Higgs-Axion conversion and anomalous magnetic phase diagram in TlCuCl$_3$

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(Dated: May 2, 2018)

What is so unique in TlCuCl$_3$ which drives so many unique magnetic properties, such as a massive Higgs mode at the magnetic critical point, long-lived paramagnons, and dimerized antiferromagnetism? To study these properties, we employ here a combination of ab-initio band structure, tight-binding parameterization, and an effective quantum field theory. Within a density-functional theory (DFT) calculation, we find an unexpected bulk Dirac cone without spin-orbit coupling (SOC). Tracing back to its origin, we identify, for the first time, the presence of a Su-Schrieffer-Heeger (SSH) like dimerized Cu chain lying in the 2D crystal structure. The SSH chain, combined with SOC, stipulates an anisotropic 3D Dirac cone where chiral and helical states are intertwined (namely, 3D SSH model). As a Heisenberg interaction is introduced, we show that the dimerized Cu sublattices of the SSH chain condensate into dimerized spin-singlet magnets. In the magnetic ground state, we also find a naturally occurring topological phase, distinguished by the axion invariant. Finally, to study how the topological axion excitation couples to magnetic excitations, we derive a Chern-Simons-Ginzburg-Landau action from the 3D SSH Hamiltonian. We find that paramagnons, which are independent of the quantum critical physics.

PACS numbers: 75.10.Jm,73.43.Lp,73.20.Mf,73.43.Nq

TlCuCl$_3$ has maintained a steady theme of research interests for more than two decades due to its unconventional magnetic properties. This material simultaneously accommodates several unusual magnetic properties, which are either individually present in other magnetic systems, or absent. TlCuCl$_3$ is a paramagnetic at ambient condition, but undergoes a quantum phase transition to an antiferromagnetic (AFM) state with small pressure,[1, 2] or with magnetic field.[3, 4] (a) The AFM phase of TlCuCl$_3$ arises from the formation of nearest neighbor quantum dimer — a spin-singlet excitation often seen in spin-liquid systems, and it does not necessarily break the translational symmetry. (b) TlCuCl$_3$ is one of the earlier systems where a Higgs mode was observed in the AFM phase, in addition to the usual Goldstone modes. (c) According to basic quantum field theory, the Higgs mass disappears at the quantum critical point.[5] But in TlCuCl$_3$, one of the Higgs mode remains massive across the AFM critical point.[6] (d) Paramagnons, gapped magnetic excitations in non-magnetic phase, usually have short lifetime, as they decay into the particle-hole continuum. But in TlCuCl$_3$, paramagnons have equally large lifetime as that of the Higgs mode across the critical point.[7] (e) In this material, Bose-Einstein condensation of magnons was experimentally achieved.[3] So, what is so special in TlCuCl$_3$ which drives such a wide variety of unusual magnetic properties in the same crystal?

Considerable experimental and theoretical studies have been devoted to understand these universal magnetic properties of TlCuCl$_3$ [1–14]. In the theoretical models, mainly the Heisenberg type spin-spin interaction is considered via various models[9, 10]. These models consistently explain the formation of spin-singlet dimers, and reproduce the experimental spin-wave dispersion[1–3]. Within the so-called $\phi^4$-theory, one can also obtain a characteristic scale of the Higgs mode’s lifetime[5, 9, 10].

To look into these questions from a materials specific, microscopic perspective, we investigate the magnetic properties of TlCuCl$_3$ constrained by its DFT band structure. To our surprise, we find that there exists an isolated Dirac cone in the bulk band structure, even in the absence of spin-orbit coupling (SOC) and magnetism. The origin of such a Dirac cone is traced back to the presence of a Cu-chain along the $c$-direction, which is reminiscent of the celebrated Su-Schrieffer-Heeger (SSH) chain, so far known in 1D polyacetylene chain.[18] The SSH chain can produce a 1D Dirac-like degenerate point at $k_z = \pm \pi/c$. However, the DFT result shows a single band crossing point at $k = (0, 0, \pm \pi/c)$. We develop a 3D SSH model for this system, which reproduces the anisotropic 3D Dirac cone with chiral (sublattice-momentum locking) states along the $k_z$-direction and helical (spin-momentum locking) state in the basal plane.

As the AFM order turns on, we find that the spin-singlet dimers are formed between the nearest neighbor Cu-sublattices of the SSH chain. This causes an inversion of the helicity between the two Cu-sublattices, driving a topologically non-trivial phase, as distinguished by a finite axion-angle ($\theta$) within the Chern-Simon theory. The interplay between the topological excitations (axions) and magnetic excitations (Goldstone, Higgs, paramagnons modes) is studied here within a microscopically derived Chern-Simons-Ginzburg-Landau (CSGL) model. We find that the CS term characteristically modifies the magnetic phase transition. In addition, the axion term gives a new contribution to the Higgs mass term which is independent of the magnetic order parameter, and hence Higgs mode remains massless across the AFM critical point. This explains why the Higgs modes remain indifferent to the magnetic critical point, and the Higgs-paramagnon conversion becomes independent of the magnetic order parameter.

DFT results. TlCuCl$_3$ crystallizes in the monoclinic P2$_1$/c space group, with 4 formula units per unit cell. We use the
experimental lattice constants of $a = 14.144$ Å, $b = 8.890$ Å, and $c = 3.983$ Å, and $\alpha = 96.32^\circ$. The top view in Fig. 1(a) shows a rectangular projection of the unit cell on the $ab$ plane. Each formula unit contains two inequivalent SSH chains along the $c$-axis as shown in Fig. 1(b), at the center and corners of the rectangle. Because of different Cl-environments, the two nearest neighbor hoppings between Cu-Cu atoms become slightly different resulting in a SSH structure.

We compute the DFT band structure using the Local Density Approximation (LDA) exchange correlation as implemented in the Vienna ab-initio simulation package (VASP)[19, 20]. LDA+U ($U = 4$ eV) method is used to deal with the strong correlation features on Cu-3d orbitals. The non-magnetic DFT band structure in Fig. 2(a) shows four bands near the Fermi level ($E_F$), stemming from the $d$-orbitals of the Cu-atoms. Each SSH chain is individually responsible for forming a 1D Dirac cone at the $k_z = \pm \pi/c$-point. The inter-chain hopping breaks the degeneracy of the bands, resulting in two gapped bands, and one single Dirac cone. The Dirac cone formation in the non-magnetic state is robust to various values of $U$ and DFT functionals. While the band degeneracy $k_z = \pm \pi/c$ is guaranteed by the chiral symmetry of the SSH chain, in the absence of SOC, there is no reason that the two sublattice bands must also be degenerate only at $k_x = k_y = 0$. However, thanks to the crystal symmetry and the crystal field splitting, TlCuCl$_3$ obtains a unique combination of the hopping integrals such that the bands are non-degenerate at every other k-points, except at $k^* = (0, 0, \pi/c)$. Our DFT band structure agrees well with a previous LMTO calculation[21], and is also reproducible with other DFT functionals.

**Tight-binding model.** Our main interest is to study the topological properties arising from the bulk Dirac cone. Since there is only one band crossing in the bulk state at $E_F$, the minimal model required to capture the essential topological properties is a two-band model forming the Dirac cone. We therefore start with a two-band tight-binding model, coming from the Cu-sublattices in a given SSH chain, and allow inter-chain hoppings in all three dimensions. We may refer the corresponding model as a 3D SSH model.

In what follows, we work in a single Cu-chain per conventional unit cell, as indicated in Fig. 1(a). We express the corresponding Hamiltonian in a 2-components spinor as $\Psi(k) = (\psi_A(k), \psi_B(k))^T$, where ‘A’, and ‘B’ stand for two Cu-atoms as

$$H_0 = \sum_{k, ij \in \{A, B\}} \left[ \xi_{ij}^A \psi_i^\dagger(k) \psi_j(k) - \mu \psi_i^A(k) \psi_i(k) \right]. \quad (1)$$

Here $\xi_{ij}^{AA} = \xi_{ij}^{BB}$, and $\xi_{ij}^{AB}$ are the intra-, and inter-sublattice dispersions, respectively. The energy eigenvalues are $E_{k_\perp}^A = \xi_{k_\perp}^{AA} \pm |\xi_{k_\perp}^{AB}|$. The two bands meet at the locii of $|\xi_{k_\perp}^{AB}| = 0$, while $\xi_{k_\perp}^{AA}$ gives an overall shift of the degenerate points in energy. In the case of an isolated 1D SSH chain, $\xi_{k_\perp}^{AB}$ is often described by $\xi_{k_\perp}^{AB} \sim (t + t'e^{-ik_x})$ (we set $a = b = c = 1$ for simplicity), where $t$, and $t'$ are the inter-sublattice hoppings along the $\pm z$-direction, respectively [see Fig. 1(b)]. A Dirac cone forms at $k_z = \pm \pi$ when $t' \approx t$. In the same spirit, we can cast the Hamiltonian in Eq. 1 into a 3D SSH model as

$$\xi_{k_\perp}^{AB} = T_{k_\perp} + T'_{k_\perp} e^{-ik_x}, \quad (2)$$

where $k_\perp = (k_x, k_y)$, $T_{k_\perp}$, and $T'_{k_\perp}$ have the same meanings as $t'$, and $t$ defined above, but due to inter-SSH chain hoppings, they acquire in-plane dispersions. $\xi_{k_\perp}^{AA}$, $T_{k_\perp}$, and $T'_{k_\perp}$ are expressed in terms of the Slater-Koster tight-binding (TB) hopping integrals between intra-, and inter-chain hoppings, and we spare the details in the supplementary materials (SM)[22].

Following the DFT result, we fit the TB dispersions to the DFT band with the constraint that $T'_{k_\perp} = T_{k_\perp}$ only at $k^* = (0, 0, \pi)$. Hence we reproduce a 3D Dirac cone, with linear dispersion in $q_x$, and quadratic dispersion in $(q_x, q_y)$, where $q = k^* - k$ and $q \ll 1$ (see inset to Fig. 2).
SOC. Although SOC is weak here, it is however sufficient to introduce helicity in the low energy spectrum. In TiCuCl₃, spins are aligned in the k₃-plane near the critical point, consistent with experiments[1–3, 7] and DFT calculation, see below. This also makes the in-plane SOC to be dominant. A full derivation of the SOC is given in the SM[22], and its non-vanishing component is given by

\[ H_{SOC} = \sum_{i,j \in (A,B)} \sum_{k, ss'} \left[ \psi_{i,s}^\dagger (k) \left( \alpha_{ij}^s \times \sigma_{ss'} \right) \psi_{j,s'} (k) \right] \]

\( s, s' \) give spin components. The components of the velocity operators are \( \alpha_{ij}^s = \alpha_{ij}^0 \left( -\partial_{k_y} \xi_{ij}^s, \partial_{k_x} \xi_{ij}^s, 0 \right) \), with \( \alpha_{ij}^0 \) being the corresponding SOC strengths. Eq. 3 allows several SOC operators are given spin components. The components of the velocity operators are \( \alpha_{ij}^s = \alpha_{ij}^0 \left( -\partial_{k_y} \xi_{ij}^s, \partial_{k_x} \xi_{ij}^s, 0 \right) \), with \( \alpha_{ij}^0 \) being the corresponding SOC strengths. Eq. 3 allows several SOC operators are

\[ H(k) = \xi_k^A 1_{4 \times 4} + \sum_{i=1}^5 d_i(k) \Gamma_i. \]

where \( \Gamma = \left( \sigma_x \otimes 1, \sigma_y \otimes 1, 1_{2 \times 2} \otimes \tau_x, 1_{2 \times 2} \otimes \tau_y, \sigma_z \otimes \tau_z \right) \), where \( \sigma \) and \( \tau \) are the Pauli matrices in the spin and sublattice basis, respectively. The components of the D-vectors are \( d = (\xi_{AA}, -\xi_{AB}, \xi_{BB}, \xi_{AB}, 0) \). The Hamiltonian is invariant under time-reversal symmetry. The parity operation constitutes of \( k \leftrightarrow -k \), and \( A \leftrightarrow B \).

Dirac Hamiltonian: Combining Eqs. 1, and 3, the full non-magnetic Hamiltonian can be expressed in terms of a spinor \( \Psi(k) = (\psi_{A\uparrow}, \psi_{B\uparrow}, \psi_{A\downarrow}, \psi_{B\downarrow})^T \) as:

\[ H(k) = \xi_k^A 1_{4 \times 4} + \sum_{i=1}^5 d_i(k) \Gamma_i. \]

where \( \Gamma = \left( \sigma_x \otimes 1, \sigma_y \otimes 1, 1_{2 \times 2} \otimes \tau_x, 1_{2 \times 2} \otimes \tau_y, \sigma_z \otimes \tau_z \right) \), where \( \sigma \) and \( \tau \) are the Pauli matrices in the spin and sublattice basis, respectively. The components of the D-vectors are \( d = (\xi_{AA}, -\xi_{AB}, \xi_{BB}, \xi_{AB}, 0) \). The Hamiltonian is invariant under time-reversal symmetry. The parity operation constitutes of \( k \leftrightarrow -k \), and \( A \leftrightarrow B \).

AFM calculations. Next, we have performed non-collinear, spin-polarized DFT calculations with and without the SOC. We find a AFM ground state with antiparallel spin between ‘A’ and ‘B’ sublattices of the SSH chain[27]. We find that the spins are quantized in the ab plane, as seen in experiment[1–3, 7], and the easy axis is almost along the diagonal direction in this plane. The DFT predicted magnetic moment along the z-direction is negligibly small, and that in the ab-plane are \( m_{A,B} = \pm 0.43 \) for the two Cu atoms, respectively. The magnetic band structure shows insulating behavior with a band gap \( \sim 1.3 \) eV.[22, 28] From the band gap and magnetic moment, we estimate the AFM coupling to be around \( J \sim 1.5 \) eV, which is close to the value estimated in neutron scattering measurement.[23]

Since the magnetic moment is small, we can treat the magnetic ground state within a mean-field order approximation.[22] We start with the nearest neighbor spin interaction term \( H_1 = J \sum_{(i,j) \in (A,B)} S_i \cdot S_j \), where \( S_i \) is the spin operator. Guided by the DFT results, we consider only the spin-spin interaction along the spin-quantization axis, and between the nearest ‘A’ and ‘B’ sublattices only. The AFM order parameter is defined as \( \phi = (m_A - m_B)/2 \), where the magnetization is \( m_{A,B} = \langle S_A^z \rangle_{A,B} \). The excitation energy gap in the band structure is \( \Delta = J \phi \). Such an order parameter have been used earlier in TiCuCl₃ and is found via self-consistent calculation to define the AFM ground state.[14] Using Hubbard-Stratonovic decomposition of the \( H_1 \), we obtain

\[ H_1 \approx J \phi \Gamma_5 = d_5 \Gamma_5. \]

Helicity inversion and topological axion insulator: The AFM order introduces a crucial change in the SOC Hamiltonian in Eq. (3). Since the spin polarization is fully reversed between the ‘A’ and ‘B’ sublattices, the corresponding SOC is also reversed, i.e., \( \alpha_{AA}^{AB} = -\alpha_{BB}^{BB} \). This induces an inversion in the helicity between the ‘A’ and ‘B’ sublattices, i.e., if the up spin is right moving in the ‘A’ sublattice, it becomes left moving in the ‘B’ sublattice, and vice versa. This helicity inversion endows the system to acquire a non-trivial topological phase.[24–26] Within the above Hamiltonian we incorporate the helicity inversion by changing \( \Gamma_{1,2} \rightarrow \sigma_x \otimes \tau_z, \sigma_y \otimes \tau_z \).

The topological invariant of the 3D AFM insulator cannot be defined by the usual \( Z_2 \) invariant or Chern number, but by an ‘axion’ invariant \( \theta \)[29, 31, 32]. The axion invariant (multiplied by \( \pi \)) for a time-reversal invariant system, and vanishes continuously as the spontaneously magnetization increases[30, 31]. The axion invariant is the solid angle enclosed in the \( d \)-space as one encircles the entire 3D Brillouin zone.[30, 32] Reminisce to the topological phase transition in a single SSH chain, we also find here that \( \theta \) becomes finite when the zeros of \( d_3(k) = \xi_{AB} \) lies inside the solid angle, giving the condition that \( T_{k,\phi} \leq T_{\phi} \), for \( k \in BZ \). Having a Dirac cone in the SOC band structure, we ensure that such a condition is automatically satisfied in the non-interacting phase. For \( \phi \rightarrow 0 \), we obtain \( \theta = \pi \).

For finite \( |\phi| > 0 \), we numerically find that \( \theta \) decreases exponentially as shown in Fig. 3a, as

\[ \theta = \pi e^{-\lambda |\phi|}, \]

where \( \lambda \propto J \), is a fitting parameter, obtained to be \( \lambda = 220 \). Owing to time-reversal symmetry breaking, the corresponding topological axion phase does not exhibit any gapless edge state.[28]

Chern-Simons-Ginzburg-Landau analysis: Finally we discuss the implications of the topological excitations to the

FIG. 3. (a) Computed values of the axion angle \( \theta \) as a function of the magnetic order parameter \( \phi \). (b) Color plot depicts values of magnetization as a function of the GL coefficient \( \alpha \), and the CS coefficient \( \gamma \) from Eq. 9. We set \( \beta > 0 \). Horizontal arrow (green) indicate a second order phase transition line where order parameter decreases continuously, while vertical arrow dictate a first-order line.
magnetic properties. The topology induced axion excitations are described by a Chern-Simons (CS) term in the effective Lagrangian.[29, 31] On the other hand, the interaction induced magnetic excitations are captured within the Ginzburg-Landau (GL) theory. The field-theory description of the competition between electromagnetic interaction and topological responses due to probe electromagnetic fields \((A_0, \mathbf{A})\) is developed earlier in the context of fractional quantum Hall effect,[33] and is termed as Chern-Simons-Ginzburg-Landau (CSGL) theory. In addition to probe fields, there may arise intrinsic ‘statistical’ gauge fields \((a_0, a)\). Thanks to the linear combination form of the intrinsic and probe gauge fields in the Lagrangian, we can combine their effects in a total gauge field as \(A_0 = a_0 + A_0\), and \(\mathbf{A} = a + \mathbf{A}\). The full Lagrangian density can be split into four parts\([33, 35]\)

\[
L_{\text{total}} = L_{\text{KE}} + L_{\text{MW}} + L_{\text{GL}} + L_{\text{CS}}.
\]

\(L_{\text{KE}}\) is the kinetic energy term which arises from the space and time-dependent parts of the field \(\phi\) and \(\theta\). \(L_{\text{MW}}\) is the Maxwell term due to EM fields. These two terms do not contribute to the magnetic phase diagram and Higgs mode we set out to discuss below and thus are not included henceforth\([22]\). The remaining GL and CS terms can be derived using the path integral description of coherent states of the total Hamiltonian \(H_0 + H_{\text{SOC}} + H_I\), and then integrating out the fermionic degrees of freedom (Grassman variables) (see SM\([22]\)) to obtain

\[
L_{\text{GL}} = \alpha|\phi|^2 + \beta|\phi|^4, \quad L_{\text{CS}} = \frac{\hbar}{\Phi_0} E \cdot B, \tag{7}
\]

Here \(\alpha\), and \(\beta\) are the typical GL-coefficients, arise from the spin-susceptibilities, explicitly evaluated with SOC in SM\([22]\), \(\Phi_0 = \hbar/e\) is the magnetic flux quantum. \(E\) and \(B\) are electric and magnetic fields (intrinsic + probe fields).

Apparently, there is no direct coupling between the scalar field \(\phi\) and the pseudo-scalar axion mode \(\theta\), rather the axion field \(\theta\) directly stems from the scalar field \(\phi\), Eq. (6). Substituting for \(\theta\) in Eq. (7), we get \(L_{\text{CS}} = \gamma e^{-\lambda|\phi|}\), where \(\gamma = \frac{\hbar}{\Phi_0} E \cdot B\) is a variational parameter. \(\gamma > 0 (\gamma < 0)\) if \(E\) and \(B\) are parallel (antiparallel) to each other, and otherwise zero. Neglecting the irrelevant space-time dependence of the order parameter, we arrive at the CSGL term, expressed exclusively in terms of the AFM field \(\phi\) as

\[
L_{\text{CSGL}} = \alpha|\phi|^2 + \beta|\phi|^4 + \gamma e^{-\lambda|\phi|} - \gamma. \tag{8}
\]

(We have added a constant term \(\gamma\) to shift the Free energy \((\propto L)\) minimum to zero at \(\phi = 0\). The magnetic phase transition, and magnetic excitations can now be studied as a function of four variational parameters \(\alpha, \beta, \gamma, \lambda\).

**Magnetic phase diagram:** Minimization of \(L_{\text{CSGL}}\) at a finite value of \(\phi = \phi_0\) is obtained at the roots of the following secular equation:

\[
2(\alpha + 2\beta|\phi_0|^2)|\phi_0| = \gamma \lambda e^{-\lambda|\phi_0|}. \tag{9}
\]

Solution of the above equation is non-trivial to manage analytically. For \(\gamma \to 0\), we recover the typical GL result of \(|\phi_0| = \sqrt{-\alpha/2\beta}\), giving a second order phase transition as \(\alpha\) becomes negative (with \(\beta > 0\)). Since we are in the vicinity of a second order phase transition, we set \(\beta > 0\), and \(\lambda = 220\) (from Fig. 3a). We study the solution of \(\phi_0\) as a function of \(\alpha\) and \(\gamma\), as given in Fig. 3b. For \(\gamma > 0\) region, we find that \(\phi_0\) decreases continuously to zero, suggesting a second order phase transition as a function of both \(\alpha\), and \(\lambda\). On the other hand, for \(\gamma < 0\), we notice that the phase boundary from finite \(\phi_0\) to zero is discontinuous, implying that the phase transition becomes first order. To understand this behavior, we expand the CS term in the leading order in \(|\phi|\) as \(\gamma \lambda|\phi|\). So, for \(\gamma > 0\), \(L_{\text{CSGL}}\) decreases with increasing \(|\phi|\), and hence its minima continuously move from \(\phi = 0\) to \(|\phi_0| > 0\) – a second order phase transition. While for \(\gamma < 0\), the increases with increasing \(|\phi|\), and then a second minima occurs at a finite \(|\phi_0| > 0\). Since \(|\phi_0|\) minima are now disjointed from the \(\phi = 0\) minima, we have a first order phase transition.

In both cases, we also observe that the phase boundary shifts from the GL limit of \(\alpha = 0\) line to finite values of \(\pm \alpha\) in the two cases, respectively. This has implications to the values of the Néel temperature, and the Higgs mass. By expanding the axion term up to \(|\phi|^2\), and assuming \(\alpha = \alpha_0(1 - T/T_{N,0})\) for \(\gamma = 0\), we obtain that the effective Néel temperature modifies as

\[
T_N = T_{N,0}(1 + \gamma \lambda^2/2\alpha_0). \tag{10}
\]

\(T_N\) increases (decreases) for \(\gamma > 0 (\gamma < 0)\). This means, \(T_N\) increases (decreases) as the applied magnetic and electric fields are parallel (antiparallel), which provides unique testbed to verify the topological nature of this magnetic ground state.

**Magnetic excitations.** Finally we study the interplay between the magnetic and topological excitations. We expand the order parameter near its expectation value as \(\phi = |\phi_0 + \delta\phi(x)|e^{i\eta(x)}\), where \(\delta\phi\) and \(\eta\) are the corresponding amplitude, and phase fluctuations, respectively. In Eq. (7), we find that \(L_{\text{CSGL}}\) depends on the amplitude \(|\phi|\) only, and thus the phase, \(\eta\), fluctuations remain gapless (Goldstone modes) even in the presence of the axion term (see SM\([22]\) for the derivation) [In fact, the Goldstone modes could be gauged out by a suitable gauge transformation of the EM fields \(A\)]. Substituting \(\phi = |\phi_0 + \delta\phi(x)|\) in Eq. (7), we can estimate the mass of the amplitude mode as \(M = 1/2 \partial_{\phi_0}^2 L_{\phi=0}\). After substituting Eq. (9) at the saddle point of the Lagrangian, we obtain the Higgs mass as

\[
M = \alpha + \lambda\alpha|\phi_0| + 6\beta|\phi_0|^2 + \lambda\beta|\phi_0|^3. \tag{11}
\]

For \(\gamma \to 0\), we recover the GL value of \(M = 4\beta|\phi_0|^2\) vanishing at the critical point where \(\phi_0 \to 0\). However, in the present case, we find that there is a finite Higgs mass even above the critical point and eventually vanishes only when \(\alpha = 0\). On the other hand, for \(\gamma > 0\), we notice in Fig. 3b, that a continuous phase transition can occur at \(\alpha > 0\), giving a non-vanishing Higgs mass at the critical point, which may be called ‘topological paramagnons’. For \(\gamma < 0\), we have a first order phase transition at \(\alpha < 0\), where the order parame-
The calculation of the Higgs mode’s lifetime is rather cumbersome. One source of Higgs lifetime is the quartic term in the Lagrangian. In this spirit, the leading term in the inverse lifetime ($\tau$) is proportional to the coefficient of the $\delta \phi^4$, which can be obtained from $\partial^2 I_{\text{CSGL}} / \partial \delta \phi^4 |_{\delta \phi = 0}$, leading to

$$\frac{1}{\tau} \propto 2(12\beta - \alpha \lambda^2)|\phi_0| - 4\lambda^2|\phi_0|^3.$$ (12)

The result suggests that the Higgs lifetime rather decreases near the critical point for $\gamma \neq 0$, while away from the critical point, as the second term becomes dominant, it tends to increase. Hence we can argue that the ‘topological paramagnons’ have much reduced decay rate, and is topologically protected.

**Conclusions and outlook:** In the present model, axion is a pseudo-scaler, and there is only one Higgs mode. Therefore, the axion-Higgs coupling can be captured well within the proposed CSGL theorem, and the corresponding Lagrangian resemblance that of the Standard Model of the particle physics. It is known that in the case of a Higgs doublet, there arises axion-Higgs cross term in the Lagrangian, and the system looses its CSGL symmetry, and one obtains a so-called Peccei-Quinn (PQ) symmetry, which violates the Standard model.[34] However, the predicted Higgs doublet is yet to be observed. Based on the above analysis, we anticipate that our work will stimulate research for the realization of PQ symmetry in condensed matter systems where topological axion and Higgs terms are intertwined.[34]

We thank Soumi Ghosh for help with some of the analysis. TD acknowledges the financial support from Science and Engineering Research Board (SERB), Department of Science & Technology (DST), Govt. of India for the Start-Up Research Grant (Young Scientist).

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Supplementary Material: Higgs-Axion conversion and anomalous magnetic phase
diagram in TlCuCl$_3$

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(Dated: May 2, 2018)

In this supplementary material, we give details of the derivations corresponding to various terms presented in the main text. In Sec. I, we show DFT calculations including spin-orbit coupling in the paramagnetic and AFM phases. Next we derive the Ginzburg-Landau-Chern-Simons theory. Subsequently, we calculate the Lagrangian minima and AFM transition temperature for the CSGL theory. In the last section, calculations of Higgs mass and corresponding lifetime are presented.

DFT CALCULATION W/ SOC

We compute the DFT band structure using the Local Density Approximation (LDA) exchange correlation as implemented in the Vienna ab-initio simulation package (VASP)[1]. The results remain characteristically the same for the GGA and other functionals. The DFT band structure also agrees well with a previous LMTO calculation. In our LDA+U calculation, the electronic wave function is expanded using plane wave up to a cutoff energy of 500 eV. Brillouin zone sampling is done by using a (8 × 8 × 8) Monkhorst-Pack k-grid. Both atomic position and cell parameters are allowed to relax, until the forces on each atom are less than 0.01 eV/Å. Projected augmented-wave (PAW) pseudo-potentials are used to describe the core electron in the calculation[2].

We found in the DFT band structure that the magnetic gap is larger than the crystal field splitting (CFS), and thus one obtains a band insulating behavior in the electronic properties. As one approaches the magnetic critical point, the magnetic gap takes over for $\Delta < \Delta_{\text{CFS}}$.

![Computed ab-initio band structure of TlCuCl$_3$ in presence of spin-orbit coupling (a) with magnetism and (b) without magnetism.](image)

SSH TERM

The expression for SSH like term in our 3D model looks like

$$
H_{AA} = C_{10} \cos((k_x + k_z)/2) + C_{11} \cos(k_y) + C_{12} \cos((k_x - k_z)/2) + C_{13} \cos(2 * k_y) + C_{14} \cos(k_x) + C_{15} \cos(k_z) + C_{16} \cos(k_x + k_z) + C_{17} \cos(k_x - k_z) + C_{18}
$$

$$
H_{AB} = e^{ik_z / 2} \left( C_0 + C_1 e^{-ik_z} + (C_2 + C_3 e^{-ik_z}) (C_4 e^{i(k_x + k_y)/2} + C_5 e^{-i(k_x + k_y)/2}) + (C_6 + C_7 e^{-ik_z}) (C_8 e^{i(k_x - k_y)/2} + C_9 e^{-i(k_x - k_y)/2}) \right)
$$
where C’s are the fitting parameters. The above equation for inter-sublattice hopping can be rewritten in the form,

\[ H_{AB} = e^{ik_z/2} T_{k \perp} (1 + \frac{T'_{k \perp}}{T_{k \perp}} e^{ik_z}) \]

where

\[ T_{k \perp} = C_0 + C_2 C_4 e^{i(k_x + k_y)/2} + C_2 C_5 e^{-i(k_x + k_y)/2} + C_6 C_8 e^{i(k_x - k_y)/2} + C_6 C_9 e^{-i(k_x - k_y)/2} \]

\[ T'_{k \perp} = C_1 + C_3 C_4 e^{i(k_x + k_y)/2} + C_3 C_5 e^{-i(k_x + k_y)/2} + C_7 C_8 e^{i(k_x - k_y)/2} + C_7 C_9 e^{-i(k_x - k_y)/2} \]

(3)

The fitting parameters are \( C_{0-18} = [0.156, 0.209, -0.076, 0.223, -0.135, -0.325, 1.466, -0.019, -0.030, 0.045, 0.002, -0.033, -0.003, 0.027, -0.002, 0.095, -0.105, -0.019] \) in eV.

FIG. 2: Figure showing effective hopping in \((x+y,z)\) plane. Here \( C'_{0}, C''_{0}, C'_{1} \) and \( C''_{1} \) correspond to \( C_2 C_4, C_2 C_5, C_3 C_4 \) and \( C_3 C_5 \) respectively. We have similar kind of hopping terms in \((x-y,z)\) plane as well.

**CHERN-SIMONS-GINZBURG-LANDAU THEORY**

Ginzburg-Landau theory

Here we develop the Ginzburg Landau theory of our Hamiltonian around the antiferromagnetic order parameter. We write the partition function for the total Hamiltonian \( H + H_1 \) written in terms of the Dirac matrices in Eq. (4) in the main text as

\[
Z = \int D[\psi, \bar{\psi}] \exp \left[ -\int_0^\beta d\tau \left( \bar{\psi}(\partial_\tau \mathbb{I}_4 \times \mathbb{I}_4 - H_k)\psi - J \sum_{\langle i,j \rangle} S_i S_j \right) \right],
\]

(4)

where \( \psi \) are 4 component Grassman variables \( \psi = (\psi_{A1}, \psi_{B1}, \psi_{A2}, \psi_{B2})^T \) (same as the Dirac spinor used in the main text), and \( \bar{\psi} \) is the conjugate of \( \psi \). \( i,j \) denote ‘A’, ‘B’ sublattices. \( S_i \) are the corresponding spin operators. We orient the spin-quantization axis along \( \sigma_z \), i.e., we only consider \( S_z^i \) component. We define the antiferromagnetic (AF) field as \( \phi = (S_A^z - S_B^z) / 2 \). Using the Hubbard Stratonovich transformation for \( H_1 \) in terms of the FM fields in the last
term of Eq. (4), we obtain
\[
\int D[\psi, \bar{\psi}] \exp \left( -J \sum_{(i,j)} S_i S_j \right) = \int D[\psi, \bar{\psi}, \phi, \bar{\phi}] \\
\times \exp \left[ -J \phi (S_\Lambda - S_0) - \phi^2 \right].
\]

(5)

Now we express \( S_i^a \) in terms of the Grassman variables as \( S_i^a = (\bar{\psi}_i \Psi_i - \bar{\psi}_i \psi_i) / 2 \). In doing so, we can write the AF term in terms of the Grassman spinor \( \phi \) as \( J \phi (S_\Lambda - S_0) = \psi (J \phi \Gamma_5) \psi \), where \( \Gamma_5 = \sigma_z \otimes \tau_z \), as defined in the main text. Substituting this identity in Eq. (5), and then inserting it back to Eq. (4), we get
\[
Z = \int D[\psi, \bar{\psi}, \phi, \bar{\phi}] \exp \left[ - \int_0^\beta d\tau \bar{\psi} (G^{-1}_0(\tau, k) - M(\phi)) \psi \right].
\]

(6)

Here we have defined the non-interacting Green’s function matrix \( G^{-1}_0(k, \tau) = \partial_\tau \delta_{4\times4} - H_k \), and the magnetization matrix as \( M(\phi) = J \phi \Gamma_5 \). Now we can go to the Matsubara frequency \( i \omega_n \) domain and integrate out fermion variables \( (\psi, \bar{\psi}) \) to get the effective Lagrangian density as
\[
\mathcal{L} = \text{Log} \left[ \text{Det} \left( \sum_{i \omega_n, k} G^{-1}_0(i \omega_n, k) - M(\phi) \right) \right] - \frac{\phi^2}{4J}.
\]

(7)

Under the saddle point approximation around the AFM, using the identity \( \text{Log}[\text{Det}[..]] = \text{Tr}[\text{Log}[[..]] \) and \( \text{Log}[x] = -\sum_{n=1}^\infty (-1)^n x^n / n \), we get the GL Lagrangian potential
\[
\mathcal{L}_{GL} = \alpha |\phi|^2 + \beta |\phi|^4 + O(|\phi|^6),
\]

(8)

where
\[
\alpha = -\frac{1}{4J} + \text{Tr} \sum_{k, k'} G_0(k) \Gamma_5 G_0(k') \Gamma_5,
\]

(9)

\[
\beta = \text{Tr} \sum_{k, k', k''} G_0(k) \Gamma_5 G_0(k') \Gamma_5 G_0(k'') \Gamma_5 G_0(k'''') \Gamma_5,
\]

(10)

where we define \( k = (k, i \omega_n) \). Exact computation of \( \alpha \) and \( \beta \) variables are difficult, but we can already grasp the essence that \( \beta > 0 \), and \( \alpha \to 0 \) when the particle-hole bubble compensates the interaction terms. These results are typical for the GL theory.

**The Chern-Simons term**

Chern-Simons term arises in the presence of electromagnetic (EM) fields. We assume the probe electromagnetic fields as \( (A_0, \mathbf{A}) \). In addition to probe fields, there may arise intrinsic ‘statistical’ gauge fields \( (a_0, \mathbf{a}) \) due to fluctuations of the bosonic fields \( \phi \). This can be seen easily. The statistical gauge field arises due to fluctuations of the order parameter, so we can write \( a_0 \propto \partial_t (\delta \phi \delta \phi) \), and \( \mathbf{a} \propto \nabla (\delta \phi \delta \phi) \), where \( \delta \phi \) is the fluctuation of the AFM field around its saddle point \( \phi_0 \). Such intrinsic gauge clearly arises from the \( |\phi|^4 \) term in the GL potential in Eq. (8), and persists above the AFM critical point. We are not particularly interested in the details of the origin of the intrinsic gauge field, except it conveys an important message that such due to spin-fluctuations in space-time dimensions, there can be CS term even in the absence of any external EM field. Readers interested in the details of the origin of such statistical gauge field can refer to Refs. [3, 8] and references therein.

Thanks to the linear combination form of the intrinsic and probe gauge fields in the Lagrangian, we can combine their effects in a total gauge field as \( A_0 = a_0 + A_0 \), and \( \mathbf{A} = \mathbf{a} + \mathbf{A} \). Due to the total EM field, we have a typical Maxwell term \( \mathcal{L}_{MW} \), and the Chern-Simons term \( \mathcal{L}_{CS} \) as defined in 3+1 dimensions as \[4, 5, 7\]
\[
\mathcal{L}_{MW} = -\frac{1}{4} F_{\mu \nu} F^{\mu \nu} - A_\mu J^\mu,
\]

(11)

\[
\mathcal{L}_{CS} = \frac{\theta}{\Phi_0} \epsilon_{\mu \nu \sigma} A_\mu \partial_\nu A_\sigma - A_\mu J^\mu.
\]

(12)
where the Einstein summation convention is implied. \( \mathcal{F}_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu \), current density \( \mathcal{J}^\mu \) is included by conservation principles and can be eliminated for the Lagrangian minimization problem of our interest. \( \theta \) is the axion angle which is related to the momentum-space non-Abelian Berry connection \( \mathcal{A}^{st}_\mu = -i(u_k^s | \partial_\mu u_k^s \rangle \langle u_k^s | ) \), where \( | u_k^s \rangle \) is the \( s^{th} \)-eigenstate of the mean-field Hamiltonian,[? ] as

\[
\theta = \frac{1}{4\pi} \int_{\text{BZ}} d^3 k e^{\mu\nu\sigma} \text{Tr} \left[ \mathcal{A}_\mu \partial_\nu \mathcal{A}_\sigma + i \frac{2}{3} \mathcal{A}_\mu \mathcal{A}_\nu \mathcal{A}_\sigma \right].
\]

(13)

By evaluating the eigenvectors of our Hamiltonian in the main text, we can obtain an algebraic, gauge independent form of the axion angle can be deduced to be:

\[
\theta = \int_{\text{BZ}} d^3 k \frac{2|d| + d_1}{4\pi (|d| + d_4)^2} e^{ijkl} d_i \partial_{k_1} d_j \partial_{k_2} d_k \partial_{k_3} d_l.
\]

(14)

where \( |d|^2 = \sum_{i=1}^5 |d_i|^2 \), and \( d_5 = J \phi \), and \( i, j, k, l \) runs from 1,2,4,5. The above integral evaluates the solid angle enclosed in the \( d \)-space as one encircles the entire 3D Brillouin zone in the \( k \)-space. Reminiscence to the topological phase transition in a single SSH chain, here also we can show that \( \theta \) acquires finite value where the zeros of \( d_3(k) \) term lies inside the solid angle, giving the condition that \( T_{k_+} \leq T_{k_+} \), for \( k \in \text{BZ} \). Having a Dirac cone in the SOC band structure, we ensure that such a condition is automatically satisfied in the non-interacting phase (\( d_5 = 0 \)). Axion angle can be calculated numerically. We are interested in studying its behavior as a function AFM field \( \phi \). Absorbing the remaining factors in the CS term into \( \gamma > 0 (\gamma < 0) \) if \( E \) and \( B \) are parallel (antiparallel) to each other.

### Chern-Simons-Ginzburg-Landau theory

The Kinetic energy term due to the AFM field is

\[
\mathcal{L}_{\text{KE}} = i \phi^* D_0 \phi + \frac{1}{2m} \phi^* D^2 \phi.
\]

(15)

Here the covariant derivative operators are \( D_0 = \partial_t + ie A_0 \), and \( D = i \nabla + e A \). Therefore the total Lagrangian density becomes\cite{3, 4} \( \mathcal{L}_{\text{total}} = \mathcal{L}_{\text{KE}} + \mathcal{L}_{\text{MW}} + \mathcal{L}_{\text{GL}} + \mathcal{L}_{\text{CS}} + \mathcal{L}_{\text{AN}} \). Here \( \mathcal{L}_{\text{AN}} \) represents the contribution from anyons arising from the fluctuation of the order parameters. The Maxwell term does not involve the order parameter or axion term, and thus also can be neglected. Neglecting space-time dependence of the order parameter, we obtain the effect CSGL term in terms of the AFM field \( \phi \) as

\[
\mathcal{L}_{\text{CSGL}} = \alpha |\phi|^2 + \beta |\phi|^4 + \gamma e^{-\lambda |\phi|}.
\]

(16)

### FREE ENERGY MINIMA AND TRANSITION TEMPERATURE

The Free energy minima occurs where the Lagrangian in Eq. (16) is also minima. Solving for \( \frac{\partial F[\phi]}{\partial \phi} = 0 \), we get

\[
2 (\alpha + 2\beta |\phi_0|^2) |\phi_0| = \gamma \lambda e^{-\lambda |\phi_0|}.
\]

This equation cannot be solved analytically, but for small \( \lambda \), we can expand the exponential up to third power in \( \phi \) to get

\[
\alpha |\phi_0| + 4\beta |\phi_0|^3 - \lambda \gamma (1 - \lambda |\phi|) + \frac{\lambda^2 |\phi|^2}{2!} - \frac{\lambda^3 |\phi|^3}{3!} = 0.
\]

(17)

It turns out that any arbitrarily small value of \( \gamma \), this leads to a minima in free energy away from \( \phi = 0 \) but very close. At high temperature, it goes arbitrarily close to 0.

The axion term leads to a correction to the Néel temperature \( T_N \). If we assume \( T_{N,0} \) is the Néel temperature without the axion term, then for a second order phase transition, we can write \( \alpha = \phi_0 (1 - T/T_{N,0}) \), where \( \alpha_0 > 0 \) is a constant. This coefficient is modified to \( \alpha' = \alpha + \gamma \lambda^2/2 \). \( \alpha' = 0 \) gives the AFM transition. Therefore, the phase transition condition becomes \( \alpha_0 \left( 1 - \frac{T}{T_{N,0}} \right) + \gamma \lambda^2/2 = 0 \), which gives \( T_N = T_{N,0} \left( 1 + \frac{\gamma \lambda^2}{2 \alpha_0} \right) \).
HIGGS MASS AND LIFETIME

Using CSGL Lagrangian, we can calculate the mass of the Higgs mode. This can be done by substituting an amplitude fluctuation term $\delta \phi$ in the Lagrangian, and calculating the coefficient of the quadratic term in fluctuation. (We do not worry about the phase fluctuation term here since it is ‘eaten’ away by the probe fields.) Replacing $|\phi| \to |\phi_0| + \delta \phi$ in Eq. (16) (assuming $\phi$ is positive), evaluating $\frac{\partial^2 L_{CSGL}}{\partial^2 \delta \phi}|_{\delta \phi=0}$ at $\delta \phi \to 0$, we get the Higgs mass as

$$\frac{\partial^2 L_{CSGL}}{\partial^2 \delta \phi}|_{\delta \phi=0} = 2 \alpha + 12 \beta (|\phi_0|)^2 + \gamma \lambda^2 e^{-\lambda |\phi_0|},$$
$$= \alpha + \lambda \alpha |\phi_0| + 6 \beta |\phi_0|^2 + \lambda \beta |\phi_0|^3. \tag{18}$$

In the last equation, we have substituted the condition for the saddle point, given above Eq. (17).

Unlike Higgs mass, its lifetime cannot be estimated exactly. One source of lifetime to the Higgs boson is the interaction term, i.e. quartic term in the Lagrangian. In this spirit, the leading term in the inverse lifetime ($\tau$) of the Higgs mass is proportional to the coefficient of the $\delta \phi^3$, which can be calculated by setting $\delta \phi = 0$ in $\frac{\partial^3 L_{CSGL}}{\partial \delta \phi^3}|_{\delta \phi=0}$ which leads to

$$\frac{1}{\tau} \propto \frac{24 \beta |\phi_0| - \lambda^3 \gamma e^{-\lambda |\phi_0|}}{\lambda},$$
$$= 2 (12 \beta - \alpha \lambda^2) |\phi_0| - 4 \lambda^2 \beta |\phi_0|^3. \tag{19}$$

Here one should take the absolute value of the right-hand side.

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