n_{i\alpha\sigma} n_{i\alpha\sigma'} + \frac{U'}{2} \sum_{\alpha \neq \beta} n_{i\alpha} n_{i\beta} \\ &\quad - \frac{J_{\text{H}}}{2} \sum_{\sigma, \sigma', \alpha \neq \beta} \psi_{i\alpha\sigma}^\dagger \psi_{i\alpha\sigma'} \psi_{i\beta\sigma'}^\dagger \psi_{i\beta\sigma} \\ &\quad - \frac{J'}{2} \sum_{\sigma \neq \sigma', \alpha \neq \beta} \psi_{i\alpha\sigma}^\dagger \psi_{i\beta\sigma'} \psi_{i\alpha\sigma'}^\dagger \psi_{i\beta\sigma}. \end{aligned} \quad (2)$$

In the above expression, U/U' are intraorbital/interorbital Coulomb interaction terms and J_{H} and J' are the Hund's coupling and pair hopping interaction, respectively. Rotational invariance in the single-atom limit dictates the relationships: $U' = U - 2J_{\text{H}}$ and $J_{\text{H}} = J'$. To bring complete rotational invariance of H_i^0 in the presence of e_g orbitals, the inclusion of additional three and four orbital terms in H_{int} should be considered. However, for large Δ_i^{CF} , such additional terms have an imperceptible influence on magnetic interactions. A discussion around this point is included in Sec. S4 of the SM [37]. Δ_i^{CF} and hopping amplitudes t_{ij} in $H_{\text{hop}} = \sum_{i \neq j, \sigma} \psi_{i\sigma}^\dagger t_{ij} \psi_{j\sigma}$ are obtained from a Wannier based tight-binding (TB) model excluding SOC effect. The strength of SOC in monolayer OsCl_3 , $\lambda = 0.355$ eV, is estimated by a band-structure fitting procedure (see Sec. S2 of the SM for details) [44]. As proposed earlier, it might be possible to experimentally tune parameters U and J_{H} by epitaxial strain in advanced growth techniques [32]. Hence we vary these two parameters to explore the possibility of a QSL state in monolayer OsCl_3 .

For a d^5 system, there are $C_5^{10} = 252$ combinations to arrange five electrons into ten d orbitals of $\psi_{i\sigma}^\dagger$ and is the dimension of the Hilbert spaces of H_i^0 . The eigenstates of this isolated Hamiltonian are obtained by exact diagonalization, the lowest two eigenstates of which are $j_{\text{eff}}=1/2$ Kramer's doublets. H_{hop} connects a two-site ground state (d^5, d^5) to an excited state with (d^4, d^6) configuration, the dimensions of whose Hilbert spaces are 210 each. We then use the second-order perturbation method, treating hopping as a perturbation, to estimate magnetic interactions between these pseudospin states. These interactions, obtained in the general form of 3×3 matrices, at X, Y , and Z bonds takes the form

$$\begin{aligned} \eta_X &= \begin{pmatrix} J + K & \Gamma' + \zeta & \Gamma' - \zeta \\ \Gamma' + \zeta & J + \xi & \Gamma \\ \Gamma' - \zeta & \Gamma & J - \xi \end{pmatrix}, \\ \eta_Y &= \begin{pmatrix} J + \xi & \Gamma' + \zeta & \Gamma \\ \Gamma' + \zeta & J + K & \Gamma' - \zeta \\ \Gamma & \Gamma' - \zeta & J - \xi \end{pmatrix}, \\ \eta_Z &= \begin{pmatrix} J + \xi & \Gamma & \Gamma' + \zeta \\ \Gamma & J - \xi & \Gamma' - \zeta \\ \Gamma' + \zeta & \Gamma' - \zeta & J + K \end{pmatrix}. \end{aligned} \quad (3)$$

Above, interactions J, K, Γ , and Γ' represent Heisenberg, Kitaev, off-diagonal terms, while ξ and ζ represent diagonal and

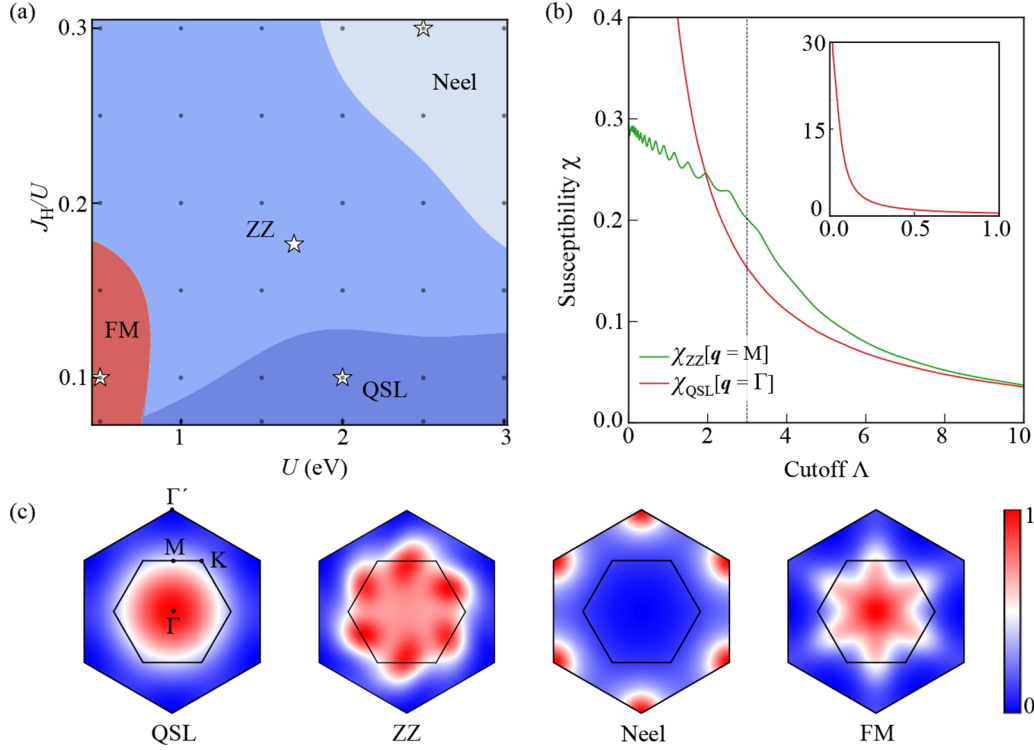


FIG. 2. (a) Magnetic quantum phase diagram in U - J_H/U parameter space obtained using a combination of second-order perturbation and pf-FRG calculations. The range considered is $U = 0.5$ – 3.0 eV and $J_H/U = 0.075$ – 0.3 are considered. Obtained magnetic phases are FM, ZZ, Néel, and QSL magnetic states. The gray dots are the calculated data points and the phase boundaries are obtained using the support vector machine (SVM) technique with a radial basis function kernel. Static spin correlation plots in (c) correspond to the points depicted by star symbols in (a). Intensity is normalized for each plot in (c) and the inner and outer hexagons in these plots denote the first and extended BZ of monolayer OsCl_3 and Γ , Γ' , M , and K points are in fractional coordinates of the reciprocal lattice. (b) The renormalization group flow of the magnetic correlations χ [$= \text{Tr}(\chi)$, where χ is a 3×3 tensor] for the two cases, ZZ (green curve) and QSL (red) states, with an inset showing the flow at low frequencies for the QSL state. Vertical dash lines show the break of the flow of χ for the ZZ state, while its smooth flow for the QSL state indicates no spontaneous symmetry breaking.

off-diagonal anisotropic terms. Only these symmetric components of interaction matrices appear on the first and third neighbor bonds, while on the second neighbor bonds, due to the absence of inversion symmetry, an additional antisymmetric component appears in the form of Dzyaloshinskii-Moriya interaction $\mathbf{D}_{ij}$ (DMI). The matrix form of DMI can be found in Ref. [28].

Details of the methodology and discussions on the validity of the low-energy $j_{\text{eff}}=1/2$ picture and atomic features, which can be observed in experiments for monolayer OsCl_3 , are presented in Secs. S3, S4, and S6 of the SM [37].

C. Magnetic interactions and quantum phase diagram

Estimated magnetic interactions up to 3NN [Fig. 1(c) depicts bonds up to 3NN] from the second-order perturbation method are used as input for the pf-FRG calculations. Interactions of magnitude $\leq |0.04|$ meV are rounded off to zero. This is justified in view of their unnoticeable influence on the magnetic ground state and helps the realization of the most relevant magnetic parameters for the system. We then analyzed the ground states by plotting the Fourier transformed static ($i\omega = 0$) magnetic susceptibility (two spin correlation function), obtained from pf-FRG calculations, in the first and

extended BZ of monolayer OsCl_3 , which is given by

$$\chi^{\mu\nu,\Lambda}(\mathbf{k}) = \frac{1}{N} \sum_{i,j} \chi_{ij}^{\mu\nu,\Lambda}(i\omega = 0) e^{i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)}. \quad (4)$$

Above, $\mathbf{r}_i$ denotes the position of the i th lattice site, N is the number of lattice sites, $\mu/\nu = x, y$, and z components of spins, and Λ is infrared cutoff (please see Sec. S5 of the SM [37] for details). The real-space magnetic susceptibility in the above equation is given by

$$\chi_{ij}^{\mu\nu,\Lambda}(i\omega) = \int_0^\beta e^{i\omega\tau} \langle T_\tau S_i^\mu(\tau) S_j^\nu(0) \rangle d\tau, \quad (5)$$

where τ is the imaginary time from functional integral construction. In the case of magnetic ordering, the ground state can be inferred from the dominant magnetic ordering vector $\mathbf{k}_{\text{max}}$ from peak susceptibility by $\chi^{\text{max}} = \max_{\mathbf{k}} \chi^\Lambda(\mathbf{k})$ in Eq. (4). A more detailed discussion can be found in Sec. S5 of the SM.

Our choice of the pf-FRG method as a quantum toolkit is based on its earlier demonstrated success in exploring quantum phase diagrams on different lattice and spin models [45–50]. Our obtained quantum phase diagram for a range of U (0.5–3.0 eV) and J_H/U (0.075–0.3) is shown in Fig. 2(a). This range of parameters is chosen so that the phase space is

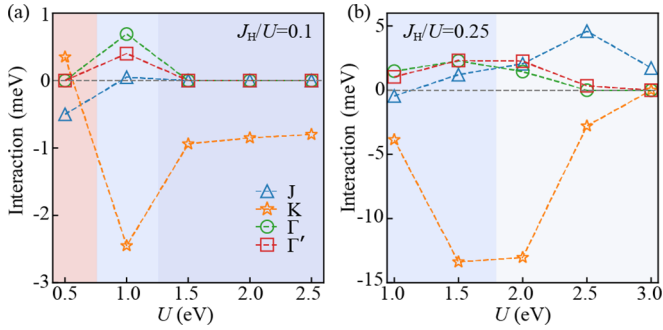


FIG. 3. Plot showing the variation of dominant magnetic interactions with U at (a) $J_H/U = 0.1$ and (b) at $J_H/U = 0.25$. Color-shaded regions from left to right in (a) are FM, ZZ, and QSL phases and in (b) are ZZ and Néel phases.

extended on either side of the points $U = 1.7$ eV and $J_H = 0.3$ eV ($J_H/U \sim 0.176$) previously considered for a $5d$ system such as iridates in the literature [28]. We use the support vector machine (SVM) with a radial basis function kernel to determine the phase boundaries from our numerical data. Taking advantage of a fairly dense $U - J_H/U$ grid, we employed the SVM classifier which naturally provides some smoothing of the phase boundaries while respecting the underlying data structure. The smooth boundaries should be understood as guides to the eye, representing the most likely transition regions between different phases based on our numerical data. Details of pf-FRG calculations are given in Sec. S5 of the SM. We will now discuss this phase diagram in detail.

Various magnetic phases, viz. FM, ZZ, and Néel, and QSL states occupy different parts of the phase diagram. At smaller values of $U = 0.5$ eV and $J_H/U = 0.075$ – 0.15 , there is a (< 1.5 meV) 1NN FM J , which is slightly larger than the AFM K coupling. For a representative point in this region [$U = 0.5$ eV and $J_H/U = 0.1$, marked by a white star in Fig. 2(a)], we obtain $J = -0.5$ meV and $K = 0.35$ meV, as illustrated in Fig. 3(a) for $J_H/U = 0.1$. This results in an FM ground state with a competing ZZ phase. This behavior manifested in a starlike shape of the static spin susceptibility (hereafter only susceptibility) plot in Fig. 2(c) (last plot). Consistent with the FM phase, the major contribution to the susceptibility comes from the zone center, while small spreads along M points in the first BZ indicate a competing ZZ AFM phase. Transition to later happens when both couplings change their respective signs with FM $K >$ AFM J as shown in the middle region of Fig. 3(a). This is consistent with a recent observation on cobaltates [16].

In the ZZ region, away from the FM-ZZ phase boundary, other terms like Γ , Γ' , ξ , ζ as well as smaller farther neighbor interactions start to emerge in the ZZ region of the phase diagram. This is shown in Fig. 3(b). However, 1NN dominant K and J terms remain FM and AFM, respectively. We emphasize here that 2NN and 3NN interactions are at least 2–3 orders of magnitude smaller than 1NN and have no qualitative effect on the magnetic ground state. Values of $U = 1.7$ eV and $J_H = 0.3$ eV bring ZZ state [white star in ZZ region in Fig. 2(a)] and estimated interactions are listed in Table I. The corresponding susceptibility plot in Fig. 2(c) (second from the left) has dominant contributions from M points of the

TABLE I. Estimated bond-dependent magnetic interactions for monolayer OsCl_3 using the second-order perturbation method at $U = 1.7$ eV, $J_H = 0.3$ eV, and $\lambda = 0.355$ eV. Values of Heisenberg J , Kitaev K , off-diagonal Γ , Γ' , and asymmetric DMI $\mathbf{D}$ terms along with diagonal and diagonal, off-diagonal anisotropic terms ξ and ζ are listed. Interactions of magnitude < 0.04 meV are rounded off to zero which include some 2NN as well as 3NN interactions.

Bond	Magnetic interactions (meV)					
	J	K	Γ	Γ'	ξ	ζ
X_1	-1.055	-14.600	5.662	-3.578	0.520	2.515
Y_1	-0.961	-17.215	6.010	-0.748	-0.213	-4.320
Z_1	-0.844	-15.127	4.749	-2.342	-1.285	-4.567
X_2	-0.294	0.533	0.206	-0.197		-0.082
Y_2	0.145	0.878	0.050	-0.150		-0.167
Z_2	-0.258	0.453	0.086	-0.246		-0.050
$\mathbf{D}_{ij}$						
X_2		(0,	0,	0)		
Y_2		(-0.350,	0,	-0.216)		
Z_2		(-0.108,	-0.125,	0)		

first BZ, consistent with the underlying ZZ magnetic order. In the absence of inversion symmetry on the 2NN bonds, a small DMI appears for these magnetic neighbors. We would like to mention that the value of U and J_H obtained from cRPA calculations also brings a ZZ state, details of which are provided in Sec. S7 of the SM. ZZ phase occupying the largest part of the phase diagram quantitatively differs in different regions and a related discussion is provided in Sec. S7 of the SM. Further increase of U (≈ 2 – 3 eV) and J_H/U (≈ 0.2 – 0.3) stabilizes the Néel AFM state, with 1NN AFM J being the dominant interaction, while all other interactions are highly suppressed. This is shown in Fig. 3(b). For a typical phase point $U = 2.5$ eV, $J_H/U = 0.3$ (marked by a white star in the Néel region), 1NN, 2NN, and 3NN J values are 3.39, 0.39, and 0.03 meV, respectively. Contribution from Γ' points of the extended BZ in the corresponding susceptibility plot shown in Fig. 2(c) (third from the left) is consistent with the underlying Néel magnetic order.

Most importantly, almost independent of the value of U in the range 1.0–3.0 eV, a substantial region of the phase diagram for $J_H/U = 0.075$ – 0.1 is occupied by the Kitaev-QSL state. In this region, we only obtained a small but finite 1NN FM K term (~ 1 meV) in our second-order perturbation calculations. The susceptibility plot for this state shown in Fig. 2(c) (leftmost) has a contribution from the whole first BZ, with the highest intensity appearing at the Γ point. No long-range magnetic ordering and hence no spontaneous symmetry breaking is expected for this state even at low temperatures. To verify this, we can plot the renormalization group flow of the magnetic correlations (χ) as a function of the renormalization group cutoff (Λ). Spontaneous symmetry breaking by the onset of a long-range magnetic ordering at a Λ results in a kink or break of χ . The smooth evolution of χ down to the lowest Λ can be seen for the QSL state from the red curve in Fig. 2(b) (see also the inset). For comparison, we also show a similar plot for the ZZ order (green curve) corresponding to

interactions listed in Table I. Clearly, χ , in this case, displays a breakdown at $\Lambda_c \approx 3.3$ signaling the onset of the ZZ order.

IV. DISCUSSION

Several remarks are in order. First, though off-diagonal magnetic coupling (e.g., Γ) is large compared to iridates [28], orders of magnitude smaller farther neighbor magnetic interactions (3NN interactions are all $<|0.04|$) distinguishes monolayer OsCl_3 these earlier proposed Kitaev-QSL candidates including iridates [16,28]. Second, strain as an external knob, proposed in Ref. [32], can be used to tune a single electronic parameter J_H since the QSL state is nearly independent of the choice of U . Similarly, strain can also be utilized to tune trigonal distortions as suggested in Ref. [11]. The strain in this case can also alter the hopping amplitudes between d orbitals. Hence it would be interesting to probe the role of strain in tuning the magnetic properties of these QSL candidate materials. Third, the required tuning of J_H to access the QSL region in the phase diagram can also be achieved through the choice of suitable elements from the Periodic Table. Advancement in material synthesis techniques can be of great help here.

V. CONCLUSION

In conclusion, considering recently synthesized monolayer OsCl_3 as an example, we have explored a route to access the Kitaev QSL state by varying electronic parameters Hubbard

U and Hund's J_H . These two parameters purportedly can be tuned experimentally with epitaxial strain. Using highly accurate *ab initio* total energy calculations, we have shown that the ZZ and FM are nearly degenerate indicating the large magnetic frustration which might be the possible reason behind the absence of long-range magnetic ordering in this material. Combining second-order perturbation and pf-FRG calculations, we presented a magnetic quantum phase diagram in U - J_H/U space. A substantial part of this phase diagram hosts the Kitaev QSL state, stabilized by 1NN Kitaev interaction only, with other magnetic phases like FM, ZZ, and Néel AFM states also appearing in different regions of the phase diagram stabilized by changing dominant magnetic interactions. Much to experimental advantage, the independence of the Kitaev-QSL region on U in our phase diagram, in turn, left only J_H for tuning with external knobs. Insights obtained from our study may be helpful in designing experiments on pertinent Kitaev-QSL candidate materials.

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Q.G. and S.K.P. contributed equally to this work.

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