

Possible proximity of the Mott insulating iridate Na_2IrO_3 to a topological phase: Phase diagram of the Heisenberg-Kitaev model in a magnetic field

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Motivated by the recent experimental observation of a Mott insulating state for the layered iridate Na_2IrO_3 , we discuss possible ordering states of the effective iridium moments in the presence of strong spin-orbit coupling and a magnetic field. For a field pointing in the $\langle 111 \rangle$ direction—perpendicular to the hexagonal lattice formed by the iridium moments—we find that a combination of Heisenberg and Kitaev exchange interactions gives rise to a rich phase diagram with both symmetry breaking magnetically ordered phases as well as a topologically ordered phase that is stable over a small range of coupling parameters. Our numerical simulations further indicate two exotic critical points at the boundaries between these ordered phases.

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I. INTRODUCTION

In the realm of condensed matter physics, spin-orbit coupling has long been considered a residual, relativistic correction of minor relevance to the macroscopic properties of a material. In recent years this perspective has dramatically changed, especially due to the theoretical prediction and subsequent experimental observation of fundamentally new states of quantum matter, so-called topological insulators,¹ that are solely due to the effect of spin-orbit coupling. The topological insulators experimentally realized so far are semiconductors, whose physical properties can be largely captured by band theory of noninteracting electrons. It is an interesting challenge, for both theory and experiment, to identify an even broader class of materials where this physics plays out even in the presence of interactions and strong correlations.² Good candidate materials for the latter are the iridates.^{3,4} These $5d$ transition metal oxides are prone to exhibit electronic correlations and form (weak) Mott insulators, while the relatively large mass of the iridium ions ($Z = 77$) gives rise to a comparably strong spin-orbit coupling, which has been found to be as large as $\lambda \approx 400$ meV.⁵ The most common valence of the iridium ions in these materials is Ir^{4+} . The d orbitals of this $5d^5$ configuration are typically split by the surrounding crystal field, and for the octahedral geometry of the IrO_6 oxygen cage, result in an orbital configuration where five electrons occupy the lowered, threefold degenerate t_{2g} level. Spin-orbit coupling will further lift this degeneracy of the t_{2g} orbitals and for strong coupling the effective $l = 1$ orbital angular momentum⁶ is combined with the $s = 1/2$ spin degree of freedom carried by the hole of this partially filled t_{2g} orbital configuration. This leaves us with two Kramers doublets of total angular momentum $j = 3/2$ and $j = 1/2$, of which the former is of lower energy and fully occupied by four electrons, while the partial filling of the latter gives rise to an effective spin-1/2 degree of freedom.

In this manuscript we focus on the iridate Na_2IrO_3 , in which NaIr_2O_6 slabs are stacked along the crystallographic c axis, and the Ir^{4+} ions in the layers form a hexagonal lattice.⁴ Recent measurements of the magnetic susceptibility provide evidence of effective spin-1/2 moments and magnetic

correlations below $T_N \approx 15$ K indicating that Na_2IrO_3 is indeed a Mott insulator.⁴ Theoretically, it has been argued^{7,8} that the interactions between the effective iridium moments in the Mott regime are captured by a combination of isotropic and highly anisotropic exchanges, which can be tracked back to the spin and orbital components of the effective momenta. A microscopic Hamiltonian interpolating between these two types of exchanges is given by

$$H_{\text{HK}} = (1 - \alpha) \sum_{\langle i, j \rangle} \vec{\sigma}_i \cdot \vec{\sigma}_j - 2\alpha \sum_{\gamma\text{-links}} \sigma_i^\gamma \sigma_j^\gamma, \quad (1)$$

where the σ_i denote the effective spin-1/2 moment of the Ir^{4+} ions, $\gamma = x, y, z$ indicates the three different links of the hexagonal lattice, and $0 \leq \alpha \leq 1$ parametrizes the relative coupling strength of the isotropic and anisotropic exchange between the moments.⁹ For $\alpha = 0$ the Hamiltonian reduces to the ordinary Heisenberg model, while in the opposite limit of highly anisotropic exchanges ($\alpha = 1$) the system corresponds to the Kitaev model.^{11,12} The latter is known to exhibit a gapless spin-liquid ground state (for equal coupling along the links) that can be gapped out into a topological phase with non-Abelian quasiparticle excitations by certain time-reversal symmetry breaking perturbations.¹¹ One such perturbation is a magnetic field pointing in the $\langle 111 \rangle$ direction, perpendicular to the honeycomb layer,

$$H_{\text{HK+h}} = H_{\text{HK}} - \sum_i \vec{h} \cdot \vec{\sigma}_i. \quad (2)$$

The main result of our manuscript is the rich phase diagram of this model, shown in Fig. 1. Besides two conventional, magnetically ordered phases we find a topologically ordered phase and two critical points, which we will discuss in detail in the remainder of the manuscript.

II. NUMERICAL SIMULATIONS

We determine the ground-state phase diagram of Hamiltonian (2) by extensive “quasi-2D” density-matrix renormalization group (DMRG)¹⁵ calculations on systems with up to $N = 64$ sites. In particular, we consider clusters of size

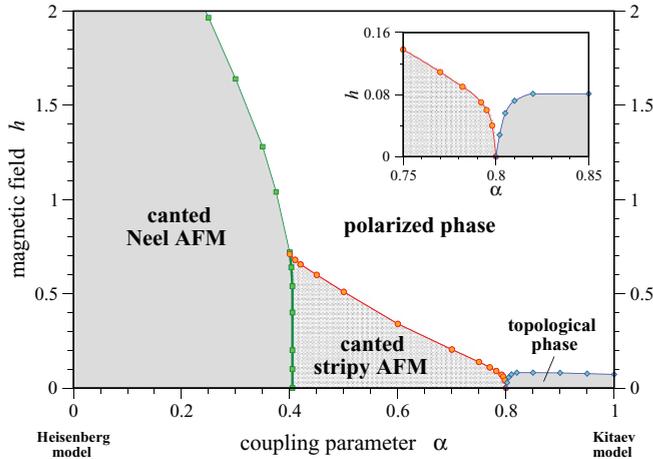


FIG. 1. (Color online) Ground-state phase diagram of the Heisenberg-Kitaev model (1) in a $\langle 111 \rangle$ magnetic field of strength h . Interpolating from the Heisenberg ($\alpha = 0$) to Kitaev ($\alpha = 1$) limit for small field strength, a sequence of three ordered phases is observed: a canted Néel state for $\alpha \lesssim 0.4$, a canted stripy Néel state illustrated in Fig. 2(c) for $0.4 \lesssim \alpha \lesssim 0.8$, and a topologically ordered state for nonvanishing field around the Kitaev limit. All ordered phases are destroyed for sufficiently large magnetic field giving way to a polarized state.

$N = 2 \times N_1 \times N_2$, which are spanned by multiples $N_1 \vec{a}_1$ and $N_2 \vec{a}_2$ of the unit cell vectors $\vec{a}_1 = (1, 0)$ and $\vec{a}_2 = (1/2, \sqrt{3}/2)$ as illustrated in Fig. 2. It should be noted that the numerical analysis of Hamiltonian (2) is a challenging endeavor, since not only the entire Hilbert space needs to be considered [due to the lack of $SU(2)$ invariance], but one also has to work with complex data types (due to the $\langle 111 \rangle$ orientation of the magnetic field). Our DMRG calculations keep up to $m = 2048$ states, which is found to give excellent convergence with truncation errors of less than 10^{-8} . We further use periodic boundary conditions in both lattice directions, which reduces finite-size effects. We have determined the phase boundaries in Fig. 1 by extensive scans of the ground-state energy, magnetization, and their derivatives in the (α, h) -parameter space.¹⁶

III. MAGNETICALLY ORDERED STATES

We start our discussion of the phase diagram shown in Fig. 1 by first recapitulating previous results⁸ for the Heisenberg-Kitaev model (1) in the absence of a magnetic field. Interpolating the relative coupling strength α between the isotropic Heisenberg limit ($\alpha = 0$) and the highly anisotropic Kitaev limit ($\alpha = 1$) a sequence of three phases has been observed:⁸ The Néel ordered state of the Heisenberg limit is stable for $\alpha \lesssim 0.4$, when it gives way to a “stripy” Néel ordered state illustrated in Fig. 2 which covers the coupling regime $0.4 \lesssim \alpha \lesssim 0.8$. In the extended parameter regime $0.8 \lesssim \alpha \leq 1$ the collective ground state is a gapless spin liquid. Near $\alpha = 1$, perturbation theory reveals that the gapless excitations of this phase are emergent Majorana fermions forming two Dirac cones in momentum space.

Including a magnetic field in the $\langle 111 \rangle$ direction a rich phase diagram evolves out of this sequence of three phases.

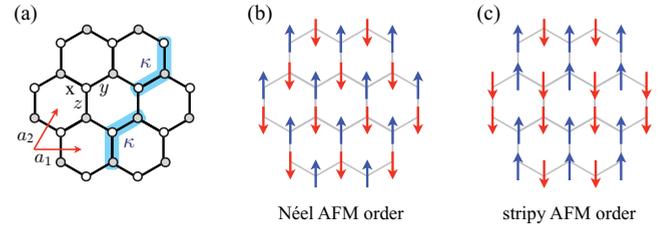


FIG. 2. (Color online) (a) The honeycomb lattice spanned by unit vectors $\vec{a}_1 = (1, 0)$ and $\vec{a}_2 = (1/2, \sqrt{3}/2)$. Illustration of magnetic states with (b) Néel order and (c) stripy Néel order.

For the magnetically ordered states we find that the orientation of the order in the Néel and stripy AFM phase cants along the $\langle 111 \rangle$ direction. To further characterize these canted states, it is helpful to analyze the independent symmetries of Hamiltonian (2). Besides the lattice translational symmetry T and a reflection symmetry I around the centers of the hexagons, there is an additional C_3^* symmetry, which is a combination of a threefold rotation around an arbitrary lattice site and a threefold spin rotation along the $\langle 111 \rangle$ spin axis.¹⁷ Both canted phases break a subset of these discrete symmetries of the Hamiltonian. The canted Néel order breaks the C_3^* and the I symmetries, which thus leads to a sixfold ground-state degeneracy in this phase. The canted stripy phase breaks both the C_3^* and translational symmetry (since the ordering pattern doubles the unit cell). As a consequence, we also find a sixfold ground-state degeneracy in this phase.¹⁸

For sufficiently large magnetic field, the order of both canted phases is destroyed and they give way to a simple polarized state. Our numerical simulations strongly suggest that the transitions between the polarized state and these canted states are continuous, which is in agreement with their spontaneous symmetry breaking. On the other hand, the transition between the two canted states at finite field strength (indicated by the bold line in Fig. 1) is found to be first order. In our simulations this is indicated by a sharp drop of the first derivative of the energy $dE/d\alpha$ as a function of the coupling parameter α across this transition—as shown in Fig. 3 for increasing strength of the magnetic field h . Approaching the endpoint of this first-order line around $h_c \simeq 0.7$ this drop smoothly vanishes, which indicates that this endpoint possibly is a tricritical point at which the two canted magnetically ordered phases and the polarized

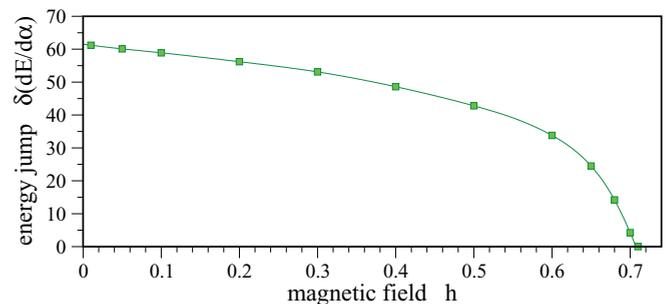


FIG. 3. (Color online) Energy jump along the first-order transition between the canted Néel and stripy AFM. Scans of the ground-state energy across this transition for various values of the magnetic field strength are provided in the supplemental material.¹⁹

phase meet. The existence of such a tricritical point can be understood within a Landau description with two distinct order parameters—corresponding to the discrete symmetry breaking of the two different magnetically ordered phases—opening a gap to magnon excitations.

IV. TOPOLOGICAL PHASE

We now turn to the spin liquid phase found for coupling parameters $0.8 \lesssim \alpha \leq 1$. For the Kitaev limit ($\alpha = 1$) it has previously been argued¹¹ that an infinitesimal field along the $\langle 111 \rangle$ direction will drive the system from the gapless spin liquid into a gapped non-Abelian topologically ordered phase. As we will discuss in the following, our numerical simulations allow us to confirm the existence of such a topologically ordered state for small magnetic field strengths not only in the Kitaev limit, but for the full extent of the gapless spin liquid phase, as indicated in the phase diagram of Fig. 1. We will further present an independent and nonperturbative way to determine the topological nature of this phase. This complements the original argument by Kitaev,¹¹ which was primarily based on a perturbation expansion showing that the leading order effect of a small magnetic field h is to introduce a topological mass term for the Majorana fermions—however, such a perturbative argument should be carefully tested when applied to a gapless state. For this purpose, we consider an additional three-spin exchange term κ , indicated by the blue bonds in Fig. 2(a), in our Hamiltonian,

$$H_{\text{HK}+h+\kappa} = H_{\text{HK}+h} - \kappa \sum_{ijk} \sigma_i^x \sigma_j^y \sigma_k^z. \quad (3)$$

In the Kitaev limit ($\alpha = 1$, $h = 0$) this Hamiltonian is exactly solvable in the same Majorana fermion representation used in the solution of the unperturbed Kitaev model.¹¹ In particular, one can prove that the three-spin exchange κ breaks time-reversal symmetry and gaps out the spin liquid phase into a topologically ordered state with non-Abelian excitations, so-called Ising anyons. To demonstrate that a small magnetic field in the $\langle 111 \rangle$ direction drives the system into the same phase, we have numerically calculated the phase diagram in the presence of both perturbations as shown in Fig. 4. The phase boundaries were again obtained by scanning the derivatives of ground-state energy and magnetization in the (h, κ) -parameter space. In particular, this phase diagram shows that one can adiabatically connect the phase for large κ and vanishing magnetic field with the phase for small, nonvanishing magnetic field and $\kappa = 0$, thus proving that the magnetic field gaps out the spin liquid into the same non-Abelian topological phase stabilized by the three-spin exchange. The only feature in the diagram is a single phase transition line which separates the topologically ordered state from the fully polarized state expected for large magnetic field strengths. For the Kitaev limit ($\kappa = 0$) this transition occurs for $h_c \simeq 0.072$. It is interesting to note that the critical field h_c initially grows with increasing κ , but then saturates to some finite value around $\kappa \gtrsim 6$. Physically, this saturation can be understood by the behavior of the gap for the Majorana fermions in the exact solution for $h = 0$. The dispersion of the Majorana fermion is given

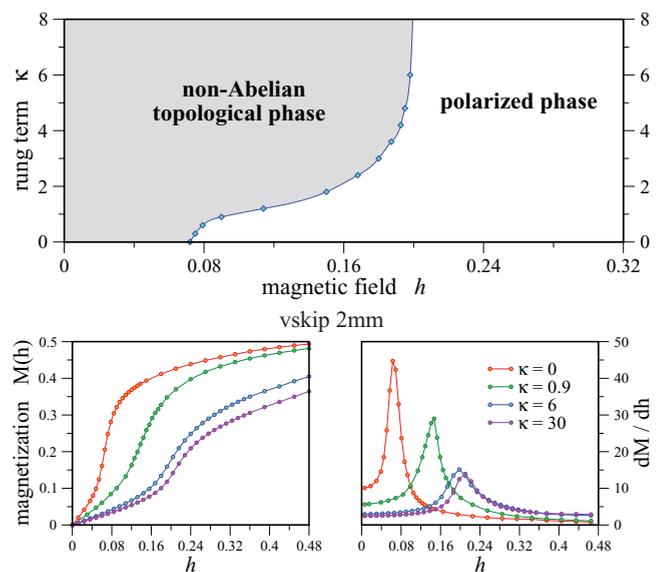


FIG. 4. (Color online) (Top panel) Ground-state phase diagram of the Kitaev model ($\alpha = 1$) in the $h - \kappa$ plane, where h is the strength of a magnetic field pointing in the $\langle 111 \rangle$ direction and κ is the strength of a time-reversal symmetry breaking three-site term. (Lower panels) Magnetization sweep and its derivative for various κ .

by $E_{\mathbf{k}} = 2\sqrt{|1 + e^{i\mathbf{k}\cdot\vec{a}_1} + e^{i\mathbf{k}\cdot\vec{a}_2}|^2 + \kappa^2 \sin^2(\mathbf{k}\cdot\vec{a}_1)}$. For small $\kappa \ll 1$, the Majorana fermion has a gap $E_g \simeq \sqrt{3}\kappa$. However, for large $\kappa \gg 1$ the gap of Majorana fermion remains finite and independent from κ , given by $E_g \simeq 2$. Since the magnetic field strength h_c required to destroy the topological phase is determined by the Majorana fermion gap at $h = 0$, the critical field h_c thus also increases and then saturates at large κ .

V. FIELD-DRIVEN TRANSITION OUT OF THE TOPOLOGICAL PHASE

We now return to the phase diagram of the Heisenberg-Kitaev model in Fig. 1 and focus on the transition between the topologically ordered state and the polarized state for large field strength. For the Kitaev limit ($\alpha = 1$) this transition occurs at a critical field strength of $h_c \approx 0.072$ and remains almost constant as the coupling parameter α is decreased. Interestingly, our numerics suggest that this field-driven phase transition might be continuous or weakly first order. In particular, we find that the second derivative of the ground-state energy $-d^2E/dh^2$ at this transition diverges with increasing system size, while the magnetization $M(h)$ does not show any discontinuity, as shown in Figs. 5(a) and 5(b), respectively.

While the limited system sizes in our study do not allow one to unambiguously determine the continuous nature of this field-driven phase transition, our numerics nevertheless provide some further insights what might cause such a continuous transition.²⁰ To this end, we plot the number of vortices in the ground state as a function of magnetic field, that is, the number of plaquettes with a nontrivial flux, in Fig. 5(c). Below the critical magnetic field (i.e., $h < h_c$), there are no vortices indicating a deconfined phase as expected in the presence of a vortex gap. At the phase transition, however, the

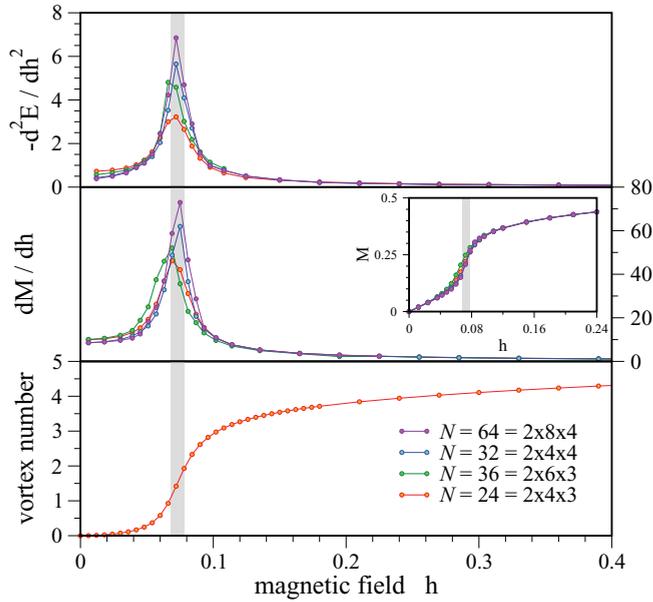


FIG. 5. (Color online) Phase transition for the Kitaev model ($\alpha = 1$) in a $\langle 111 \rangle$ magnetic field of strength h . (a) Second derivative of the ground-state energy $-d^2E/dh^2$, (b) magnetization $M(h)$ and its first derivative $dM(h)/dh$ for different system sizes. (c) Vortex number.

vortices appear to condense and the number of vortices in the ground state quickly increase above the critical field strength. The nature of the phase transition might thus be framed in terms of a confinement-deconfinement transition of a non-Abelian gauge field, akin to the confinement-deconfinement transition in the Abelian discrete gauge theory. An example of the latter is the Z_2 gauge theory (for example, the toric code in a magnetic field²¹), for which it is well known that flux condensation leads to a confinement transition.²²

We note that although such a field-driven phase transition might share many similarities for topological states with Abelian and non-Abelian topological order, there also exist important differences. For the Abelian Z_2 gauge theory, it has been shown that the critical field strength is of the same magnitude as the single vortex gap and as a result, the closing of the gap for a single vortex will naturally lead to vortex condensation. In the non-Abelian case, however, the single vortex gap, estimated around ~ 0.27 , is considerably larger than the critical field strength. There are two potential reasons for this discrepancy in the non-Abelian case. First, the non-Abelian nature of the vortices gives rise to a macroscopic degeneracy which might in turn enhance the low energy quantum fluctuations. Second, in the limit of high vortex density, the vortex core energy can be much smaller than in the single vortex case and thus further enhance the quantum fluctuations.

VI. A SECOND CRITICAL POINT

Finally, we note that there appears to be a second critical point in our phase diagram around $\alpha \approx 0.8$ and $h = 0$, where the stripy AFM phase and the gapless spin liquid meet. We find that in the presence of the magnetic field the transition lines of the field-driven phase transition out of the corresponding

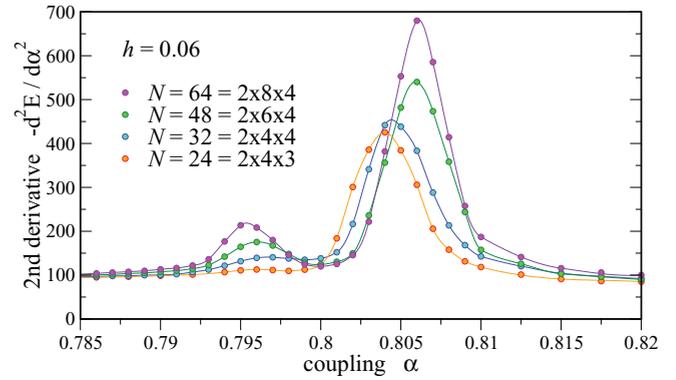


FIG. 6. (Color online) Constant field scan in the vicinity of the (critical) point separating the stripy AFM from the topological phase.

canted and topologically ordered states bend in and merge only in the zero-field limit as depicted in the inset of Fig. 1. To show that there is indeed no direct transition between the canted stripy Néel state and the topological phase we have made extensive scans in the coupling parameter α in the vicinity of this putative critical point for small field strength. As shown in Fig. 6 for $h = 0.06$, the second derivative $d^2E/d\alpha^2$ of the ground-state energy clearly shows two peaks proliferating with increasing system size indicative of two well-separated phase transitions. However, the underlying effective theory for such a multicritical point is not known, and will be left for further study.

VII. OUTLOOK

Having established the rich phase diagram of the Heisenberg-Kitaev model in a magnetic field, it is interesting to speculate where one would place the iridate Na_2IrO_3 . While experiments⁴ report indications of an AFM ordered ground state below $T_N \approx 15$ K, the precise nature of the order remains open. Given the considerable suppression of the ordering temperature T_N in comparison with the Curie-Weiss temperature $\Theta_{\text{CW}} \approx 116$ K,⁴ which is typically interpreted as an indicator of frustration, an alternative explanation would be the proximity to a quantum critical point, such as the multicritical point $\alpha \approx 0.8$ in the context of our phase diagram. This would bring the material in close proximity to the spin liquid phase for $\alpha \gtrsim 0.8$ and the topological phase found for a magnetic field pointing in the $\langle 111 \rangle$ direction. To further substantiate this possibility, it is desirable to study the finite-temperature phase diagram of our model system and to consider the effects of disorder, such as site mixing between the Ir and Na sites.⁴ Finally, it would be interesting to bring the Mott physics discussed in this manuscript in competition with the topological insulator phase suggested in Ref. 23.

ACKNOWLEDGMENTS

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- ¹⁸We note that there are other independent reflection symmetries, such as the reflection symmetry about the plane perpendicular to the $(1-10)$ direction and through the hexagon center. However, we note that the canted Néel phase remains invariant under the combination of such a reflection symmetry and its C_3^* , I symmetries. Similarly, the canted stripy AFM phase does not break the combination of such a reflection symmetry and its C_3^* translational symmetries. Consequently, the presence of such additional, independent symmetries does not affect our results.
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