Chiral Spin Order in Kondo-Heisenberg Systems

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We demonstrate that low dimensional Kondo-Heisenberg systems, consisting of itinerant electrons and localized magnetic moments (Kondo impurities), can be used as a principally new platform to realize scalar chiral spin order. The underlying physics is governed by a competition of the Ruderman-Kittel-Kosuya-Yosida (RKKY) indirect exchange interaction between the local moments with the direct Heisenberg one. When the direct exchange is weak and RKKY dominates, the isotropic system is in the disordered phase. A moderately large direct exchange leads to an Ising-type phase transition to the phase with chiral spin order. Our finding paves the way towards pioneering experimental realizations of the chiral spin liquid in systems with spontaneously broken time-reversal symmetry.

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Interactions between magnetic moments usually lead to some kind of magnetic order where rotational symmetry is broken and the order parameter is linear in spins [1]. This is what happens in ferromagnets, antiferromagnets, and all sorts of helimagnets. Villain has demonstrated [2] that, in addition to the magnetic order, helical magnets possess a vector chiral order parameter. It is bilinear in spins and is related to the mutual orientation of neighboring spins. This chiral order breaks the discrete symmetry and can exist even without magnetic order [3]. The discovery of the vector chiral order has given rise to the idea that there could exist an order which includes a combination of three spins. The corresponding order parameter is a mixed product of three neighboring spins; see $\mathcal{O}_c$ in Eq. (1) and Refs. [4,5]. It breaks time-reversal and parity symmetries. Such an order parameter is considered as the key quantity for description of exotic magnetic phases [4]. In contemporary language, $\mathcal{O}_c$ is referred to as "scalar chiral spin order," and the state of matter with (spontaneously) broken time-reversal and parity symmetries but with conserved spin rotational symmetry is called chiral spin liquid (CSL) [6]. The seminal example possessing the CSL symmetry is the Kalmeyer-Laughlin model [7–10]. Its wave functions demonstrate the topological behavior inherent in the fractional quantum Hall effect. Thus, the Kalmeyer-Laughlin model links spin liquids and topologically nontrivial states [11–16] and can be called "topological CSL." An increasing interest in the topological CSL [17–23] is stimulated, in part, by a search for exotic (anyon) superconductivity [24,25] and by the physics of skyrmions [26–29]. The latter can be realized in magnets with the chirality resulting from the lattice structure or from the Dzyaloshinskii-Moriya interaction [30–33].

Although the concept of CSL and its order parameter were introduced in the 1980s, it still remains unclear whether such a state can exist in realistic systems where time-reversal symmetry is not explicitly broken. Numerous theoretical suggestions include spin systems with a complicated set of either Heisenberg exchange interactions extended far beyond nearest neighbors [34–36] or multispin interactions [15,16], moat-band lattices [37], and even laser-driven Mott insulators [38]. This list can be continued, but, to the best of our knowledge, the question is still open and a reliable experimental evidence of CSL governed by the spontaneously broken time-reversal symmetry is still absent.

The goal of this Letter is to demonstrate that this uncertainty can be removed by realizing CSL in Kondo-Heisenberg systems (KHS) [39–43], which consist of localized spins and itinerant electrons. Their coexistence leads to a competition between the direct Heisenberg spin exchange and Ruderman-Kittel-Kosuya-Yosida (RKKY) generated by the electrons, Fig. 1: the short-range Heisenberg exchange prefers a commensurate Néel order and RKKY prefers an incommensurate plane spiral order. Thus, the system is magnetically frustrated. When the Heisenberg interaction exceeds some critical value, see

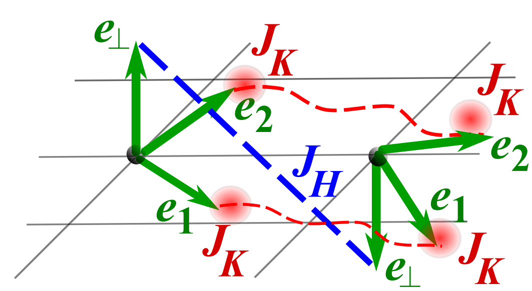

FIG. 1. Competition between spin interactions in KHS. The spin on each lattice site is decomposed in terms of an orthonormal triad $\boldsymbol{e}_{1,2,3}$ (green arrows) with $\boldsymbol{e}_\perp = (-1)^{N(r)}\boldsymbol{e}_3$; see Eq. (3). The RKKY exchange interaction (red lines) is mediated by electrons (red circles) and favors helical-like configuration of the vectors $\boldsymbol{e}_{1,2}$. The Heisenberg exchange interaction (blue line) favors antiparallel orientation of $\boldsymbol{e}_\perp$ on neighboring lattice sites. Coupling constants $J_{K,H}$ are introduced in Eq. (2).

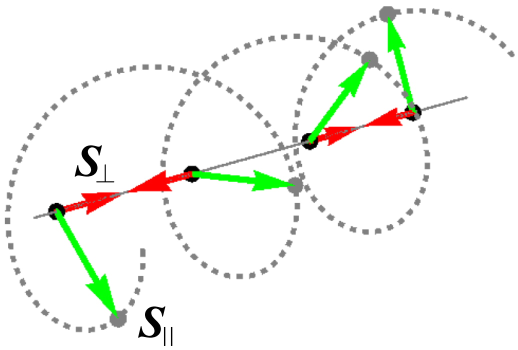

FIG. 2. Chiral configuration of spins in the CSL phase. The dotted line is the helix. The green and red arrows show helical $\boldsymbol{S}_\parallel$ and antiferromagnetic $\boldsymbol{S}_\perp$ spin components, respectively; see Eq. (3). For simplicity, we disregard helix deformations on the scale of several lattice constants which are caused by the thermal fluctuations of the triad $\boldsymbol{e}_{1,2,3}$.

Eqs. (5) and (6), the compromise between the two interactions is reached via an Ising-type phase transition leading to formation of a 3D spiral, Fig. 2. It is accompanied by the spontaneous breaking of the chirality and by an appearance of the CSL order. This is our main result.

We emphasize that the scalar chirality is necessary for the quantum effects mentioned above, but it does not require them and can exist in spin systems where the magnetic order is destroyed not by quantum but by thermal fluctuations. We shall demonstrate that the CSL state can emerge in classical (quasi-)two-dimensional systems when the spin susceptibility of the electrons has a sharp maximum at some nonzero wave vector $\boldsymbol{Q}$ incommensurate with the lattice. The easiest way to model this is to assume that the Fermi surface has nested portions. In the second order in the spin-electron coupling constant, the Fourier transform of the RKKY exchange is proportional to the spin susceptibility of itinerant electrons and, hence, is strongly enhanced at $\boldsymbol{Q}$. Without loss of generality, we can consider KHS with the spins situated on a 2D lattice with a short-range antiferromagnetic Heisenberg exchange. The spins interact with electrons with a nested Fermi surface. Thermal fluctuations in 2D prevent long-range spin order in the SU(2) symmetric system, but do not prevent the chiral one. When the Heisenberg exchange overwhelms the RKKY interaction, the scalar chiral order (SCO) emerges as the only nontrivial order parameter:

$$\mathcal{O}_c = (\boldsymbol{S}(\boldsymbol{r}_1), [\boldsymbol{S}(\boldsymbol{r}_2) \times \boldsymbol{S}(\boldsymbol{r}_3)]), \tag{1}$$

where $\boldsymbol{S}$ are the spin operators located on neighboring lattice sites $\boldsymbol{r}_{1,2,3}$. The energetically favorable spin configuration is presented in Eq. (3). Such 3D spiral of spins is the only configuration which preserves the constraint on the spin length and, at the same time, contains Fourier components with $\pm\mathbf{Q}$ and Néel wave vectors. We predict that $\mathcal{O}_c$ acquires a nonzero expectation value below a certain temperature breaking parity and time-reversal symmetries. Unlike noncollinear magnets, which have other order parameters (e.g., linear in spins), the thermodynamic CSL phase is fully characterized by $\mathcal{O}_c$.

We will now explain how to justify our predictions. We consider the model combining the Kondo lattice Hamiltonian and the Heisenberg interaction between the local moments, $\hat{H} = \hat{H}_K + \hat{H}_H$, where

$$\hat{H}_K = \sum_{\boldsymbol{k}} \epsilon(\boldsymbol{k}) \hat{c}^\dagger(\boldsymbol{k}) \hat{c}(\boldsymbol{k}) + J_K \sum_{\boldsymbol{r}} \hat{c}^\dagger(\boldsymbol{r}) \boldsymbol{\sigma} \hat{c}(\boldsymbol{r}) \boldsymbol{S}(\boldsymbol{r}),$$

$$\hat{H}_H = J_H \sum_{\boldsymbol{r},\boldsymbol{a}} \boldsymbol{S}(\boldsymbol{r}+\boldsymbol{a}) \boldsymbol{S}(\boldsymbol{r}), \qquad \boldsymbol{S} = \{S_x, S_y, S_z\}. \tag{2}$$

Here $\hat{c}^T \equiv (c_\uparrow(\boldsymbol{r}), c_\downarrow(\boldsymbol{r}))$ are electron operators at lattice site $\boldsymbol{r}$, $\hat{c}(\boldsymbol{k})$ is Fourier-transformed $\hat{c}(\boldsymbol{r})$, $\boldsymbol{\sigma} = \{\sigma_x, \sigma_y, \sigma_z\}$ are Pauli matrices, $S_{x,y,z}(\boldsymbol{r})$ are components of the spin-s operator $\boldsymbol{S}$ located on lattice site $\boldsymbol{r}$, and $J_{K,H}$ are coupling constants of the isotropic exchange interaction which are much smaller than the bandwidth, $sJ_K,\ sJ_H \ll D$. The Heisenberg exchange acts between nearest neighbors; i.e., $\boldsymbol{a}$ are the smallest vectors of the lattice. To model the above discussed maximum of the electron spin susceptibility, we assume that the dispersion $\epsilon(\boldsymbol{k})$ is nested with a wave vector $\boldsymbol{Q}$ being incommensurate with the lattice: $\epsilon(\boldsymbol{k}) = -\epsilon(\boldsymbol{k}+\boldsymbol{Q})$. We emphasize that this is just a simple model providing the susceptibility maximum and nesting should not be considered as a strict requirement for our theory. The electron band is far from half filling. We concentrate on the regime where RKKY suppresses the Kondo screening such that the latter can be neglected; see Ref. [44] for details. For the sake of simplicity, we will not distinguish the crystalline and the spin lattices. To simplify the calculations, we choose the 2D dispersion relation $\epsilon(\boldsymbol{k}) = k_x^2/2m_x - 2t_y \cos(k_y a_y)$ [45], which is parametrized by the effective mass in the x direction m_x and by the hopping integral along the y direction t_y. Results will be simplified for the case of a square 2D lattice with equal lattice constants $a_x = a_y = a_0$.

A one-dimensional Kondo chain [a 1D version of the model Eq. (2) with $J_H = 0$] was studied in Refs. [44,46]. It has been shown that, in the case of densely located spins, the physics is dominated by the backscattering processes which generate the RKKY exchange and suppress the Kondo screening. We have obtained nonperturbative solutions for cases of the easy-axis and of the easy-plane anisotropy of the Kondo exchange. In the latter case, the local spins assemble into a quasi-long-range vector chiral (or "helical") order [47]. The spontaneously chosen helix orientation (left or right handed) breaks the helical symmetry of the conduction electrons which results in a symmetry protection of the ideal transport.

In this Letter, we concentrate on magnetic properties of KHS. Because of thermal fluctuations, the helical spin ordering does not occur when the SU(2) symmetry is present. Therefore, KHS is in a disordered phase at

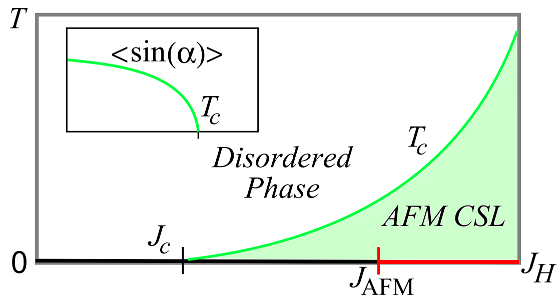

FIG. 3. Phase diagram of the isotropic 2D KHS on the plane T vs J_H at $T \ll s|J_K|$. The green line is the critical line; see Eq. (7). It separates the disordered phase and CSL (green area). $J_{\rm AFM}$ marks the transition from CSL to the antiferromagnetic phase (red line) at $T = 0$. Inset: Temperature dependence of the mean value $\langle \sin(\alpha) \rangle$. Note that the SCO parameter is proportional to this quantity; see Eq. (9).

$J_H < J_c \sim (J_K^2/D)\log(D/|J_K|)$. When J_H exceeds J_c, an Ising-type phase transition occurs and the spins form SCO; see the phase diagram on Fig. 3.

To establish the existence of the CSL it suffices to calculate the ground state energy of our model in the proper spin background. These calculations are similar to those for the 1D Kondo chain [44,46]. We outline them for KHS skipping algebraic details. Firstly, we change from the Hamiltonian to the action and single out slow fermionic modes located at the right and left sheets of the open Fermi surface [45], with an ultimate aim to develop an effective low-energy field theory for the spins. To do this, we separate fast and slow spin degrees of freedom which can be conveniently done with the help of the parametrization:

$$\begin{aligned} \boldsymbol{S}(\boldsymbol{r}) &= \boldsymbol{S}_\parallel(\boldsymbol{r}) + \boldsymbol{S}_\perp(\boldsymbol{r}), \\ \boldsymbol{S}_\parallel &= s\cos(\alpha)[\boldsymbol{e}_1\cos(\boldsymbol{Q}\cdot\boldsymbol{r}) + \boldsymbol{e}_2\sin(\boldsymbol{Q}\cdot\boldsymbol{r})], \\ \boldsymbol{S}_\perp &= s\sin(\alpha)(-1)^{N(\boldsymbol{r})}\boldsymbol{e}_3, \qquad (\boldsymbol{e}_i\boldsymbol{e}_j) = \delta_{ij}. \end{aligned} \tag{3}$$

The triad of mutually orthogonal unit vectors $\boldsymbol{e}_{1,2,3}$ and the angle α depend on the coordinate $\boldsymbol{r}$ and change slowly over the lattice distance a_0. To be definite, we choose the antiferromagnetic Heisenberg exchange on a bipartite lattice such that $N(\boldsymbol{r})$ is a sum of all lattice coordinates for a given site.

We are interested in the state where $\sin(\alpha)$ acquires a nonzero average below some transition temperature and the triad of vectors $\boldsymbol{e}_{1,2,3}$ remains disordered, at least at finite temperatures. As we shall see, the fluctuations of angle α always remain massive. Its mean value will be found from minimizing the free energy.

To calculate the ground state energy, we first neglect space variations of the $\boldsymbol{e}_i$ vector fields and integrate out the electrons [48]. We will comment on the space variations below while deriving the Landau free energy for the fluctuations. The spin configuration Eq. (3) gaps out only half of the electronic modes and another half remains gapless. A similar effect has been predicted by us for the 1D Kondo lattice where the anisotropy is of the easy-plane type and one helical sector of the fermions is gapped [44,46]. However, in the SU(2)-symmetric system, the axis of the spin spiral fluctuates in space which does not allow a global identification of gapped and gapless fermionic modes. The density of the ground state energy for the uniform and static configuration reads as

$$\begin{aligned} E_0/s^2 &= J_H\sum_{\boldsymbol{a}}(-1)^{N(\boldsymbol{a})}\sin^2(\alpha) \\ &\quad + \cos^2(\alpha)\{\tilde{J}_H(\boldsymbol{Q}) - \rho(\epsilon_F)J_K^2\ln[D/|sJ_K\cos(\alpha)|]\}, \end{aligned} \tag{4}$$

where $\tilde{J}_H(\boldsymbol{q}) = J_H\sum_{\boldsymbol{a}}\cos(\boldsymbol{q}\cdot\boldsymbol{a})$ is the Fourier transform of the Heisenberg exchange interaction, $\rho(\epsilon_F)$ is the density of states (per one unit cell of the lattice) at the Fermi energy. We emphasize that, if the Fermi surface is nested, the specific choice of the dispersion relation has an influence only on $\rho(\epsilon_F)$, but neither the structure of Eq. (4) nor its further analysis depends on details of $\epsilon(\boldsymbol{k})$. In the case of a square 2D lattice, $(-1)^{N(\boldsymbol{a})} = -1$ such that $J_H\sum_{\boldsymbol{a}}(-1)^{N(\boldsymbol{a})}$ simplifies to $\tilde{J}_H(\boldsymbol{G})$ with $\boldsymbol{G} = \{\pi/a, \pi/a\}$. We will use the contracted notation $\tilde{J}_H(\boldsymbol{G})$ for $J_H\sum_{\boldsymbol{a}}(-1)^{N(\boldsymbol{a})}$, implying that $\tilde{J}_H(\boldsymbol{G}) < 0$.

$E_0(\alpha)$ has three extrema, one at $\alpha = 0$ and the other two at α defined by the following equation:

$$|\cos\alpha| = \mathcal{C}(J_H) \equiv \frac{e^{-1/2}D}{s|J_K|}\exp\left[\frac{\tilde{J}_H(\boldsymbol{G}) - \tilde{J}_H(\boldsymbol{Q})}{\rho(\epsilon_F)J_K^2}\right]. \tag{5}$$

The fluctuations of α are massive in both cases. Since $|\cos(\alpha)| \le 1$, the nontrivial minimum defined in Eq. (5) appears only at sufficiently strong J_H. The critical value can be found from the equation

$$\mathcal{C}(J_c) = 1 \Rightarrow J_c \sim \rho(\epsilon_F)J_K^2\log(D/s|J_K|). \tag{6}$$

If $J_H < J_c$, the minimum of the energy is located at $\alpha = 0$ and the system is in the disordered phase with $\mathcal{O}_c = 0$ [49]. When $J_H > J_c$, the effective potential Eq. (4) has two equivalent minima corresponding to different signs of $\alpha \neq 0$ defining signs of the finite SCO parameter; see Eq. (9). This corresponds to the CSL phase. Since the vacuum is doubly degenerate, the SCO parameter at $T = 0$ reflects broken Z_2 symmetry and there is an Ising-like phase transition at finite temperature T_c. We can estimate T_c by the height of the potential barrier in the effective potential Eq. (4):

$$T_c \sim E_0|_{\cos(\alpha)=1} - E_0|_{\cos(\alpha)=\langle\cos(\alpha)\rangle}, \qquad J_H > J_c. \tag{7}$$

For J_H close to J_c, Eq. (7) simplifies to

$$T_c \sim \rho^{-1}(\epsilon_F)[(J_H - J_c)/J_K]^2. \tag{8}$$

At $T < T_c(J_H)$ and $J_H > J_c$, $\mathcal{O}_c$ acquires the finite value [49]:

$$\mathcal{O}_c = s^3\langle \sin[\alpha(\boldsymbol{r})]\cos[\alpha(\boldsymbol{r})]^2\rangle[(-1)^{N(\boldsymbol{r}_3)}\sin(\Delta_{12}) + (-1)^{N(\boldsymbol{r}_1)}\sin(\Delta_{23}) + (-1)^{N(\boldsymbol{r}_2)}\sin(\Delta_{31})],$$
$$\Delta_{jj'} \equiv (\boldsymbol{Q}, \boldsymbol{r}_j - \boldsymbol{r}_{j'}). \quad (9)$$

To describe fluctuations of the vector fields, we have to integrate over the fermions and make a usual gradient expansion keeping only leading terms [50]. This yields the Landau free-energy density for the disordered and for the chiral phases. At low temperatures and on the square 2D lattice, we obtain

$$\mathcal{F} = \frac{1}{8}\sum_{j=1,2,3}\sum_{\nu=x,y}\mathcal{R}_{j,\nu}(\partial_\nu \boldsymbol{e}_j)^2, \qquad (\boldsymbol{e}_i, \boldsymbol{e}_j) = \delta_{ij},$$
$$\mathcal{R}_{1,\nu} = \rho(\epsilon_F)v_x^2\delta_{\nu,x} - 2\langle\cos^2(\alpha)\rangle(sa_0)^2 J_H \cos(\boldsymbol{Q}\cdot\boldsymbol{a}_\nu),$$
$$\mathcal{R}_{2,\nu} = \mathcal{R}_{1,\nu},$$
$$\mathcal{R}_{3,\nu} = \rho(\epsilon_F)v_x^2\delta_{\nu,x} - 2\langle\sin^2(\alpha)\rangle(sa_0)^2\tilde{J}_H(\boldsymbol{G}), \quad (10)$$

where v_x is the x projection of the Fermi velocity. The stiffness tensor $\mathcal{R}_{j,\nu}$ is generically anisotropic. Its anisotropy is not universal and depends, in particular, on a specific choice of $\epsilon(\boldsymbol{k})$ and on temperature.

Equation (10) has a form of a nonlinear σ model with the symmetry $SU(2) \times U(1)$. Similar σ models were studied in the context of noncollinear antiferromagnetism [51–53]. Nonlinearity of the theory Eq. (10) comes from the orthonormality of the vectors $\boldsymbol{e}_j$. In 2D, this interaction generates a finite correlation length ξ [54]. In the renormalization procedure, this manifests itself as a continuous decrease of the stiffness components $\mathcal{R}_{j,\nu}(\Lambda)$ with the decrease of the momentum cutoff Λ. As a result, the fluctuations acquire a correlation length which is exponentially large in $\mathcal{E}_{\rm UV}/T$; $\mathcal{E}_{\rm UV}$ is the UV regularizer [55,56].

We consider the finite temperatures implying that thermal fluctuations dominate over the quantum ones at length scales $L > \xi > v/T$, where v is a characteristic velocity of the spin excitations. In this case, one can treat the fields $\boldsymbol{e}_i$ as time independent and there is no need to promote the free-energy description to the full dynamical theory. The thermal fluctuations prevent a breaking of the SU(2) symmetry of Eq. (10) and the magnetic order can occur only at $T = 0$; see Fig. 3. This leaves us with SCO as the only possible order at $J_H > J_c$ and $T \neq 0$.

One has to distinguish two regimes where Eq. (10) can be used. (1) The model with $\alpha = 0$ corresponds to the disordered phase and can be used in the temperature interval between the Ising transition temperature and the fermionic gap: $T_c \ll T \ll sJ_K$. (2) The model with $\alpha \neq 0$ corresponds to CSL and should be used well below the Ising transition, $T_{\rm min} < T \ll T_c$, where one can neglect fluctuations of $\langle \sin\alpha\rangle$.

Although all quantum effects in CSL are very interesting, we leave their systematic study for a forthcoming paper. At present, we can make only a preliminary guess: We note that the charge and the spin degrees of freedom are deeply connected in our approach [57]. The Kondo lattice model considered in Refs. [44,46] has the same property. Based on this analogy and on the fully quantum theory of Refs. [44,46], we surmise that nontrivial excitation of the KHS are slow spinons dressed by localized electrons.

To summarize, we have found that increasing the direct Heisenberg exchange in the Kondo-Heisenberg model with the nested Fermi surface leads to a phase transition to the state with spontaneously broken scalar chirality. The corresponding chiral order parameter, $\mathcal{O}_c$ in Eq. (1), breaks time-reversal and parity symmetry. This symmetry is Z_2 and the transition belongs to the universality class of the Ising model.

We believe that KHS can be used as a principally new platform to realize SCO in nonexotic experimental setups. Our finding paves the way towards removing the doubt of whether the chiral spin liquid with the scalar chirality can exist in the realistic systems where the time-reversal symmetry is not explicitly broken.

Broken time-reversal and parity symmetries can reveal themselves in the optical measurements through, for instance, the Kerr effect or measurements of nonlinear optical responses. The second harmonic response is particularly sensitive to the presence of global inversion symmetry. There are two other, though not definite, experimentally detectable indicators which can complement the optical experiments and confirm formation of CSL, namely, peculiar magnetic and electronic responses of the antiferromagnetic KHS with the nested Fermi surface. Firstly, the energetically favorable spin configuration, Eq. (3), suggests that correlation functions of all spin components have $\boldsymbol{Q}$ harmonics. Therefore, spin susceptibilities possess the Bragg peaks not only on the Néel vector but also on the wave vectors $\pm\boldsymbol{Q}$. These new peaks are smeared out by smooth fluctuations of the spin $\boldsymbol{Q}$ components, including the fluctuations of the triad $\boldsymbol{e}_{1,2,3}$ and of the angle α. The triad fluctuations are (almost) insensitive to the Ising phase transition at $J_H > J_c$, $T \to T_c$. However, the fluctuations of α are suppressed in the CSL phase and, therefore, the peaks become sharper at $J_H > J_c$, $T < T_c$. On the other hand, the response of the itinerant electrons will experience a drop when the probe frequency and the temperature are below $sJ_K > T_c$. Such a drop is related to the fact that one half of the electrons acquire a gap while the other half remain gapless. This decrease in the number of carriers is expected to alter the electric properties of a sample, cf. Ref. [58]; more details will be presented elsewhere. Thus, the full characterization of CSL predicted in the present Letter can be achieved via a combination of the optical measurements with measurements of the spin and electron responses at various temperatures.

A model described by the Kondo part of our Hamiltonian, $\hat{H} = \hat{H}_K$ at $J_H = 0$, has been considered in Ref. [59] on a triangular lattice. It has been demonstrated that, for a particular band filling providing two independent nesting vectors of the Fermi surface, the chiral order is formed. We would like to stress that our approach is much more general and does not require any special fine-tuning. In particular, details of the band dispersion are not important for our general predictions. The only crucial ingredient is the strong maximum of the spin susceptibility of the itinerant electrons. A nested Fermi surface is just a simple way to achieve it and should not be considered as a strict requirement for our theory imposing restrictions on its experimental verification. Possible candidates for the experimental realization of KHS with the spontaneously broken chirality are proximity-coupled layers of metals and Mott insulators. At present, we know at least one system which is structurally similar to what we propose. This is Sr_2VO_3FeAs [60], a naturally assembled heterostructure made of well-separated layers of an iron-based metal SrFeAs and Mott-insulating vanadium oxide. One can search suitable materials among similar systems.

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