Three dimensional quantum spin liquid in 3D-honeycomb iridate models and phase diagram in an infinite-D approximation

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Motivated by the recent synthesis of insulating Li$_2$IrO$_3$, where Ir$^{4+}$ $S_{Ir} = 1/2$ moments form a 3D lattice termed the H-1 harmonic honeycomb structure, we study magnetic Hamiltonians on this and other threefold co-ordinated lattices including the hyperhoneycomb lattice. Experimentally measured magnetic susceptibilities suggest that Kitaev interactions, predicted for the ideal 90° Ir-O-Ir bonds, are sizable in these materials. We first consider pure Kitaev interactions, which leads to an exactly soluble 3D quantum spin liquid (QSL) with emergent Majorana fermions and Z$_2$ flux loops. Unlike 2D QSLs, the 3D QSL is stable to finite temperature, with $T_c \approx K/100$. On including Heisenberg couplings, exact solubility is lost. However, by noting that the shortest closed loop $\ell$ is relatively large in these structures, we construct an $\ell \rightarrow \infty$ approximation by defining the model on the Bethe lattice. The phase diagram of the Kitaev-Heisenberg model on this lattice is obtained directly in the thermodynamic limit, using tensor product states and the infinite system time-evolving-block-decimation (iTEBD) algorithm. Both magnetically ordered and gapped QSL phases are found, the latter being identified by an entanglement fingerprint.

I. INTRODUCTION

Recently there has been growing interest in studying quantum phases of matter that are characterized by long range entanglemen$,^2$ in contrast to conventional symmetry broken states. In particular, gapped quantum phases that feature long range entanglement exhibit remarkable emergent properties such as excitations with unusual statistics and fractional quantum numbers. These properties are known to occur in two dimensional phases such as the fractional quantum Hall states, which are realized in 2D electron gases in strong magnetic fields. In solids, frustrated insulating magnets are believed to be prime candidates for avoiding conventional ordering in favor of a long range entangled phase of matter – the quantum spin liquid phase. Recently there has been mounting evidence for gapped spin liquids, phases which are long range entangled$^3$, on two dimensional geometrically frustrated lattices such as the Kagome lattice$^4$. However, frustration need not arise from geometry alone. In quantum magnets of heavy elements, spin-orbit coupling leads to anisotropic interactions that may engender quantum disordered ground states even in the absence of the usual geometrical frustration. A prime example is the honeycomb lattice – a bipartite lattice on which both ferromagnetic and antiferromagnetic Heisenberg couplings host ordered ground states. However, a peculiar set of anisotropic interactions proposed by Kitaev$^5$, where neighboring spins are coupled by Ising interactions along an axis that is set by the spatial orientation of the bond, has been shown to be in a quantum spin liquid phase. Furthermore, this is demonstrated via an exact solution – in contrast to the numerical tour de force required for identifying the spin liquid phase in the Kagome antiferromagnet$^6$. Interestingly, the requirement for obtaining an exactly soluble spin liquid is not specific to the honeycomb lattice. Instead, the key ingredients are the three fold coordination of the sites and the peculiar Ising interaction with rotating axes. If such a network would be created in three dimensions, it would lead to an example of a 3D quantum spin liquid. Such long range entangled quantum phases in 3D are less well explored than their 2D counterparts. While basic constraints on long range entangled quantum phases in 3D have been discussed$^7$, few suggestions for materials candidates exist. An exception is the 3D hyperkagome material$^8$, for which a spin liquid ground state with bosonic$^9$ or fermionic$^{10}$ spinon excitations has been proposed. Related U(1) spin liquids$^{11,12}$ have been proposed for quantum spin ice materials$^{13}$ on the pyrochlore lattice. Here we discuss a 3D example of quantum spin liquid behavior.

![FIG. 1. The H-1 lattice of iridium ions in Li$_2$IrO$_3$. The recently synthesized H-1 harmonic honeycomb lattice$^{14}$ space group #66 Cccm, has an eight-site unit cell with a base-centered orthorhombic Bravais lattice. Sites are threefold co-ordinated and form both 6-site hexagons and longer 14-site loops; the H-1 lattice is distinct from the hyperhoneycomb (H-0) lattice of Fig. 5. In the limit of superexchange via ideal oxygen octahedra, the magnetic Hamiltonian is dominated by Kitaev-type couplings, leading to an exactly soluble model of a 3D quantum spin liquid.](image-url)
induced by spin-orbit coupling in a 3D model with Kitaev exchanges, and explore a possible physical realization.

At first sight, the Kitaev interactions seem rather unphysical. However, as pointed out by Jackeli and Khaliullin they may actually be realized under certain circumstances in iridium oxides. An Ir$^{4+}$ ion at the center of an oxygen octahedron is expected to be in a Kramers doublet state $J = 1/2$, with the doublet wave function set by the spin-orbit coupling. This leads to unusual magnetic exchange interactions. For example, when a pair of Ir$^{4+}$ moments are coupled via an intermediate oxygen with a 90° bond, the usual Goodenough-Kanamori-Anderson rules would have predicted a ferromagnetic Heisenberg exchange. However, due to the special nature of the Kramers doublets, the coupling was shown[22] to be ferromagnetic, but of the Ising type, with the spin component involved being perpendicular to the bond's iridium-oxygen plane. Other exchange paths around the Ir-O-Ir-O square and involving higher energy states including the Ir$^{4+}$ $e_g$ orbitals[23][24] also generate this type of coupling, with either sign. For the compound Na$_2$IrO$_3$ in which Ir forms independent honeycomb lattices, these mechanisms were argued to lead to couplings identical to Kitaev's honeycomb model, although additional spin interaction, minimally a Heisenberg term, is also expected. An appropriate minimal model for the low energy magnetic Hamiltonian is then the nearest neighbor Kitaev-Heisenberg mode[23].

In the $C2/m$ layered structure[25] of Na$_2$IrO$_3$, and even more dramatically in the Cccm 3D-Li$_2$IrO$_3$ structure we discuss below, space group symmetries single out the subset of Ir-Ir bonds which are oriented along a particular axis. Recent ab initio work[26] has found that already for Na$_2$IrO$_3$, the magnitude of both Kitaev and Heisenberg couplings can be quite different between these symmetry-distinguished subsets of bonds. Allowing the couplings to take a different value on the symmetry-distinguished “c-bonds” compared to the remaining “d-bonds” produces the bond-anisotropic Kitaev-Heisenberg Hamiltonian,

$$H = \sum_{c\text{-bond}(ij)} \left( K_c S_i^{\gamma(i)} S_j^{\gamma(i)} + J_c \vec{S}_i \cdot \vec{S}_j \right) + \sum_{d\text{-bond}(ij)} \left( K_d S_i^{\gamma(i)} S_j^{\gamma(i)} + J_d \vec{S}_i \cdot \vec{S}_j \right).$$

The geometry of IrO$_6$ octahedra implies that the spin component $\gamma_{ij}$ coupled in a Kitaev term $K_S^{\gamma(i)} S_j^{\gamma(i)}$ is, on any bond, one of the Ir-O Cartesian axes $x$, $y$ or $z$.

The additional Heisenberg interactions are important; indeed, the ground state of Na$_2$IrO$_3$ is magnetically ordered and not a quantum spin liquid. The “zigzag” (wavevector $M$) magnetic ordering seen[27,28] in Na$_2$IrO$_3$, as well as other measured magnetic and electronic properties, are consistent with Kitaev-Heisenberg as well as more conventional Hamiltonians with SU(2) rotation symmetry[29–31]. Recent resonant inelastic x-ray scattering measurements exhibit flat disper-
sion features particularly reminiscent of strong Kitaev exchange. Since its original formulation and solution, much research has been done on elucidating properties of the Kitaev-Heisenberg model. As a model containing a quantum spin liquid, it has been especially interesting to investigate its behavior under charge doping. Note that other starting scenarios for Na$_2$IrO$_3$ have also been proposed, which paint a picture of it different from a Mott insulator.

In addition to the original proposals by Jackeli and Khaliullin, there have been many related proposals for anisotropic exchange in Na$_2$IrO$_3$ and related iridium compounds. The key point in this context is that the exchanges as described for Na$_2$IrO$_3$ apply equally well for any lattice of edge-sharing IrO$_6$ octahedra with roughly cubic local symmetry, as long as any distortion from cubic symmetry is weaker than the spin orbit coupling.

Recently, single crystals of insulating Li$_2$IrO$_3$ were successfully synthesized in a novel polytype crystal structure of edge-sharing IrO$_6$ octahedra, in which iridium ions form the $H$-1 harmonic honeycomb 3D lattice shown in Fig. 1. The lattice is locally honeycomb-like, consisting of strips of hexagon plaquettes and preserving threefold connectivity for every site. The lattice geometry suggests an extension to a harmonic honeycomb structural series, parametrized by an integer $n$ denoting the number of adjacent hexagon strips. The synthesized Li$_2$IrO$_3$ $H$-1 lattice is the $n = 1$ member of the series; the hyperhoneycomb lattice of Fig. 5 is the $n = 0$ member ($H$-0); and the layered honeycomb structure is described by $n = \infty$.

The 3D-Li$_2$IrO$_3$ crystals undergo a magnetic transition at 38K, as evidenced by large anisotropic peaks in magnetic susceptibility. The low temperature magnetic ordering is not currently known. As also pointed out in the experimental analysis, the bond-anisotropic Kitaev-Heisenberg model Eq. 1 is sufficient for capturing the large susceptibility anisotropy observed in experiment; within this scenario, large ferromagnetic Kitaev exchanges are necessary to fit the experimental data. A susceptibility fit is shown in Fig. 3 we elaborate on the magnetic couplings required for this fit in section IIIIC below. We study the Hamiltonian Eq. 1 with parameters extracted from the fit, classically as well as using the fully quantum large-$\ell$ approximation discussed below, and find in both cases a ground state with Stripy-X magnetic order. This collinear antiferromagnetic order, with a magnetic unit cell twice as large as the crystal unit cell, is schematically shown in Fig. 4. We predict that such a magnetic pattern may qualitatively capture the low temperature phase of 3D-Li$_2$IrO$_3$.

In parallel with this work, a few other studies of 3D Kitaev-Heisenberg models have appeared. Various properties of the hyperhoneycomb lattice model’s magnetic phases and exact spin liquids were studied while the magnetic phases at finite fields and temperature were explored using classical and semi-classical techniques.

**II. SUMMARY OF RESULTS**

We begin by analyzing the relevance of the Kitaev interactions to the $H$-1 lattice 3D-Li$_2$IrO$_3$ material. We discuss the interplay of chemistry and geometry in the A$_2$IrO$_3$ structures, aiming to understand the newly synthesized $H$-1 lattice and possibilities for other 3D harmonic honeycomb lattices of edge-sharing IrO$_6$ octahedra. We analyze in detail the argument, based on fitting magnetic susceptibility, that the magnetic properties of 3D-Li$_2$IrO$_3$ should be described by the bond-anisotropic Kitaev-Heisenberg model Eq. 1. Its key is the geometrical contrast between the $\hat{c} \sim x - \hat{y}$ axis distinguished by the $Ccmm$ space group symmetry, and the $z$-axis highly anisotropic susceptibility observed in experiment; the $S^z S^z$ Kitaev coupling of $\hat{c}$-bonds can replicate this rela-
The H-0 (hyperhoneycomb) lattice. Left: The unit cell has four sites, with a face centered orthorhombic Bravais lattice; the space group is #70, $Fddd$. Observe the threefold connectivity and the 10-site decagon loops, inspiring a large-$\ell$ loop length approximation. Right: Dark orange bonds of type $z$ (top right) or $x, y$ (bottom right) carry $u_{i,j} = -1$ within the QSL $Z_3$ gauge sector, producing a $Z_2$ closed flux loop excitation which encircles the bonds.

Accepting Eq. 1 as a minimal Hamiltonian, we proceed to study its spin liquid phase in the Kitaev limit through the Majorana fermion exact solution. We extend the previous analysis of a 3D Kitaev model, which studied an H-0-like graph,[20] and focus on the Li$_2$IrO$_3$ H-1 lattice. We compute the spin-spin correlators (or bond energies), finding variations of a few percent across dimensionality, geometry and symmetry-distinguished bonds. Computing the majorana spectrum with its nodal contour as a function of lattice geometry and bond anisotropy, we find the same phase boundary between the gapped and gapless spin liquids as a function of bond-strength anisotropy, independent of lattice, dimensionality (2D, 3D or infinite-D) and independent of whether the bond-anisotropy preserves or breaks the lattice symmetries.

We give a simple but general counting argument based on Euler’s formula that explicitly illustrates the lack of monopoles in (3+1)D $Z_2$ gauge theory, showing that closed flux loops rather than individual fluxes are the gauge-invariant objects. The energy of the flux loop excitations is described by a loop tension, which we compute within the zero temperature exact solution as $\tau = 0.011 |K|$. This tension combines with the extended nature of the loops to control the finite temperature behavior of the model, producing the finite temperature loop proliferation transition which confines the Majorana fermions. Together with the robustness of fermionic statistics (when 2D flux attachment is impossible), this stability to finite temperature hallmarks the features unique to three dimensional fractionalization.

Computing the quantum phase diagram of the full frustrated Hamiltonian is exponentially difficult; while such problems have been tackled in two dimensions, an unbiased phase diagram computation of the three dimensional model is currently impossible. We are able to capture it by employing a limit inspired by the H-0 hyperhoneycomb lattice, whose shortest loops are $\ell = 10$ decagons. Treating $\ell$ as a large control parameter and taking it to infinity, we reach the loopless Bethe tree lattice, which is infinite dimensional but preserves the key $z = 3$ connectivity. This $\ell \to \infty$ approximation is not analytically tractable, but does admit an entanglement-based numerical solution using tensor product states (TPS). Gapped states can be efficiently represented as a TPS on a tree lattice; on the tree, as in 1D systems, the full entanglement between two halves of the system is carried by the single bond connecting them. We employ the time evolving block decimation algorithm which works directly in the thermodynamic limit (iTEBD) [21] which has been previously extended to the Bethe lattice for magnetic phases[22] and other non-fractionalized phases.[23,24] The iTEBD straightforwardly captures the FM and Neel magnetic orders as well as their duals[21,22] the stripy and zigzag magnetic orders. The large-$\ell$ phase diagrams (section VI) are summarized in Fig. 2.

However, quantum spin liquids are generally difficult to identify positively since they lack an order parameter. Studies in 2D have relied on the topological entanglement entropy[25,26] but it cannot be extracted from bipartitions of the tree lattice. To complement the TPS computation we investigate the gapped Kitaev QSLs on the loopless tree using the Majorana exact solution, computing the entanglement entropy from the fermion and gauge sectors on each bond as a function of anisotropy. We find that the entanglement from the $Z_2$ gauge theory sector is partially quenched, on the weak-anisotropy bonds, due to the entanglement cutoff of the TPS computation. The remaining entanglement serves as a fingerprint which, alongside the negative-signature of vanishing magnetic order, we use to identify the QSL phase within the iTEBD computation. Ours is the first positive-signature capture of a fractionalized phase in the quantum large-$\ell$ limit.

This solution of the QSLs with their adjacent phases in the quantum large-$\ell$ approximation augments the ground state and finite temperature analysis within the solvable H-1 lattice three dimensional QSL, yielding a remarkably complete picture of a fractionalized phase in a potentially realizable solid state system.

III. RELEVANCE OF KITAEV INTERACTIONS TO THE H-1 LATTICE LITHIUM IRIDATE

A. Chemical bonding with IrO$_6$ octahedra

Oxides with octahedrally coordinated transition metals can bond in a variety of ways, sharing octahedral corners, edges, faces or a combination of these. For the iridates, edge sharing octahedral bonds are generally 25%
smaller than corner sharing ones, measuring $\sim 3\,\text{Å}$ (for edge sharing) compared to $\sim 4\,\text{Å}$ (for corner sharing) between nearest neighbors. In the latter if one oxygen is shared by exactly two iridium then four-fold symmetric structures generally arise, as in perovskite and layered perovskite structures. Compounds with edge sharing octahedra also occur in a variety of structures; the triangular lattice NaCoO$_2$, the hyperkagome Na$_3$Ir$_3$O$_8$ and the layered honeycomb Na$_2$IrO$_3$ are all examples.

In the layered honeycomb iridate, two oxygens are shared by exactly two iridium, and every iridium is coordinated by three others, belonging to a single plane. In a fixed coordinate system, there are multiple choices for the orientation of the triangular Ir-Ir-Ir plaquette, which are locally indistinguishable from the perspective of any given iridium atom. In general the octahedral symmetry will not be perfect and the distortion may favor the situation of the layered honeycomb where all of the Ir-Ir bonds lie within a common plane.

However, for sufficiently high local symmetry approaching the full $O_h$ group, alternatives to the layered geometry become increasingly favorable. Consider compounds with chemical formula Li$_2$IrO$_3$; this substitution of Na by Li is known to lead to much smaller distortions, since the Ir and Li ions are more similar in size. With the decreased octahedral distortion, multiple spatial orientations of the bonds should be more likely to occur. This can result in complex structures, such as the $\mathcal{H}$-1 lattice of 3D-Li$_2$IrO$_3$.

\section*{B. Symmetry and geometry of the harmonic honeycomb lattices}

The class of lattices we will consider, the harmonic honeycomb structures, possess bonds with various orientations comprising all but one of the possible orientations for edge-sharing octahedra. (Coordinates for $\mathcal{H}$-1 and $\mathcal{H}$-0 are given in Appendix~[E]) This scenario is shown in Fig.~[6] two opposite octahedra edges are forbidden from bonding, and distinguish the spatial direction parallel to these edges. The other edges on the same square create Ir-Ir bonds lying along the special $c$ axis, resulting in the bond anisotropy described above. The $c$ axis is thus distinguished for all harmonic honeycomb lattices; this is also reflected in the symmetry properties of each particular lattice. For example in the $\mathcal{H}$-1 lattice, the space group $Cccm$ has a single mirror plane, whose normal is the $c$ direction.

Recall that Kitaev spin coupling along the Bloch sphere cubic axis $\gamma \in \{x,y,z\}$ set by the spatial cubic vector normal to that Ir-Ir bond and the shared edge, shown here by the appropriate color (red, green, blue) of the shared edge. The 3D harmonic honeycomb lattices host a preferred axis for Ir-Ir bonds (thick blue bond); the perpendicular edge (X’ed out gray lines) is not shared by any two octahedra.

\section*{C. Capturing 3D-Li$_2$IrO$_3$ susceptibility with a bond-anisotropic Kitaev-Heisenberg Hamiltonian}

The symmetry distinction between $z$ and $x,y$ type bonds implies that if the Kitaev coupling is strong, the magnetic susceptibility should have a distinctive $z$-axis response compared to its $x,y$ axes response, at least at temperatures above the magnetic transition. If the Kitaev coupling $K_\gamma$ on the $z$-bonds is more ferromagnetic than the Kitaev coupling $K_d$ on the $x,y$-bonds, it suggests an anisotropic susceptibility with larger response along $z$. Exactly such an anisotropy is observed in the 3D-Li$_2$IrO$_3$ experiment\cite{11}. However, to preserve the strong $z$-axis susceptibility which is observed also below the ordering transition, the resulting magnetic order should not have spins aligned along the $z$ axis. This places a condition on the magnetic coupling, to disfavor magnetic order alignment along $z$, which is partially at odds with the condition necessary to favor susceptibility anisotropy with large $\chi^z$.

To achieve strong anisotropy in the susceptibility $\chi$, the Heisenberg couplings must be dominated by anisotropic single-spin-component exchanges, in this case large ferromagnetic Kitaev exchanges. Since the low temperature phase is not a ferromagnet, the Heisenberg couplings must be antiferromagnetic. This region of parameter space hosts two types of magnetic order,
Stripy-Z and Stripy-X, with different symmetry properties. With no additional anisotropies, Stripy-Z nominally hosts spins aligned along the z axis and can thus be ruled out. A more general property of the Stripy-Z phase is that, because of the two symmetry-inequivalent z-type bonds, it should generically exhibit a nonzero net moment. We therefore focus on Hamiltonians within the Stripy-X phase.

The constraints on the couplings can be seen explicitly by treating the Hamiltonian classically, and extracting susceptibility by classical mean field theory (details are given in Appendix [3]). Within the mean field treatment of the Stripy-X phase (in the regime $K < 0$, $J > 0$), the transition temperature is given by $T_N = (J_c + |K_d|)/4$. The susceptibility peaks at this temperature, taking values

$$\chi^{bb}(T_N) = (g^b)^2 \mu_B^2/(2(J_c + J_d) - (|K_c| - |K_d|))$$

and with $\chi^{cc}$ similar to $\chi^{aa}$. The observed susceptibility anisotropy then suggests a large value for the difference $|K_c| - |K_d|$. However, the stability of the Stripy-X phase against Stripy-Z order is controlled by the constraint

$$|K_c| - |K_d| < 2(J_c - J_d).$$ (3)

There is a window of parameters which fit the data. One possibility for the couplings, as shown in Fig. [3], is (in meV): $K_c = -17$, $K_d = -7$, $J_c = 6.3$, $J_d = 0.8$. The Hamiltonian with this set of parameters was also studied beyond the classical limit, using tensor product states within the infinite dimensional large-$\ell$ approximation, and determined to lie within the Stripy-X quantum phase.

This parameter regime of the fit, large ferromagnetic Kitaev exchange and small antiferromagnetic exchange, is consistent with Jackeli and Khaliullin’s original proposal[22,23] and with the recent Na$_2$IrO$_3$ ab initio[24]. The extent of the anisotropy is qualitatively similar to the Na$_2$IrO$_3$ ab initio prediction as well: the parameters computed for Na$_2$IrO$_3$ are[23] $K_c = -30.7$, $K_d = -23.9$, $J_c = 4.4$, $J_d = 2.0$ meV, and larger anisotropy is expected for the H-1 lattice because the special c bonds directly form the special axis of its $Cccm$ space group.

Note that for the model Hamiltonians we consider, the principal axes of the susceptibility tensor are $x, y, z$ rather than the crystallographic axes $a, b, c$. Terms arising from the global symmetries of the crystal will likely change the principal axes to match the crystallographic ones. To compare with experiment without adding any such additional terms, we measure the susceptibility tensor along the crystallographic axes as shown in Fig. [3].

The weakly anisotropic $g$-factors, experimentally determined at high temperature for each of the crystallographic axes, are then incorporated into each axis of $\chi$. We use the $g$-factors $g^{x+y} = g^{z} = 1.95$, $g^{x-y} = 2.35$. Note that the overall scale of the $g$-factors needed to fit the susceptibility, which was measured experimentally on a single crystal, carries an uncertainty associated with the uncertainty of estimating the number of Li$_2$IrO$_3$ formula units in the crystal.

The primary conclusion of this analysis is the argument that the bond-anisotropic Kitaev-Heisenberg Hamiltonian is appropriate for describing current experimental data on 3D-Li$_2$IrO$_3$ and requires quite large Kitaev exchanges $K >> J$. A secondary conclusion is a prediction for the low temperature magnetic order of 3D-Li$_2$IrO$_3$, namely the Stripy-X order of the fitted Hamiltonian, shown in Fig. [2]. We do not expect the spin direction to be locked to $\hat{x}$ in the real crystal, though we expect it to lie within the $(a, c)$ (or equivalently $(x, z)$) plane.

IV. THREE DIMENSIONAL QUANTUM SPIN LIQUIDS

Let us now tune the Heisenberg couplings $J$ to zero, taking the limit of a pure Kitaev Hamiltonian. Though this limit does not describe the experiments on Li$_2$IrO$_3$, it offers a wide range of interesting phenomena associated with 3D fractionalization, which may turn out to be experimentally accessible at a future date.

A. Solution via Majorana fermion mapping

Kitaev’s solution[12] of the honeycomb spin model relies on a local condition — each site touches three bonds carrying the three different Kitaev labels — and hence may be generalized to lattices with $z = 3$ coordination number. In order to discuss important subtleties which will arise later (in infinite dimensions), let us briefly review the solution here. The $S = 1/2$ algebra is represented in an enlarged Hilbert space via four majorana fermions

$$S_i^a \rightarrow \frac{1}{2} \chi_i^a \chi_i^a, \quad \{\chi_i^a, \chi_i^b\} = 2\delta_{i,i'}\delta_{a,a'}. \quad (4)$$

The enlarged Hilbert space Kitaev Hamiltonian $\tilde{H}$ is then a free majorana fermion $\chi_i^0$ minimally coupled to a $Z_2$ vector potential $a_{i,j}$ with $e^{i\pi a_{i,j}} \equiv u_{i,j} \equiv i\chi_i^a\chi_j^b$ living on links $\langle ij \rangle$. The gauge field operators $u_{i,j}$ all commute with each other and with $\tilde{H}$, so $\tilde{H}$ may be diagonalized by solving a free fermion problem for each gauge field configuration $\{u_{i,j} = \pm 1\}$. Here $u_{i,j}$ is identified as a gauge field because, while in the enlarged Hilbert space it is a simple $Z_2$ bond variable set by the majorana fermion occupancy, there is a set of site operators $D_i \equiv \chi_i^0\chi_i^1\chi_i^2\chi_i^3$ which are the identity within the physical spin Hilbert space but act as a lattice gauge transformation on the link variable $u_{i,j}$. Projection to the physical spin Hilbert space is implemented by symmetrizing over all local gauge transformations $D_i$, with the projection operator $P = \prod_i (1 + D_i)/2$.

Let us now discuss the consequences of this majorana fermion solution for the 3D trivalent lattices. Some of
the phenomenology was previously explored\cite{56} for a 3D lattice whose connectivity graph matches the hyperhoneycomb's. The projection from gauge to physical Hilbert space is aided by gauge invariant operators whose eigenvalues, commuting with the Hamiltonian, label physical sectors of states. These closed Wilson loops are the usual \( Z_2 \) fluxes piercing the elementary plaquettes. Flipping \( u_{i,j} \) on a bond inserts flux in adjoining plaquettes. As discussed earlier, the 3D lattices possess symmetries as well as graph connectivity which distinguish one bond type, \( z \), from the other two bond types \( x \) and \( y \). On the hyperhoneycomb lattice, flipping \( u_{i,j} \) on a \( z \) type bond creates fluxes on the eight adjacent plaquettes, while flipping \( u_{i,j} \) on \( x \) or \( y \) type bonds changes the flux on the only six adjacent plaquettes. On the \( \mathcal{H}-1 \) lattice, the elementary plaquettes come in multiple forms, consisting of \( \ell = 6 \) hexagons together with larger \( \ell = 14 \) plaquettes.

**B. Extended flux loop excitations in the 3D QSL**

The gauge field sector on the 3D-lattice Kitaev model is a 3+1D \( Z_2 \) lattice gauge theory\cite{57}. The product of \( u_{i,j} \) around a minimal closed contour gives the flux through an elementary plaquette of the lattice. The product of fluxes on plaquettes surrounding an elementary volume element multiplies to the identity: this is equivalent to the fact that there are no magnetic monopoles in the \( Z_2 \) theory. Each elementary volume carries a zero monopole charge and thus acts as a constraint, forcing the number of flux lines piercing the volume to be even. These constraints ensure that the flux lines only appear within closed flux loops.

It is important to note that while in the 2D honeycomb case the magnetic fluxes are the gauge invariant result of projecting the gauge theory, in the case of three spatial dimensions, individual fluxes are not gauge invariant. Rather, only closed flux loop configurations are the physical gauge invariant excitations of the model. The individual fluxes cannot be gauge invariant labels of sectors of the Hamiltonian since they don’t even correctly count the physical degrees of freedom of the gauge theory. The constraint of closed loops fixes this counting: the closed magnetic loop configurations exactly label the gauge invariant sectors of the \( \{ u_{i,j} \} \) after projection.

This can be seen as follows (explicitly verifying this statement in the \( \mathcal{H}-1 \) and \( \mathcal{H}-0 \) lattices is also straightforward). Viewing the space-filling lattice (with periodic boundary conditions) as a large simplicial complex, we may count the number of simplices of every dimension – sites, bonds, plaquettes and enclosed volumes. A formula related to the Poincare formula for the Euler characteristic then shows that

\[
N_{\text{sites}} - N_{\text{bonds}} + N_{\text{plaquettes}} - N_{\text{volumes}} = 0.
\]  

The combination \( N_{\text{plaquettes}} - N_{\text{volumes}} \) is of interest here, since every plaquette is associated with a flux, but each enclosed volume presents a condition on the adjacent fluxes (they must multiply to the identity). This constraint, due to the lack of monopoles in the \( Z_2 \) gauge theory, is responsible for the flux lines forming closed flux loops. The number of independent such loops is given by the number of possible flux lines minus the number of constraints, ie \( N_{\text{flux loops}} = N_{\text{plaquettes}} - N_{\text{volumes}} \).

Furthermore let us restrict to our case of interest where sites have coordination number \( z = 3 \) and so \( N_{\text{bonds}} = (3/2)N_{\text{sites}} \). Then the formula becomes

\[
N_{\text{flux loops}} = N_{\text{sites}}/2
\]

as required: the gauge field flux sector hosts half of the spin degrees of freedom, while the majorana fermion particle sector hosts the other half.

Lieb’s flux phase theorem\cite{59}, which shows that the 2D honeycomb ground state has zero flux per \( \ell = 6 \) hexagon, suggests that the \( \ell = 6, \ell = 10 \) and \( \ell = 14 \) loops of the 3D \( \mathcal{H}-1 \) and \( \mathcal{H}-0 \) lattices, whose length is equal to 2 mod 4, should also carry zero flux in the ground state. We have checked numerically that the ground state on small finite systems lies in the sector with no flux loops.

**C. Majorana fermion excitations**

The Kitaev QSL possesses emergent quasiparticles which are fermionic, arising out of the interacting bosonic spin model. The emergent fermions are as real as physical electrons, but carry no usual electric charge; and moreover are majorana (self-adjoint), related to the particle-hole-symmetric excitations of superconductors. As in the 2D honeycomb model, in which the fermion dispersion possesses graphene’s Dirac nodes, the fermionic dispersion in the 3D lattices is gapless for the isotropic model. The sublattice symmetry present in all the 3D harmonic
honeycomb lattices – they are all bipartite – ensures that
time reversal remains a symmetry in the QSL phase, and
the majorana fermion spectrum is particle-hole symmet-
ric about the 1D nodal lines (as in 2D, nodes carry codi-
menion 2). In the 3D harmonic honeycomb lattices the
nodal lines form a closed 1D contour. Indeed this is the
dispersion of a nodal 3D superconductor: the majorana
fermions are gapless along a 1D ring of points in the 3D
momentum space. The shape of this closed contour for
the \( \mathcal{H} \)-1 lattice is set by the two equations \( \mathbf{k} \cdot \mathbf{a} = 0 \) and
\( \cos (k \cdot a/2) + \cos (k \cdot b/2) = 1/2 \), in the notation of Ap-
pendix [E] and as shown in Appendix [F]. This contour of
low energy excitations is plotted in Fig. 8.

Within each sector with its associated flux loop con-
figuration, we may study how the majorana fermions
propagate. The fermions are charged under the gauge
field, and hence interact with the magnetic loop excita-
tions through an Aharonov Bohm effect, analogous to
that occurring between electrically charged electrons and
conventional E\&M magnetic flux lines. The interaction
is as follows: when a fermion winds through the interior
of a magnetic flux loop, it encircles one flux line and re-
ceives a minus one (\(-1\)) phase to its single particle wave
function.

D. Spin-spin correlators

The spin-spin correlators may be computed straight-
forwardly within the fermion mapping; as in 2D, they
are only nonzero between spins on nearest-neighbor sites
and then only between spin components matching that
bond’s Kitaev label. Hence the nonzero spin correlators
\( G \) are also equivalently the energy \( E \) carried by the bond
(divided by the coupling), specifically \( G = E/K \). For
notational simplicity we quote correlators \( G \) for \( K < 0 \),
in which case the correlators are positive; for \( K > 0 \),
correlators simply gain a minus sign. Here we report re-
results at the isotropic point of the Hamiltonian (though of
course lattice symmetry still comes into play). We find
that the average energy per bond is
\[ E_0^{3D,0} = -0.1284|K| \]
for the 3D \( \mathcal{H} \)-0 and
\[ E_0^{3D,1} = -0.1290|K| \]
for the 3D \( \mathcal{H} \)-1, only 2% higher than the 2D honeycomb result
\[ E_0^{2D} = -0.1312|K| \].

For the hyperhoneycomb \( \mathcal{H} \)-0 lattice, the \( z \) bonds and
\( x, y \) bonds correlators are
\[ G_z = 0.1314, \quad G_{x,y} = 0.1268. \] (7)
The harmonic \( \mathcal{H} \)-1 lattice has two symmetry-distinct
types of \( z \) bonds: those within hexagons (“\( z[\mathbf{h}] \)”) and
those within length-14 loops (“\( z[\mathbf{f}] \)”). The correlators are
\[ G_{z[\mathbf{h}]} = 0.1337, \quad G_{z[\mathbf{f}]} = 0.1269, \quad G_{x,y} = 0.1283. \] (8)
The large correlations on hexagon-\( z \) bonds could be ex-
plained as strong resonances within a hexagon, combined
with a lattice symmetry effect that, for both \( \mathcal{H} \)-0 and \( \mathcal{H} 
-1 \), gives stronger correlations on the special-axis \( z \) bonds.
Surprisingly, this global symmetry effect is almost as
powerful as the hexagon resonances: it produces \( x,y \)
-bond correlators which are only slightly stronger than
those on the cross-hexagon-stripe \( z[\mathbf{f}] \) bonds.

E. Nodal contour under Toric Code anisotropies
and broken symmetries

Increasing the coupling strength \( K \) on one bond type
is an anisotropy which preserves exact solvability of the
model, in 3D as well as 2D. Increasing \( K \) on bonds of
one Kitaev-type shrinks down the nodal contour, until
it vanishes and gaps out the fermions when the Kitaev
exchange for any one bond type becomes larger than the
sum of the other two. However, consider that the \( \mathcal{H} \)-0
and \( \mathcal{H} \)-1 lattice symmetries already distinguish \( z \)-bonds
and their axis \( \hat{c} \sim \hat{x} - \hat{y} \) as a special direction; increas-
ing the strength of \( z \) bonds is an anisotropy which is
generically expected to arise given the crystal symme-
tries. Increasing \( z \) bond strength shrinks the nodal con-
tour around the origin \( \hat{k} = 0 \) until the majoranas gap
out. But anisotropies for \( x \) or \( y \) type bonds do break a
symmetry of the isotropic model. When increasing bond
strength on \( x \) or \( y \) bonds, the nodal contour goes through a
Van Hove singularity as it expands to touch the BZ sur-
face, and then becomes a gradually shrinking loop around
the BZ corner. However, while these details of the nodal
contour are different between symmetry-breaking \( x, y \)
and non-symmetry-breaking \( z \) anisotropy, the resulting

FIG. 8. Momentum space contour of majorana gap-
less nodes. In the gapless 3D spin liquid phase, the major-
afermions are gapless at a set of wavevectors which forms
a 1D contour in the 3D momentum space. Here the con-
tour is shown for the \( \mathcal{H} \)-1 model (\( \mathcal{H} \)-0 is visually identical)
in the parent simple orthorhombic coordinate system of Ap-
pendix [E]. Coupling strength anisotropies shrinks this contour
until it collapses to a point and then gaps out, yielding the
gapped spin liquid phase, smoothly connected to one of two
3D toric codes.
phase diagram of the spin liquid phase is that shown in Fig. 7, identical to that of the symmetric 2D honeycomb lattice. The same phase diagram was also found in infinite dimensions, in the computations on finite open trees discussed below.

Each of the 3D lattices supports two different types of limits of large anisotropy, $z$ and $x/y$ types, which are associated with different three dimensional Toric Code models living on different $z = 4$ lattices. Each of these 3D Toric Code models is a pure $\mathbb{Z}_2$ gauge theory, with commuting plaquette terms formed by sites on a particular $z = 4$ lattice set by the type ($z$ or $x, y$) of anisotropy. The Toric Code lattices are easily constructed by collapsing the strong-coupled bond into a site. The Toric Code $\mathbb{Z}_2$ flux operators act on plaquettes of reduced size: the $\mathcal{H}$-0 decagons turn into Toric Code hexagon plaquettes, while $\mathcal{H}$-1 hexagons (as in 2D) turn into Toric Code square plaquettes.

Breaking time reversal symmetry with an external magnetic field induces oriented imaginary second neighbor hopping of the majorana fermions. The sign (orientation) of this imaginary hopping, necessary for majorana fermions, is set (as in 2D) by the sign of the permutation of the two Kitaev bond labels traversed. Except for certain multi-parameter fine tuning, we find that breaking time reversal fully gaps out the entire majorana nodal contour.

F. Fractionalization in 3D: extended loops and finite temperature confinement transition

Enlarging spatial dimensionality from two to three dimensions changes the nature of the spin liquid phase; the two most interesting differences involve fermions and finite temperature. In the two dimensional spin liquid away from the exactly solvable point, the flux excitations gain dynamics and interact with the majorana fermions; the low energy excitations could then be bound fermion-flux pairs, composite particles with simple bosonic statistics. In contrast, consider the three dimensional spin liquid; here fluxes are not pointlike particles but rather closed magnetic loops, so the emergent fermions cannot merely bind a (point-like) flux to transmute into bosons, and thus their 3D fermionic statistics are more robust. While fermions can e.g. bind into Cooper pairs to disappear from the lowest energy theory, a fundamental excitation in the model still necessarily preserves fermionic statistics. The fermions remain until a phase transition either confines them or transmutes them into bosons via a more complicated mechanism such as that recently explored in transitions between symmetry protected topological phases.

Three dimensional spin liquid phases generally admit a key characteristic distinguishing them from 2D spin liquids: the 3D spin liquid phases survive to finite temperatures. Such is true for the Kitaev 3D spin liquid phase, which undergoes a distinct entropy-driven phase transition to a classical paramagnet. In 2D QSLs a finite density of fluxes exists at any nonzero temperature; the fermions gain a phase of ($-1$) when encircling each of the fluxes and the resulting destructive interference results in a $T = 0$ confinement transition to the paramagnet phase. But in the 3D QSL, magnetic fields appear in extended loop excitations, whose energy is proportional to their length via an effective loop tension. The loop energy diverges with its length. At finite temperature there is a finite density only of short loops, whose small cross-sectional area renders them invisible to the fermions. A finite probability for flux-encircling paths occurs only with macroscopically large loops, which cost diverging energy and hence appear at vanishing density. Entropy however favors longer loops, and so the free energy at finite temperature $T$ for a loop of length $L$ appears as (for long $L$)

$$F(L; T) = (\tau + \delta\tau(T) - \delta T) L$$

where $\delta$ is the entropy contribution to the loop tension, roughly the natural logarithm of the coordination number of the dual lattice (where magnetic loops live), $\delta \approx \log(2)$; $\tau$ is the zero temperature flux loop tension; and $\delta\tau(T)$ is the contribution to the effective loop tension at finite temperature due to interactions mediated by the gapless fermions.

Because the entropy is likely the dominant contribution and appears with a negative sign, the effective magnetic loop tension renormalizes to lower values at finite temperature. At a temperature $T_c$ the tension becomes negative and proliferates arbitrarily large magnetic loops in a transition analogous to Kosterlitz-Thouless flux unbinding, which then confine the fermions. We estimate the critical temperature $T_c$ by computing the zero temperature value of the magnetic loop tension $\tau$ in the isotropic Hamiltonian, finding the result

$$\tau = 0.011/K$$

for both $\mathcal{H}$-1 and $\mathcal{H}$-0 in different geometries and for different loops roughly independent of the loop shape, underlying bond/plaquette type, and for large loop lengths of up to 30 cross-sites (on $\mathcal{H}$-0), suggesting $T_c \sim |K|/100$.

V. QUANTUM PHASE DIAGRAM IN AN INFINITE-D APPROXIMATION

The Kitaev-Heisenberg model suffers from the “sign problem” of frustrated quantum Hamiltonians: unbiased algorithms for computing its phase diagram require computational costs scaling exponentially with system size, a problem greatly exacerbated in a three dimensional lattice. unbiased reliable computations of the phase diagram on the three dimensional lattices are not possible at present time.
A. Duality results for the magnetic phases

Even on the 3D lattices, definitive conclusions for the magnetically ordered phases can still be made due to a general feature, the Klein duality, exhibited by Kitaev-Heisenberg model. The following discussion applies to any bipartite lattice, including the tree lattice in infinite dimension, as well as all 3D harmonic honeycomb lattices. Since these lattices are bipartite, simple Neel antiferromagnetic order is the expected ground state for the Heisenberg antiferromagnet Hamiltonian. The Neel AF and FM orders map under the Klein duality to three dimensional generalizations of stripy and zigzag orders. Assuming that the unfrustrated Neel order is indeed the ground state for AF Heisenberg exchange (as may be verified by quantum Monte Carlo at the sign-problem-free SU(2) point), we conclude that all four of these magnetic phases must be present in the phase diagram.

B. Loop length as a control parameter

To capture the full quantum phase diagram including the quantum spin liquid phases, we employ a limit inspired by the geometry in the hyperhoneycomb lattice. Its shortest loops are the 10-site decagons. Treating this loop length \(\ell = 10\) as a large parameter and formally taking it to infinity, we find the loopless \(\ell \to \infty\) Cayley tree or Bethe lattice with \(z = 3\) connectivity in infinite dimensions. The tree lattice approximation \(\ell \to \infty\) enables a solution using entanglement-based methods originally developed for 1D systems, which rely on efficient representations of matrix or tensor product states. The key for such efficient representations is that entanglement is carried by bonds: cutting a single bond serves as an entanglement bipartition, and a singular value decomposition fully determines the entanglement spectrum which can be associated with this bond.

This tree lattice is infinite dimensional in the sense that for finite trees with \(N\) sites, a finite fraction of sites \(f_s \approx (z - 2)/(z - 1)\) is on the boundary. But note that this is an opposite limit of infinite dimensionality from the one commonly taken in mean field theories, which assume infinite connectivity \(z \to \infty\): here we crucially fix \(z = 3\). Entanglement based algorithms within our infinite-D approximation can work with the low coordination number \(z = 3\) and low spin \(S = 1/2\), capturing the associated strong quantum fluctuations.

C. iTEBD on the tree lattice

The large loop \(\ell \to \infty\) limit of the hyperhoneycomb (or higher harmonic honeycomb) lattice, which yields the infinite-D Bethe tree lattice, admits a numerical solution of the gapped phases in the phase diagram. The key is that cutting a single bond gives an entanglement bipartition (as shown in Fig. 9) with an entanglement spectrum which is associated with that bond. Hence gapped states can be represented efficiently as tensor product states, and the full machinery of entanglement based algorithms can be used. We choose to use a variant of the infinite system size time-evolving block decimation algorithm (iTEBD)\(^{61}\). The iTEBD algorithm has been previously used to study various Hamiltonians on tree lattices, with phase diagrams containing magnetic phases\(^{62–64}\), nonmagnetic phases\(^{65}\) and even a symmetry protected topological phase related to the AKLT Hamiltonian\(^{66}\). The iTEBD algorithm is especially useful here since it works directly in the thermodynamic limit, avoiding the issues which plague finite trees.

Specifically, each update step in the algorithm, such as an imaginary time evolution step in iTEBD, must be followed by an operation which restores the state into a correctly normalized tensor product state. This requires cutting the TPS into two parts and computing the entanglement spectrum across the cut, via a singular value (i.e. Schmidt) decomposition. These singular values are associated with the bond and placed between the adjacent site tensors when one contracts the TPS in order to measure observables. The tree lattice offers all these properties and hence entanglement based algorithms developed for 1D systems may be adapted to the tree\(^{65}\).

The iTEBD algorithm performs imaginary time evolution (i.e. soft projection to the ground state) within a restricted set of tensor product states, allowing it to find a good approximation to gapped periodic ground states with sufficiently local entanglement. Since it works on an infinite system, it always chooses one minimally entangled ground state, i.e. it can exhibit spontaneous symmetry breaking. To enable such symmetry breaking consistent with the expected magnetic ordering, we choose a unit cell with 8 site tensors and 12 bond (Schmidt) vec-
tors as shown in Fig. 9, employing 24 update cycles in each imaginary time evolution step. The key parameter for TPS algorithms is the bond dimension $\chi$, serving as a cutoff on the number of entangled states. The computational costs scale polynomially in $\chi$, but for computations on the tree the exponent is fairly high, with scaling of $\chi^6$. The Kitaev-Heisenberg model harbors additional computational complexity due to its lack of spin rotation symmetry, the large unit cell necessary to describe its magnetic phases and the emergent small energy scales in its quantum spin liquid phases. Our results are roughly independent of $\chi$ for $\chi \geq 6$; we report data for computations using $\chi = 12$, after verifying convergence through $\chi = 6, 8, 10$, and run $2 \times 10^7$ singular value decompositions (SVDs) for each parameter point. To preserve normalized tensors during the imaginary time evolution, we intersperse evolution steps with zero imaginary time (i.e. pure SVD steps), as well as work with short time steps which are gradually reduced to $10^{-6}$ in inverse energy. The algorithm enables us to capture any periodic state consistent with our 8-site unit cell whose entanglement is sufficiently local, as is the case for the magnetically ordered phases we expect to find as well as for the gapped quantum spin liquids.

D. Parametrized Hamiltonian

The bond-anisotropic Kitaev-Heisenberg Hamiltonian, Eq. (1) involves one overall scale and and three free parameters. In computing the quantum phase diagram via tensor product states, we will focus on two of these parameters. The Kitaev exchange, generated by spin-orbit coupling, may be especially sensitive to the bond anisotropy; we therefore focus on the effects of bond anisotropy on the Kitaev term, leaving the study of the large-$\ell$ quantum phase diagram with Heisenberg term bond anisotropy for future work. Note however that we have performed calculations on the full Hamiltonian Eq. (1) in the neighborhood of the experimentally extracted parameter values shown in Fig. 3, finding the magnetic Stripy-X phase and a nearby phase boundary to the magnetic Stripy-Z phase. We shall now record the resulting two-parameter Hamiltonian, together with a few different useful parametrizations of its couplings, which we use to present various figures. In particular, we define polar coordinates with $r = 1 - a$ and two different angle parameters, $\phi$ or the alternative $\theta$. The Hamiltonian is:

$$H = \sum_{\langle ij \rangle} \left[ K_{\gamma ij} \vec{S}_i^\gamma \cdot \vec{S}_j^\gamma + J \vec{S}_i \cdot \vec{S}_j \right]$$

$$K_{\gamma ij} = K \times \begin{cases} (1 - a) & \text{on } \gamma_{ij} = x, y \text{ bonds} \\ (1 + 2a) & \text{on } \gamma_{ij} = z \text{ bonds} \end{cases}$$

$$K = 2 \sin(\phi), \quad J = \cos(\phi); \quad \theta = \pi/2 - \phi.$$  

The Kitaev-Heisenberg spin Hamiltonian, with the angular $\phi$ parametrization relating the strengths of Kitaev and Heisenberg coupling, is extended with this symmetry-allowed anisotropy, parametrized by $-1/2 \leq a \leq 1$.

Let us here also note the extension of the Klein duality discussed in section IIIA above, to the case of nonzero anisotropy. Recall that the Klein duality relates parameters by $\tan \phi' = -(1 + \tan \phi)$ for the isotropic case $a = 0$. The transformation exposes a hidden ferromagnet even with anisotropy, at $\phi_{\text{hidden FM}} = -\arctan[1/(1-a)]$. Observe that the anisotropy reduces the symmetry at the hidden-FM point from SU(2) to U(1): the dual Hamiltonian is no longer Heisenberg but rather is an easy-axis ferromagnet. The key observation, that its ground state is an exact product state, remains unchanged.

For the pure Kitaev Hamiltonians, $a = 1/4$ is the transition point between the gapless ($-1/2 \leq a \leq 1/4$) and gapped ($1/4 < a \leq 1$) spin liquid phases. In addition to the isotropic case $a = 0$ we focus on a particular anisotropy value within the gapped QSL regime, $a = 1/2$. We sample other values of the anisotropy as well in order to generate the global phase diagram shown in Fig. 2.

E. Magnetically ordered phases

Let us begin our analysis of the tensor product state computation by discussing the magnetic phases captured by the iTEBD algorithm on the tree lattice. We use a variety of measures to identify phases and the phase diagram. Magnetically ordered phases can be captured directly by their magnetic order parameter, since the iTEBD always outputs a single symmetry broken ground state. This analysis is shown in Fig. 10 for the isotropic model, and in the appendix in Fig. 13 for $a = 1/2$ anisotropy. The four magnetic phases expected from the discussion in section IIIA above are observed. Phase transitions are also identifiable, as always, through the first and second derivatives of the energy. As a simple benchmark we have verified that the energy is always bounded from above by the energy of the expected classical product state and from below by the optimal energy for any given site in its surrounding cluster, as may be seen in Fig. 13. Phase transitions are also signaled by peaks in the entanglement carried by the various bonds in the tensor product state, and finally of course the phases can be identified using the spin-spin correlation functions; these two measures are shown in Fig. 17. We also verify that the entanglement correctly vanishes at the exact (hidden-)ferromagnet points.

The particular parameters of the direct first order phase transitions between the magnetic phases should be insensitive to dimensionality and loop length $\ell$ for sufficiently large $\ell$, since the quantum fluctuations on top of these classical phases need to propagate a distance of $\ell$ sites to distinguish one lattice from another. The smallest value for $\ell$ we encounter is $\ell = 6$, so quantum fluctuations in these magnetic orders must traverse at least six nearest-neighbor bonds to distinguish the honeycomb from the $H$-1 or $H$-0 lattices. Hence we expect that the
2D honeycomb, 3D harmonic honeycomb and infinite-D tree lattices will exhibit similar magnetic transitions. Indeed the parameters we find for the tree lattice within iTEBD are essentially indistinguishable from those of the 2D honeycomb model. As a function of anisotropy, the location of magnetic transitions can also be compared to a classical mean field theory. We find similar behavior, with larger differences closer to the isotropic point; see Appendix C for details.

![Energy and Magnetism](image)

**FIG. 10.** *Kitaev-Heisenberg isotropic phase diagram via iTEBD.* The magnetic order parameters of the four phases are directly observed by iTEBD, working in the thermodynamic limit. The stripy and FM phases surround an exact solution with saturated magnetic order parameter \( m = \hbar / 2 \); Neel and zigzag phases exhibit a moment reduced by quantum fluctuations. The energy per bond (purple) also provides the phase transitions, as well as benchmarking (see Fig. 11). The QSL phases exist around the Kitaev points at \( \theta / \pi = 0, 1 \) but here are gapless and cannot be numerically captured with finite entanglement.

**F. The quantum spin liquid within iTEBD in infinite-D**

Turning to the phase diagram of the QSL phases and their immediate surrounding, we first must restrict ourselves to the regime with sufficiently strong anisotropy so that the emergent majorana fermions are gapped, at \( a > 1/4 \). The gapped spin liquids can be well approximated by the tensor product states we use. In Fig. 11, we show the spin liquid phase for \( K < 0 \) and the nearby stripy and ferromagnetic orders. The spin liquid can be identified by its lack of magnetic order parameter as well as by its interesting entanglement as we discuss later; phase transitions are again seen in energy derivatives and as peaks in the entanglement entropies. The extent of the spin liquid in this computation is small, covering about a tenth of a percent of the phase diagram, but nonzero; more importantly, the extent of the spin liquid is the same throughout the full range of bond dimensions we study.

While the QSL phase lacks an order parameter and thus avoids a direct identification of the type in Fig. 10, the exact solution of the Kitaev model on the infinite-D tree allows us to uncover the unique fingerprints of the exact QSL, and use them to unequivocally identify the QSL phase within iTEBD. Each such set of fingerprints can be computed as a function of anisotropy for the pure Kitaev model across the entire gapped phase \( 1/4 < a < 1 \).

We can of course compute the energy curve as a function of \( a \) within the majorana solution, and find agreement as shown in Fig. 18 but energies are notoriously lousy fingerprints for spin liquid phases. We also compute the spin-spin correlators within the iTEBD and find that they match the correlators we compute within the exact solution, as shown again in Fig. 18. This is more robust but, we only study correlators across a few neighbors, grossly insufficient for identifying the QSL.

Instead, the most valuable set of fingerprints is furnished by the entanglement entropy carried by each bond. The entanglement spectrum is an inherent part of the tensor product state description and is easily accessible from the iTEBD. Spurious “accidental” symmetry breaking exhibited by the iTEBD ground state, caused by the large unit cell and the merely finite imaginary time evolution duration, complicates the bond entanglement entropies but still permits comparison with the entanglements computed in the exact solution. This comparison is shown in Fig. 12, confirming that the iTEBD algorithm is indeed capturing the emergent Majorana fermions of the quantum spin liquid fractionalized phase in infinite dimensions.

**FIG. 11.** *Gapped spin liquid at \( K < 0 \) and surrounding magnetic phases, via \( \chi = 12 \) iTEBD.* Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotropy, here \( a = 1/2 \) iTEBD. Sufficient bond anisotrop
the iTEBD computation only if we assume that the gauge field sector contributes entanglement only on bonds set as strong by the anisotropy parameter. To understand this key subtlety, we now turn to the study of the exact Kitaev $Z_2$ quantum spin liquid on the loopless tree lattice, focusing first on the fermion sector followed by the more subtle $Z_2$ gauge field sector.

G. Majorana fermions on finite open trees

The spectrum of Majorana fermions hopping on the infinite tree can be computed exactly using recursion on propagators. However, we will mainly compute entanglement entropies associated with a bipartition and thus choose to work with finite trees. The spectrum of a finite tree adjacency matrix has an extensive number of zero modes, which may be counted for any finite tree by noting that the number of bonds is $N_b = N_s - 1$, reduced from the expected $N_s z/2$ by a fraction $f_b \approx (z-2)/z$; i.e., about $N_s/3$ of the eigenvalues are finite size boundary effects. For a site-centered tree they may be counted exactly using Lieb’s sublattice imbalance theorem by observing the unbalanced occupation in the bipartite tree’s $A$ and $B$ sublattices, $[N_B - N_A]/N_s = (z-2)/z + 1/N_s$. On a bond-centered tree, in addition to the identically zero boundary eigenvalues there is an isolated low-lying eigenvalue whose gap vanishes with increasing system size, which is also associated with the boundary. We may thus take the bulk tree thermodynamic limit by discounting these boundary eigenvalues of finite tree adjacency matrices.

Using the bulk fermion correlation function and the reduced correlator for a bipartition associated with cutting the central bond in a bond centered-tree, we compute the entanglement spectrum and entropy of the bipartition, which resides on this central bond. An alternative approach for computing the entanglement entropy entails subtracting the $T = 0$ thermal entropy of the finite open tree from the naively computed entanglement entropy of the bipartition, which again yields the entanglement entropy of the single bond cut without the $T = 0$ thermal entropy of the boundary zero modes. See details in appendix A. The approaches agree, yielding the entanglement entropy contributed by the fermion sector of the exact quantum spin liquid.

H. $Z_2$ gauge theory on the loopless tree

Entanglement is also contributed by the $Z_2$ gauge sector of the tree Kitaev model; in order to describe this contribution we shall now discuss the unusual subtleties which arise in a $Z_2$ gauge theory on a loopless lattice. We begin by noting that the gauge theory is necessarily well defined even on the loopless tree lattice, since it arises from a well defined spin model. For $N_s$ sites there are $N_s/2$ gauge-invariant sectors after projecting the gauge fields, which combine with the $N_s/2$ Majorana fermion degree of freedom to give the $N_s$ doublet degrees of freedom for the lattice of $S = 1/2$. In 2D the $N_s/2$ sectors are labeled by fluxes; in 3D, by magnetic field loops; and in infinite dimensions, they may be associated with $N_s/2$ particular infinite magnetic field lines extending across the (infinite) lattice. These field lines stretching across the system are intimately related to a more familiar set of infinite products of operators: in 2D topologically ordered phases, field lines can wind around the periodic boundary conditions. The resulting flux piercing the torus costs an energy which vanishes in the thermodynamic limit, and these operators generate the topological ground state degeneracy on the torus. On the tree lattice there is an extensive number of such operators, contributing an extensive ground state degeneracy $2^{N_s/2}$.

This degeneracy may also be seen by counting conserved quantities associated with infinite products of local operators within the original quantum spin Hamiltonian. Working either within the original spin model or within the gauge theory, we must count the number of such independent paths on the tree lattice. A moment’s thought shows that independent paths may be counted as paths from a given boundary site to any other boundary site on a finite open tree. For the purposes of this counting the open tree may be compactified by identifying all boundary sites, in which case the strings again form conventional closed loops carrying a flux. In any case the counting gives exactly $N_{\text{boundary sites}} - 1 = N_s/2$ operators, in agreement with the gauge field mechanism for $2^{N_s/2}$ degeneracy. Thus on the tree lattice within a full thermodynamic limit, the gauge theory collapses to an extensive degeneracy of $2^{N_s/2}$ states.

A $Z_2$ gauge theory contributes $\log 2$ of entanglement
for every two bonds in the entanglement cut, or \( \log(2)/2 \) entanglement per bond.\(^{14}\) Intuitively, the gauge field carries half of the information content of a physical gauge-invariant \( \mathbb{Z}_2 \) link variable. An additional global term of the topological entanglement entropy is generally expected to arise in the gauge theory, but does not appear on the tree lattice single-bond entanglement biartitions: the only entanglement is that associated with the bond. Thus on the tree lattice we expect the single bond entanglement cut to carry \( \log(2)/2 \) entanglement from the gauge sector, in addition to any fermionic entanglement.

However, when comparing to the iTEBD result, we find that the iTEBD choice of ground state within the gauge theory’s degenerate manifold effectively quenches the \( \mathbb{Z}_2 \) gauge sector entanglement on weak bonds, giving gauge sector entanglement only on strong bonds, which retain the gauge field entanglement \( \log(2)/2 \). This is reasonable since there are two \( S = \log(2)/2 \)-carrying weak bonds per two sites, giving exactly the \( \log 2 \) value of entanglement associated with the twofold degeneracy also found per two sites. Thus for the iTEBD ground state on the tree, unlike for the unique ground state on the planar honeycomb, the entanglement entropy on various bonds is continuous in the Toric Code limit \( a \to 1 \), with weak bonds carrying vanishing entanglement like for the disjointed singlets Hamiltonian \( a = 1 \).

The finite bond dimension \( \chi \) entanglement cutoff of the iTEBD algorithm is likely playing the key role here, collapsing the extensive degeneracy of the gauge theory on the tree into a particular minimally entangled state which is then chosen by iTEBD as the ground state. It will be interesting to explore whether this mechanism, of a ground state selected from an extensive degenerate manifold through a minimal-entanglement constraint, changes its role as the bond dimension increases.

Armed with the understanding of these subtleties, we thus find that aside from some spurious spontaneous symmetry breaking due to the infinite system size explored by the iTEBD algorithm, the entanglement entropy of the resulting iTEBD QSL ground state (as well as its energy and correlators) exhibit close agreement with these predictions of the exact QSL solution on a finite tree, as shown in Figs. \(^{12,18}\)

\section*{VI. CONCLUSION}

In this work we have discussed three-dimensional honeycomb-like crystal structure for iridates with edge sharing \( \text{IrO}_6 \) octahedra, including the recently synthesized\(^{11}\) 3D harmonic honeycomb \( \text{Li}_2\text{IrO}_3 \). In this crystal the iridium ions form a 3D \( \mathcal{H}-1 \) “harmonic honeycomb” lattice, whose local threefold coordination and hexagon plaquettes are identical to those of the 2D honeycomb lattice. Effective spin 1/2 moments arise on the \( \text{Ir}^{4+} \) as the Kramer’s doublet set by the octahedral crystal fields and iridium’s strong spin-orbit coupling. We model magnetic interactions using a minimal Hamiltonian with Heisenberg \( (J) \) and Kitaev \( (K) \) couplings, which are expected to dominate in the limit of ideal octahedra. Anisotropy in the strength of couplings between \( z \) bonds and the \( x, y \) bonds is expected from the crystal symmetries, and enables a fit to the experimental susceptibility measurements, which suggests strong Kitaev exchange. We discuss an exact solution to the pure Kitaev model in 3D, some of whose most interesting features are unique to three dimensionality. These features include the extended magnetic flux loop excitations as well as the existence of a finite temperature deconfined phase, neither of which can occur in the 2D honeycomb model.

We compute the phase diagram upon including the Heisenberg term, as a function of both the relative strength of \( J \) to \( K \) and the Kitaev coupling bond anisotropy parameter. Our approximation of choice is to study this system on the Bethe lattice, the tree with no closed loops. This is expected to capture the basic physics on the hyperhoneycomb lattice and its related 3D harmonic honeycomb lattices, due to the long length of their shortest closed loop \( (\ell = 10 \text{ for } \mathcal{H}-0 \text{ or } \ell = 6, 14 \text{ for } \mathcal{H}-1) \). This large-\( \ell \) approximation admits no analytical solution, but rather is numerically tractable via a class of entanglement-based algorithms. We use a TPS representation of the ground state, which is then determined using the iTEBD algorithm directly in the thermodynamic limit. Both the magnetically ordered phases as well as the gapped quantum spin liquid phases are obtained using this technique. We study the exact solution on the tree to analytically compute physical properties and entanglement entropies of the majorana fermion sector and the \( \mathbb{Z}_2 \) gauge theory sector of the spin liquid on the Bethe lattice. Comparing to the entanglement from iTEBD gives a clear positive signature of the quantum spin liquid phases captured by the iTEBD numerics. The exact 3D quantum spin liquid together with this large-\( \ell \) approximation provide a controlled study of 3D fractionalization. Although experimentally the 3D harmonic honeycomb \( \text{Li}_2\text{IrO}_3 \) appears to be magnetically ordered\(^{10}\), the significant Kitaev couplings indicated by experiments are promising, and suggest future directions to realize QSLs by tuning magnetic interactions via pressure or chemical composition.

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APPENDICES

Appendix A: Entanglement entropy from the majorana fermions on the tree

At the exact QSL point we wish compute the entanglement entropy (and the energy) for the ground state on the tree, in order to compare this exact result to the iTEBD computation. Within the gapped phase of the pure Kitaev (anisotropic) Hamiltonian, the system can be exactly mapped to a free majorana fermion problem with a gapped spectrum. We can thus compute quantities on finite trees independently of the iTEBD algorithm, within the majorana fermion mapping. Computing energies is straightforward and we find convergence to the thermodynamic limit using the boundary-eliminating procedure described above on trees with up to 9 layers. To describe the entanglement entropy results, let us first recall the computation of entanglement spectrum and entropy for free fermion systems. Operating on a bond-centered finite tree, we compute the correlation function by occupying half of the majorana spectrum. The reduced correlation function associated with a cut through the central bond is found by restricting the site indices of the correlator to lie within one of the two partitions. Each eigenvalue \( c_i \) of the reduced particle correlator also has an associated hole eigenvalue \( 1 - c_i \). The entanglement entropy of the bipartition can be computed from the particle and hole correlators, with a factor of \( 1/2 \) for majoranas, as \( S_E = (1/2) \sum_i [c_i \log c_i + (1 - c_i) \log (1 - c_i)] \).

To eliminate tree finite size effects for computing the entanglement entropy in the fermion sector of the spin liquid, we use two approaches. In the first approach, we carefully determine which of eigenvalues of the open tree adjacency matrix are associated with the bulk, using the counting procedure described above, and keep only the eigenstates associated with these eigenvalues when computing the correlation function for the entanglement bipartition. In the second approach, we subtract the \( T = 0 \) thermal entropy of finite \( L \)-layered trees (with open boundary conditions) from the reduced density matrix entanglement entropy of each such tree under a bipartition through the center bond. This difference gives purely the entanglement entropy associated with the single bond cut, without the thermal entropy of the numerous zero modes of the boundary. We find agreement between the two approaches as the system size is increased (and the isolated boundary eigenvalue of the bond-centered tree vanishes).

Appendix B: Mean field for Stripy-X order

We briefly recall the self consistency equation for the mean field moment, \( 2|\vec{m}_i| = \tanh \left( \frac{\vec{B}_i}{(2T)} \right) \) and \( \vec{m}_i = \vec{B}_i \), where \( \vec{B}_i \) is the mean field coupling to spin \( S_i \). Above the transition temperature the mean field produces the Curie-Weiss law, \( \chi^{rr} = \left( g^2 \mu_B^2 / (4T + \sum_j J_{ij}^{rr}) \right) \); the experimentally relevant units may be restored by noting that \( \mu_B = 0.672 \) kelvin/tesla. In the stripy-x order, the classical mean field \( \vec{B}_i \) takes the form

\[
\vec{B}_i = g\mu_B \vec{B} - (K_{d,m}^z \hat{x} + K_{d,m}^y \hat{y}) + K_{c,m}^{z[i]} \hat{z} + J_{c} \vec{m}_{n[i]} + J_{d} \vec{m}_i
\]

and every site carries one of two magnetizations, \( \vec{m}_i \) or \( \vec{m}_{n[i]} \).

Appendix C: Comparison of magnetic transitions in iTEBD and mean field theory

Magnetic phases can be approximately described within a classical mean field theory. Such classical product states over sites, with no quantum fluctuation or entanglement, correspond to tensor product states with bond dimension \( \chi = 1 \). On the Bethe lattice, we have captured the magnetically ordered phases using tensor product states with various \( \chi \). We find that increasing \( \chi \) to a value as low as \( \chi = 4 \) is sufficient for capturing most of the quantum fluctuations near a first order transition between adjacent magnetic phases. The location in parameter space of these transitions can be compared to the classical transition point. Classically, the transition occurs at \( \phi = n\pi - \arccot[2 + a] \), for anisotropy \( a \), with \( n = 1 \) for the zigzag-FM transition and \( n = 2 \) for the striped-Neel transition. This comparison is shown in Fig. 15. For concreteness, we also draw sample magnetic configurations on the hyperhoneycomb lattice, shown in Fig. 16.
FIG. 14. Gapped spin liquid for $K > 0$ and surrounding magnetic phases within iTEBD. See the description in Fig. 11. Here we show the QSL at $a = 1/2$, $K > 0$, which competes with the zigzag and Neel orders. Different entropy curves occur here compared to the $K < 0$ QSL since while both the FM and stripy orders are effectively ferromagnets with nearly saturated ordered moments, the Neel and zigzag phases involve substantial quantum fluctuations.

Appendix D: Harmonic honeycomb series

In the notation for the $n$-harmonic honeycomb lattice, the integer $n$ counts the number of hexagons forming the width of each fixed-orientation planar strip. Or equivalently, going along the direction of the special $c$ axis, the integer $n + 1$ specifies the number of $z$-bonds between switches of the $x,y$ bonds orientation. Odd-$n$ lattices possess a mirror plane perpendicular to the special axis, slicing through the midpoint of the (odd number of) hexagons; even-$n$ lattices possess no mirror reflections, only glide planes. In this manuscript we focus on two lattices: the $n = 1 \ H^{-1}$ lattice, space group $Cccm$, recently synthesized as a polytype of $\text{Li}_2\text{IrO}_3$, with $\ell = 6$ hexagon as well as $\ell = 14$ sized minimal loops; and the $n = 0$ hyperhoneycomb or $\ H^{-0}$ lattice, space group $Fddd$, with $\ell = 10$ decagon minimal loops. The two lattices are shown in Figures 1 and 5 respectively.

The “hyperhoneycomb” terminology for the $\ H^{-0}$ lattice may be understood through the following connection to the hyperkagome lattice (also related to the hyperoctagon lattice). Consider the 2D kagome and honeycomb lattices. The kagome is the medial lattice — formed by connecting bond midpoints – of the honeycomb lattice. This relation naturally suggests the existence of 3D honeycomb-like lattices which can be similarly associated with the 3D hyperkagome, namely the three dimensional lattice of corner-sharing triangles formed by iridium ions in $\text{Na}_4\text{Ir}_3\text{O}_8$. Indeed, the medial lattice of the $\ H^{-0}$ lattice has a graph (or, an adjacency matrix) which is locally identical to that of the hyperkagome: both feature corner-sharing triangles which combine to form $\ell = 10$ decagon loops. These decagons arise from the $\ell = 10$ minimal loops of the $\ H^{-0}$ lattice.

Appendix E: Coordinates for the $\ H^{-0}$ and $\ H^{-1}$ harmonic honeycomb lattices

We will write vectors in the Cartesian coordinate system defined by the axes from an iridium to its neighboring oxygen ions. We use units in which these Ir-O bonds have length 1. Nearest neighbor bonds in the resulting Ir lattice then have length $\sqrt{2}$.

Consider the symmetric octahedra idealization of an
iridate lattice, for which the Kitaev Hamiltonian is most likely to be relevant. For the $\mathcal{H}$-0 and $\mathcal{H}$-1 lattices, the parent simple orthorhombic crystallographic vectors can be taken to be

$$a = (2, 2, 0), \quad b = (0, 0, 4), \quad c = (6, -6, 0). \quad (E1)$$

1. $\mathcal{H}$-0 lattice (space group $Fddd$)

The hyperhoneycomb lattice is face centered orthorhombic, space group number 70, i.e. $Fddd$. To connect to the symmetry discussion in the main text, we may use the basis vectors of the parent simple orthorhombic lattice, $a, b, c$. The points in the unit cell may be given as coefficients of the three vectors $a, b, c$. Together with an overall factor of 1/24, these are

$$(3, 3, 5); (9, -3, 11); (21, -3, 19); (15, 3, 13). \quad (E2)$$

2. $\mathcal{H}$-1 lattice (space group $Cccm$)

The $\mathcal{H}$-1 lattice has space group #66, $Cccm$. Its Bravais lattice is base-centered orthorhombic. To describe the position of the sites, let us again use the basis vectors $a, b, c$ of the parent simple orthorhombic lattice. The points in the unit cell may be given as coefficients of these three vectors, by

$$(0, 0, 0); (0, 0, 1/6); (1/2, 0, 1/4); (1/2, 0, 5/12); \quad (E6)$$

$$(0, 0, 1/2); (0, 0, 2/3); (0, 1/2, 3/4); (0, 1/2, 11/12). \quad (E6)$$
These are the eight points in the base-centered (reduced) unit cell; the other eight points in the unit cell of the parent simple-orthorhombic lattice are found by adding $(1/2, 1/2, 0)$ to these points.

Appendix F: Lattice tight-binding model and Majorana spectrum

In the Kitaev spin liquid at its exactly solvable parameter point, the emergent Majorana Fermion hops within the nearest-neighbor tight binding model on the lattice. Its band structure (fixed to half filling) is given by the nearest-neighbor tight binding model on the lattice. For the $\mathcal{H}$-1 (Cccm) lattice, this matrix is

$$K \times \begin{pmatrix}
0 & 1 & 0 & 0 & 0 & 0 & V_3 U_3^* \\
1 & 0 & V_1^* & 0 & 0 & 0 & 0 \\
0 & V_1 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 & V_1 & 0 & 0 \\
0 & 0 & 0 & V_1^* & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & V_2 \\
V_2 U_3^* & 0 & 0 & 0 & 0 & 1 & 0
\end{pmatrix}$$  \hspace{1cm} (F1)

where $U_3 = \exp [i \mathbf{k} \cdot c]$, $V_{1,2} = 1 + U_{1,2}$ and $U_{1,2} = \exp [i (\mathbf{k} \cdot (\mathbf{a} \pm \mathbf{b})/2)]$ with the $+/-$ sign corresponding to $U_1, U_2$ respectively. The zeros of the spectrum appear at the contour of momenta characterised by the two equations $\mathbf{k} \cdot \mathbf{c} = 0$ and $\cos (\mathbf{k} \cdot \mathbf{a}/2) + \cos (\mathbf{k} \cdot \mathbf{b}/2) = 1/2$.

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