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Robust non-Abelian spin liquid and possible intermediate phase in antiferromagnetic Kitaev model under magnetic field

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We investigate the non-Abelian topological chiral spin liquid phase in the two-dimensional (2D) Kitaev honeycomb model subject to a magnetic field. By combining density matrix renormalization group (DMRG) and exact diagonalization (ED) we study the energy spectra, entanglement, topological degeneracy, and expectation values of Wilson loop operators, allowing for robust characterization. While the ferromagnetic (FM) Kitaev spin liquid is already destroyed by a weak magnetic field with Zeeman energy $H_*^{\text{FM}} \approx 0.02$, the antiferromagnetic (AFM) spin liquid remains robust up to a magnetic field that is an order of magnitude larger, $H_*^{\text{AFM}} \approx 0.2$. Interestingly, for larger fields $H_*^{\text{AFM}} < H < H_{**}^{\text{AFM}}$, an intermediate gapless phase is observed, before a second transition to the high-field partially-polarized paramagnet. We attribute this rich phase diagram, and the remarkable stability of the chiral topological phase in the AFM Kitaev model, to the interplay of strong spin-orbit coupling and frustration enhanced by the magnetic field. Our findings suggest relevance to recent experiments on RuCl₃ under magnetic fields.

Introduction. The search for highly entangled quantum states of matter such as quantum spin liquids (QSLs) has intensified in recent years ¹⁻⁴. The peculiarity of QSLs lies not only in the absence of magnetic long-rang order even at zero temperature, but more importantly in exhibiting fractionalized excitations and topological ground state degeneracy. Among various theoretically proposed QSLs, a remarkable example is the Kitaev model of spins with nearest-neighbor interactions on the two-dimensional (2D) honeycomb lattice⁵. This model is solved exactly by mapping it into a model of Majorana fermions coupled to an emergent static \mathbb{Z}_2 gauge field. The ground state is either a gapless spin liquid or, with weak time reversal breaking, a gapped spin liquid phase. The latter harbors a non-Abelian anyon known as an Ising anyon, a descendant of vortices in two-dimensional p + ip superconductors⁶. The exact solution of the apparently simple Kitaev model has motivated a search for the physical realization of non-Abelian OSL⁷⁻⁹.

The highly anisotropic and spatially dependent spin interaction in the Kitaev model can conceivably appear in Mott insulators with strong spin-orbit coupling and j = 1/2 local moments. In particular Jackeli and Khaliullin¹⁰ proposed a mechanism for Kitaev interaction in transition metal oxides with edge-sharing oxygen octahedra. By now, in addition to various three-dimensional compounds^{11–13}, a variety of two dimensional layered honeycomb lattice magnets¹⁴ have been discovered, including Na₂IrO₃^{15–17}, α -Li₂IrO₃^{16,17}, a hydrated variant H₃LiIr₂O₆¹⁸, and RuCl₃¹⁹.

Aside from the spin liquid candidates $Na_4Ir_3O_8$ (hyperkagome^{11,20}) and $H_3LiIr_2O_6$ (honeycomb¹⁸), these compounds are magnetically ordered at sufficiently low temperatures, indicating the presence of additional spin interactions beyond the Kitaev model. Nonetheless, various experimental and theoretical works suggest the magnetic ordered states are proximate to a spin-liquid phase^{9,21–32}. To understand the nature of quantum phases realized in materials, it is helpful to compare experimental findings with expected signatures of perturbed Kitaev Hamiltonians. However,even with the large body of available experimental data, the sign

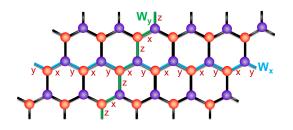


Fig. 1: (Color online) The honeycomb lattice and Wilson loop operators. The honeycomb lattice is spanned by unit vectors (1,0) and $(1/2, \sqrt{3}/2)$ with lattice size $N = L_x \times L_y \times 2$. Green and blue loops denote Wilson loop operators along vertical and horizontal periodic boundary conditions on the torus, respectively.

of the Kitaev interaction in the honeycomb magnets remains an open question^{25–31,33–43}. In models with strong spin orbit coupling, the Curie-Weiss temperature may not reflect the dominant interaction due to cancellation among the various spin-orbit-coupled exchanges; for instance T_{CW} may even vanish⁴⁴. Interestingly for RuCl₃ it has recently been argued that natural models with nearest-neighbor exchanges involve strong Γ exchange or have dominant likely *antiferromagnetic* Kitaev exchange^{26,27,45}.

For the pure Kitaev model, the different signs of the Kitaev exchange are related by a sublattice dependent transformation, leading to identical energy spectrum. However, under an external magnetic field, ferromagnetic (FM) and antiferromagnetic (AFM) Kitaev models are expected to behave differently. Previous theoretical studies mainly focused on the FM Kitaev model, and found that the non-Abelian spin liquid phase only survives up to a very small magnetic field $H_*^{\rm FM} \approx 0.02$ by Jiang *et al.*⁴⁶. To our knowledge, except semiclassically⁴⁷, the AFM Kitaev model in a magnetic field has not yet been studied.

In this Letter, we study the AFM Kitaev model in a magnetic field by performing extensive exact diagonalization (ED) and density matrix renormalization group (DMRG) simulations. The energy spectra, the expectation value of Wilson

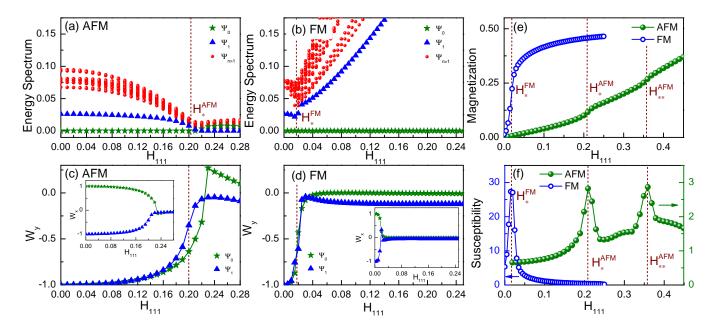


Fig. 2: (Color online) Panels (a, b): For the AFM (a) and FM (b) Kitaev models in a magnetic field, the pair of topological ground states (approximately degenerate on this $N = 4 \times 3 \times 2$ torus) are separated from higher energy states by an energy gap, within the topological phase $H < H_*$ where $H_*^{\text{AFM}} \approx 0.2$ and $H_*^{\text{FM}} \approx 0.02$. Panels (c, d): Wilson loop operators W_y (main panel) and W_x (inset) for the two lowest energy states, for AFM (c) and FM (d) models. The two states have $W_y = -1$ but are distinguished by $W_x = \pm 1$. Panel (e): The magnetization curves show the transitions and the stark difference between the AFM and FM models. Panel (f): The second order derivative of ground state energy with respect to field, or equivalently the magnetic susceptibility: note the difference in magnitudes between AFM and FM models. In the AFM case, the transition to the polarized high-field phase is achieved only at a second peak with $H_{**} \approx 0.36$.

loop operator and the ground state degeneracy as a function of the magnetic field are computed and compared with exact analytical results at zero field. We find the presence of the non-Abelian QSL phase in the AFM Kitaev model over a wide range of magnetic field up to $H_*^{\rm AFM} \approx 0.2$, an order-of-magnitude larger than that of the FM Kitaev model. Moreover, before a second transition at $H_{**}^{\rm AFM} \approx 0.36$ to the high-field partially-polarized paramagnet , an intermediate gapless phase is observed for fields $H_*^{\rm AFM} < H < H_{**}^{\rm AFM}$.

Model and Method.—We consider the Kitaev honeycomb model subject to an external magnetic field **H** along the $\langle 111 \rangle$ direction. The Hamiltonian is given by

$$H = \sum_{\langle i,j \rangle} K_{\gamma} S_i^{\gamma} S_j^{\gamma} - \sum_i \mathbf{H} \cdot \mathbf{S}_i.$$
(1)

Here, $\gamma = x, y, z$ denote the three distinct nearest neighbor links $\langle i, j \rangle$ of the hexagonal lattice [see Fig. 1], S^{γ} represents effective spin-1/2 degrees of freedom sitting on each vertex and interacting via exchange K_{γ} . The ground state at $\mathbf{H} = 0$ corresponds to the Kitaev limit, which exhibits two kinds of QSLs depending on the relative coupling strength. When one of the three coupling K_{γ} is much larger than the others, the system is a gapped \mathbb{Z}_2 spin liquid with Abelian excitations, while around the isotropic point of equal couplings, the system is a gapless spin liquid⁵. The latter can turn into a non-Abelian topological phase under time-reversal symmetry breaking perturbations⁵, e.g., by adding a three-spin chirality term⁴⁸ or by applying an external magnetic field⁵ or by decorating the honeycomb lattice⁴⁹.

We use both exact diagonalization (ED) and density matrix renormalization group (DMRG) to study the Hamiltonian (1) with isotropic coupling $K_{\gamma} \equiv K$, as a function of an external magnetic field **H**. We compare the phase diagrams with AFM (K > 0) and FM (K < 0) Kitaev couplings.

In the present calculation, we consider a system of size $N = L_x \times L_y \times 2$ [see Fig. 1], where L_x and L_y represent the number of unit cells along x and y directions, respectively. Our present DMRG calculations keep enough states to ensure the truncation error of the order or smaller than 10^{-9} and perform DMRG sweeps until the measured quantities are converged.

Non-Abelian topological phase.—We first compute the energy spectra of the model Hamiltonian Eq. (1) as a function of the magnetic field. Figure 2 shows the low-energy spectra in different momentum sectors for a system size $N = 4 \times 3 \times 2$ on the torus, with antiferromagnetic (K = +1) or ferromagnetic (K = -1) Kitaev couplings. For the antiferromagnetic case, we find two lowest energy states in $(\pi, 0)$ and (0, 0) momentum sectors, which are separated from the higher energy states by a finite gap for a range of magnetic field $0 \le H_{111} \le 0.2$. In contrast, in the case with ferromagnetic Kitaev coupling, the spectra shown in Fig. 2 (b) indicates that the topological phase only survives in a much smaller regime at $H_{111} \le 0.02$. Meanwhile, while naively one would expect a transition directly to the partially-polarized phase (which is smoothly con-

nected to the fully polarized $H_{111} = \infty$ limit), as is indeed seen in the FM Kitaev model [see Fig. 2 (b), (e) and (f)], here for the AFM Kitaev model, as shown in Fig. 2 (a),(e) and (f), an intermediate gapless phase (discussed further below) is observed at $H_*^{\text{AFM}} < H < H_{**}^{\text{AFM}}$ before a transition to polarized paramagnet at $H_{**}^{\text{AFM}} \approx 0.36$. In both AFM and FM cases, the critical field is also identified by sharp peaks in the second order derivative of the ground state energy or equivalently the magnetic susceptibility[see Fig. 2 (f)]. Similarly to the FM case⁴⁶, the field-driven phase transitions in the AFM case might be continuous or weakly first-order.

We now demonstrate the topological nature of the two lowest-energy states below the critical field H_* . First, we note that the Kitaev model at zero field with periodic boundary conditions has topological ground state degeneracy in two dimensions. Different ground states are characterized by two Wilson loop operators W_y and W_x associated with non-contractible loops along y and x directions respectively. As illustrated in Fig. 1, the definitions of W_y and W_x are given by

$$W_y = -\left\langle \prod_{i=1}^{2L_y} \sigma_i^y \right\rangle; W_x = -\left\langle \prod_{i=1}^{2L_x} \sigma_i^z \right\rangle.$$
(2)

where σ^y and σ^z are Pauli matrices⁵⁰, i.e. twice the spin–1/2 operators. The loops along y direction only cover $\gamma = x, z$ links while the loops along x direction only cover $\gamma = x, y$ links. It is straightforward to verify that these Wilson loop operators commute with each other and also with the Hamiltonian in the Kitaev limit. Each operator squares to identity, hence its eigenvalue is either +1 or -1. The ±1 eigenvalue of Wilson loop operator corresponds to the Z_2 fluxes or equivalently the periodic/antiperiodic boundary conditions for the emergent Majorana fermions.

The expectation values of Wilson loop operators $W_{x,y}$ are measured for these two lowest energy states in the model (1). As shown in Fig. 2 (c) and (d), these Wilson loop operators take exact quantized values in Kitaev limit and nearly quantized values for a finite range of magnetic fields below the critical value. This indicates that the emergent \mathbb{Z}_2 gauge theory remains a good description of perturbed Kitaev model away from static limit. Importantly, below the critical field, the two lowest energy states have nearly the same value of $W_y \simeq -1$ but distinct values of W_x , with $W_x \simeq +1$ for the state in momentum $(\pi, 0)$ sector and $W_x \simeq -1$ for the state in (0,0) sector, as shown in the inset of Fig. 2 (c). These results are fully consistent with our expectation that the degeneracy between different topological sectors of the Kitaev phase in thermodynamic limit is split by finite size effect in a quasione-dimensional geometry. For a three-leg system studied in this work, the splitting between $W_y = 1$ and $W_y = -1$ sectors is strong enough that the two lowest energy states both have $W_u \simeq -1$. As we shall show below, these two lowest energy states become degenerate as L_x increases. Their many-body momenta $k_x = 0$ and π indicate that as a onedimensional system the three-leg AFM Kitaev model spontaneously breaks translational symmetry breaking and doubles the unit cell in thermodynamic limit. This is analogous to the

charge-density-wave states obtained by placing $\nu = 1/3$ fractional quantum Hall states on a thin torus.

The expectation values of Wilson loop operators decay rapidly near the critical field and becomes negligible above the critical field. The near quantization of Wilson loop operators (and its lack of) provide another strong evidence for the topological (non-topological) nature of the phases before (after) the phase transition.

For the AFM Kitaev model, we further use DMRG to calculate the ground state degeneracy at $H_{111} \lesssim 0.2$ for large L_x to confirm its topological nature. In Fig. 2 (a), we find a small energy split between two lowest energy states. To confirm these two states are exactly degenerate states in the thermodynamic limit, we perform DMRG calculation on torus by targeting three lowest energy states with increasing L_x . As shown in Fig. 3 (a), we find that the energy difference between two lowest states $E_1 - E_0$ becomes vanishingly small when the system length $L_x\gtrsim 8$, while the lowest two states are separated from higher energy states by a finite gap indicated by $E_2 - E_0$ [see Fig. 3 (a)]. Based on these calculation, the two-fold ground state degeneracy of such topological phase is identified. Meanwhile, we also checked the Wilson loop operator W_y for different $L_y = 3$ system size by DMRG, as shown in Fig. 3 (b), we find the topological phase is very robust and independent of system length.

The gapped feature of the topological phase can also be confirmed by the Von Neumann entanglement entropy $S_{\rm VN}$ defined by $S_{\rm VN} = -Tr (\rho_A \ln \rho_A)$, where ρ_A is the reduced density matrix of part A for the bipartition of the system into A and B, ρ_A is got by tracing out the degrees of freedom of B part. Here, we consider the cut parallel to y direction and measure the value of $S_{\rm VN}$ for each cut at L_A . For the gapped state, the Von Neumann entropy should be independent on the positions of each cut and display flat behavior. As shown in Fig. 4 (b), we calculate a long cylinder by DMRG and find the flat $S_{\rm VN}$ as a function of L_A , implying the existence of the well defined gap in the topological phase. All of these confirm the stability of the topological phase.

In the absence of magnetic field, the Kitaev model is exactly solvable in terms of static fluxes and Majorana fermions⁵. We also analyze the exact solution on finite systems as well as infinite ladders. In each topological sector defined by a particular set of values of the Wilson loop operators, the ground state energy is simply given by the energy of the filled fermi sea, i.e. the sum of all negative Majorana eigenvalues. Figure 4 (a) shows the Majorana dispersion for infinite cylinders in $W_y = -1$ sector with fixed width $L_y = 3$. We also compared the exact solution on finite-sized systems with DMRG and ED results, which are consistent with each other.

Interestingly, the three-leg system with $W_y = -1$ as we identified here is a one-dimensional topological superconductor of Majorana fermions in the thermodynamic limit $L_x \rightarrow \infty$. This is shown by computing sgn[Pf[H[0]]Pf[H[π]]], i.e. the sign of the product of Pfaffians of the quadratic Majorana Hamiltonian matrices at 1D momenta k = 0 and $k = \pi$. We find a negative value for this topological index, correspond to a 1D topological superconductor⁵¹. Therefore, we expect the presence of boundary Majorana zero modes for open bound-

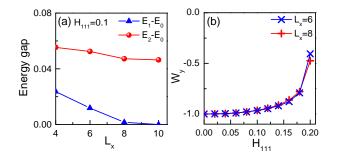


Fig. 3: (Color online) (a) The DMRG calculation of the lowest three energy states as function of L_x at $H_{111} = 0.1$ on torus. The energy difference between two lowest energy sates becomes vanishingly small with increasing system size while they are separated from higher energy sectors by a finite gap. (b)The DMRG results of the Wilson loop operator W_y on torus for the antiferromagnetic (AFM) Kitaev model with $L_x = 6, 8$.

ary conditions in the L_x direction. These boundary zero modes can be regarded as a descendent of non-Abelian anyons in the Kitaev phase in two dimensions, and their presence should be robust against perturbations such as the magnetic field. Indeed, we find the lowest two states are exactly degenerate on cylinders by DMRG, as shown in the Fig. 4 (b) for $H_{111} = 0.1$, and the two-fold degeneracy in the entanglement spectrum on cylinders, confirming the existence of Majorana zero modes on the boundary.

Discussion and Summary.—In this letter, we report a robust non-Abelian phase in the antiferromagnetic Kitaev model under magnetic field. Based on extensive DMRG and ED simulations, we identify its topological features by the energy spectra, entanglement, topological degeneracy, and Wilson loop operators. We find that the topological phase in the antiferrmagnetic Kitaev model is much more stable to increasing magnetic field than the one in ferromagnetic Kitaev model. This can be partially understood from the low field magnitude of magnetic susceptibility (Fig. 2 (a,b) insets), which in turn have a simple interpretation. While at zero field the AFM and FM Kitaev models are exactly equivalent by a majorana sign transformation on one honeycomb sublattice⁵, since their spin correlations are identical except opposite in sign, the FM Kitaev model is nearly a ferromagnet, while the AFM model has similar strong response to a staggered magnetic field but a weak response to a uniform field. This difference between the AFM and FM Kitaev coupling can also be seen approaching from the infinite field limit⁴⁷ based on semiclassical spin wave analysis. Our findings suggest that, in materials with dominant antiferromagnetic Kitaev interactions, a spin liquid phase if present may be observable under application of fairly

substantial magnetic fields, in contrast to previous expectations.

Moreover, when the gapped chiral topological order is destroyed by the large field in the AFM case, before entering the polarized phase an intermediate gapless phase is found. Within the intermediate gapless phase of the AFM model, the DMRG algorithm converges to a state exhibiting mod-

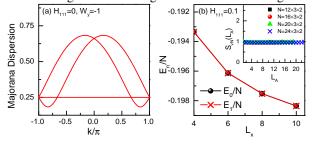


Fig. 4: (Color online) (a)Majorana dispersions on infinite ladders $(L_y = 3)$ for $W_y = -1$ sector at $H_{111} = 0$, which show finite gap and quantized value of Wilson loop operators. These are consistent with the numerics for finite fields (see main text). (b) The energy density of two degenerate states on cylinders at $H_{111} = 0.1$, the inset shows the Von Neumann entanglement entropy for long cylinders, where the flat feature indicates the existence of the finite gap.

ulations in spin density around the partially-polarized mean (about $10\% \sim 20\%$ of full amplitude), which appear to be pinned by the open boundaries (see Supplementary Material⁵² for details). Together with the gapless spectrum [Fig. 2 (a)] and the large entanglement, these observations serve as evidence that this gapless phase involves long range correlations or entanglement, and thus it cannot be captured reliably in the 2D limit. A possible connection to experiments remains an open question.

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