A Spin-Orbital Singlet and Quantum Critical Point on the Diamond Lattice: FeSc$_2$S$_4$

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We present a theory of spin and orbital physics in the A-site spinel compound FeSc$_2$S$_4$, which experimentally exhibits a broad “spin-orbital liquid” regime. A spin-orbital Hamiltonian is derived from a combination of microscopic consideration and symmetry analysis. We demonstrate a keen competition between spin-orbit interactions, which favor formation of a local “Spin-Orbital Singlet” (SOS), and exchange, which favors magnetic and orbital ordering. Separating the SOS from the ordered state is a quantum critical point (QCP). We argue that FeSc$_2$S$_4$ is close to this QCP on the SOS side. The full phase diagram of the model includes a commensurate-incommensurate transition within the ordered phase. A variety of comparison to and suggestion for experiments are discussed.

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The search for quantum spin liquids – materials in which local moments are well formed but continue to fluctuate quantum mechanically down to zero temperature – is a fundamental challenge in condensed matter physics. In transition metal compounds with relatively high (e.g. cubic) symmetry, a richer possibility has also been contemplated, in which not only spin but also orbital states of localized electrons fluctuate. Such a “Spin Orbital Liquid” (SOL) was proposed in LaTiO$_3$ [1]. Probably the best candidate for a SOL is FeSc$_2$S$_4$ a spinel compound (with the general structure AB$_2$X$_4$), in which only the A sites form a magnetically/orbitally active diamond sublattice. In recent years, a variety of such A-site spinels, e.g. CoAl$_2$O$_4$ and MnSc$_2$S$_4$, were also found to be frustrated [2, 3, 4, 5, 6], forming a “spiral spin liquid” [7] at certain temperature range. FeSc$_2$S$_4$ stands out markedly amongst this class of compounds in exhibiting a much broader liquid regime, extending down to the lowest measured temperatures.

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In the letter we describe a theoretical study of the spin and orbital physics of FeSc$_2$S$_4$ [8, 9, 10, 11, 12]. Here, the magnetic Fe$^{2+}$ ion is in a 3$d^6$ configuration, with a local $S = 2$ moment as well as a two-fold orbital degeneracy (associated with one hole in the $e_g$ doublet), i.e. Fe$^{2+}$ is Jahn-Teller active. However, no indication of long range spin or orbital order is detected down to 50mK, which makes the frustration parameter $f \gtrsim 1000$ ($f \equiv \Theta_{\text{CW}}/T_N$ and $\Theta_{\text{CW}} = -45.1$K is the Curie-Weiss temperature), one of the largest values ever observed. We argue that the unusual behavior of FeSc$_2$S$_4$ arises from proximity to a Quantum Critical Point (QCP) between a spin-orbital singlet phase induced by atomic spin-orbit coupling and a magnetically and orbitally ordered phase favored by exchange (see Fig. 1).

The spin-orbital Hamiltonian in FeSc$_2$S$_4$ — The naïvely large (10-fold) spin and orbital degeneracy of the isolated Fe$^{2+}$ ions must be split by additional effects. One natural mechanism is exchange. To study this problem, we will first derive a “Kugel-Khomskii” type spin-orbital exchange Hamiltonian [13], in which the spin and orbital state on each Fe$^{2+}$ are described by spin $S_i$ ($S = 2$) and pseudo-spin $\tau_i$ ($\tau = 1/2$). The study of a structurally identical A-site spinel compound MnSc$_2$S$_4$ (no orbital degeneracy) indicates [7, 14, 15] that a minimal superexchange Hamiltonian should include both the nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions. Unfortunately, direct derivation of the superexchange Hamiltonian from the “parent” Hubbard model is not very practical due to the complicated exchange paths A-X-B-X-A [16] which involve five ions for both NNs and NNNs. However, the Fd3m space group symmetry of FeSc$_2$S$_4$ rather strongly constrains the exchange Hamiltonian. One can show that the most general spin-orbital exchange Hamiltonian allowed by symmetry for NNs and NNNs has the following simple form (neglecting the spin anisotropic terms),

$$
H_{ex} = \frac{1}{2} \sum_{ij} [J_{ij} S_i \cdot S_j + 8K_{ij} \tau_i \cdot \tau_j + \tilde{K}_{ij} \tau_i^x \tau_j^y + (L_{ij} \tau_i \cdot \tau_j + \tilde{L}_{ij} \tau_i^y \tau_j^x)] S_i \cdot S_j,
$$

(1)

FIG. 1: (color online). The schematic phase diagram versus $T$, temperature and $x$, the ratio of exchange to spin orbit interaction (see text). Here the labels for the phases are: “SOS”, spin-orbital-singlet; “CAF+OO”, commensurate antiferromagnet with orbital order; “IC”, incommensurate spin and orbital order. “QC” stands for the quantum critical regime.
where $J_{ij} = J_1$ or $J_2$ when $ij$ are NN and NNN sites, respectively (and similarly for $K_{ij}$, $L_{ij}$, $L_{ij}$).

To further constraint the couplings, we must treat the microscopic physics. Integrating out the intermediate states on B and X sites, we consider processes involving electron motion between two Fe$^{2+}$ ions. In general, superexchange may occur between electrons from either the $t_{2g}$ or $e_g$ levels. Only processes involving the latter, however, distinguish the orbital pseudo-spin. Now note that in Eq. (1), there are no terms linear in the pseudo-spin operators $\tau$. Therefore, microscopic processes which might individually contribute to such terms for a single pair of Fe$^{2+}$ ions must cancel when summed together. Orbital contributions from superexchange processes between $e_g$ and $t_{2g}$ are precisely of this form, and can therefore be neglected. Thus to obtain the orbital part of the exchange Hamiltonian, we only need to focus on terms involving motion of two $e_g$ electrons. Again, symmetry restricts the form of Hamiltonian describing the effective transfer between NNs and NNNs:

$$H_t = \sum_{ij} \sum_{m\sigma} t_{ij} d^\dagger_{ima} d_{jms} ,$$  
(2)

where $d^\dagger_{ima}$ and $d_{ima}$ are the creation and annihilation operators of a hole at $m$th $e_g$ level with spin $\sigma$, and $t_{ij} = t_1, t_2$ when $i, j$ are NNs and NNNs, respectively. Together with the on-site Hubbard-U terms, one can derive the pseudo-spin part of the exchange Hamiltonian by the standard perturbation theory. Combined with the contribution to the spin part from the $t_{2g}$-$t_{2g}$ and $t_{2g}$-$e_g$ exchange, we obtain a simplified version of Eq. (1) with

$$K_{ij} = \tilde{L}_{ij} = 0, \quad L_{ij} = 2K_{ij},$$  
(3)

with $K_{1,2} = t_{1,2}^2/(4U) > 0$. The simplified Hamiltonian contains 4 dimensionful parameters ($J_1, J_2, K_1, K_2$). Moreover, from the above analysis, we expect crudely $J_{1,2} \gg K_{1,2}$ as the $K$’s only come from the $e_g$-$e_g$ exchange.

The exchange Hamiltonian $H_{ex}$ has spin-rotational symmetry. A second mechanism to split the large ionic degeneracy is spin-orbit coupling. For an isolated Fe$^{2+}$ ion, the physical spin and pseudo-spin can couple via the symmetry allowed term [17],

$$H_0^x = -\frac{\lambda}{3} \sqrt{3} r_i^x \left[(S_i^x)^2 - (S_i^y)^2\right] + r_i^z \left[3(S_i^z)^2 - S_i^2\right].$$  
(4)

Note that, for an isolated single Fe$^{2+}$ ion, $H_0^x$ results in a non-degenerate ground state with a gap $\lambda$ to the first excited triplet. The ground state is a highly entangled state of spin and orbital degrees of freedom: a “spin orbital singlet”. It is remarkable that a 3d ion with large spin $S = 2$ can form such a highly entangled quantum ground state.

Such spin orbital singlet formation competes with exchange, so it is helpful to have a microscopic estimate of $\lambda$. This interaction arises at second order in the $LS$ spin-orbit interaction $\lambda_0(L \cdot S)$ due to the nonvanishing matrix elements of $L$ between $e_g$ and $t_{2g}$ orbitals. Standard second order perturbation theory gives [17] $\lambda = 6\lambda_0^2/\Delta_{e} > 0$, with $\Delta$ the crystal field splitting between $e_g$ and $t_{2g}$ levels. Taking the atomic spin-orbit coupling constant $|\lambda_0| \approx 100$cm$^{-1}$ and $\Delta_e \approx 2500$cm$^{-1}$ [18][19] yields $\lambda \approx 36K$. It is noteworthy that this is comparable to $\Theta_{CW}$. If we assume $K_1 = K_2 = 0$, by the high temperature mean field theory (including both $\sum H_0^i$ and $H_{ex}$) one finds $\Theta_{CW} = -\frac{\sqrt{3}(S_0^z)}{4J_1 + 12J_2}$. Thus $\Theta_{CW}$ is a measure of the strength of exchange, and we conclude that exchange and spin-orbit coupling are competitive in FeSc$_2$S$_4$.

From the above analysis, our complete Hamiltonian for FeSc$_2$S$_4$ is, in the first approximation

$$H = \sum_i H_{t}^i + H_{ex} .$$  
(5)

Minimal model for FeSc$_2$S$_4$ — We begin the analysis by considering a simplified exchange Hamiltonian by appealing to the neutron scattering measurements [9], which indicate the low energy magnetic excitations are localized near $k = 2\pi(1,0,0)$. This is precisely the ordering wavevector associated with a simple collinear Néel state on an FCC sublattice, and suggests the dominance of second neighbor exchange $J_2$. Therefore we begin by studying the “minimal model” with $J_1 = K_1 = K_2 = 0$ and $J_2 > 0$ antiferromagnetic. The two FCC sublattices of the diamond lattice decouple in this case, and $J_2$ can be viewed as a nearest-neighbor antiferromagnetic exchange within either of these sublattices. (Pathological effects of this decoupling can be removed by including very small $J_1$).

Here there is a single dimensionless parameter $x \equiv J_2/\lambda$. For $x \gg 1$, the exchange is dominant, and since the $S = 2$ spins are rather classical, we expect them to order magnetically. The ground states of the NN FCC sublattice are well-known. In real space, they consist of simple collinear antiferromagnetic Néel states within each $\{100\}$ plane, with an arbitrary choice of axis for each such plane. In momentum space, this allows for spiral states with wavevector $k = 2\pi(1,\delta, 0)$ (and symmetry-related wavevectors) with arbitrary $\delta$.

The $\lambda$ term splits this degeneracy. For an arbitrary magnetically ordered state, in which $\langle S_i \rangle \neq 0$, the spin orbital Hamiltonian Eq. (4) induces an “orbital field” that induces an orbital moment on each site. The magnitude of this orbital field is maximal when the spin is along one of the axial directions [100]. This selects commensurate states with $\delta = 0, 1/2$. Within the minimal model, the remaining degeneracy is lifted by the weak effects of quantum fluctuations [20], which favors collinear spin states. Note that this selects $\delta = 0$, which corresponds to the experimentally observed low energy excitations in FeSc$_2$S$_4$.

In contrast to the commensurate ordered phase (with collinear orbital order) found for $x \gg 1$, for $x \ll 1$, the ground state is a spin orbital singlet, with a gap to all excited states. This is a generalization of a “quantum paramagnet” discussed intensively in spin-only models. The gap decreases
where Eq. (5) has actually a singlet phase, according to numerous properties of the ideal QCP follow directly. The gap \( D \) constrain Eq. (9). This is an Euclidean multi-component \( \Phi \) field theory of a free relativistic scalar field, up to logarithmic corrections. A comparison with known experimental results is given at the end of this letter.

Having established the fundamental nature of the phase diagram and QCP, we turn to a discussion of more subtle effects.

\[
\langle S_i \rangle = m [\cos(\mathbf{p} \cdot \mathbf{r}_i) \hat{x} + \sin(\mathbf{p} \cdot \mathbf{r}_i) \hat{y}],
\]

with \( \mathbf{p} = 2\pi(1, 0, 0) \) or \( \mathbf{p} = 2\pi(1, 1/2, 0) \), we predict by mean field theory that at \( T = 0 \) a continuous second order transition occurs at \( x_c = 1/16 \), and the staggered magnetization vanishes for \( x > x_c \) like

\[
m \sim 8\sqrt{2(x - x_c)}. \tag{7}
\]

In the vicinity of the QCP, one may obtain a Landau expansion of the effective action by standard methods [21]. The order parameters are the (real) staggered magnetizations \( \Phi_a \) at wavevectors \( 2\pi\hat{x}, 2\pi\hat{y}, 2\pi\hat{z} \) (for \( a = 1, 2, 3 \)):

\[
\langle S_i \rangle = \sum_{a=1,2,3} \Phi_a (-1)^{x_i^a}, \tag{8}
\]

where the \( x_i^a \) are the usual half-integer coordinates of the FCC sites using a unit length conventional cubic unit cell. The symmetry-allowed form of the effective lagrangian (in imaginary time \( \tau \)) is

\[
\mathcal{L} = \sum_a \left[ \partial_\tau \Phi_a \right]^2 + v^2 |\nabla \Phi_a|^2 + r |\Phi_a|^2 \right]
+ \text{Sym} \left[ g_1 (\Phi_1^x)^4 + g_2 (\Phi_1^y)^2 (\Phi_1^z)^2 + g_3 (\Phi_1^z)^2 (\Phi_1^x)^2 \right] + g_4 (\Phi_1^y)^2 (\Phi_1^z)^2 + g_5 \Phi_1^x \Phi_1^y \Phi_1^z \Phi_1^y, \tag{9}
\]

where “Sym” indicates symmetrization with respect to both wavevector (lower) and spin (upper) indices, and we have for simplicity neglected presumably unimportant anisotropy of the gradient terms. Note that the effective Hamiltonian in Eq. 3 has actually independent cubic “internal” spin symmetry and cubic “external” space group symmetry, which both constrain Eq. 9.

This is an Euclidean multi-component \( \Phi^4 \) field theory of standard type, which in \( D = d + 1 = 4 \) space-time dimensions is in its upper critical dimension. Thus MFT is expected to be qualitatively correct, up to logarithmic corrections. Numerous properties of the ideal QCP follow directly. The gap \( \Delta \) vanishes upon approaching the QCP from the spin orbital singlet phase, according to \( \Delta \sim \sqrt{x_c - x} \). Similarly, the Néel temperature vanishes approaching from the other side, \( T_N \sim \sqrt{x - x_c} \). Other critical properties are readily obtained from the theory of a free relativistic scalar field, up to logarithmic corrections. A comparison with known experimental results is given at the end of this letter.

Having established the fundamental nature of the phase diagram and QCP, we turn to a discussion of more subtle effects.

Commensurate to incommensurate transition in the ordered phase — We first consider the effects of exchanges other than \( J_{1,2} \) in the ordered phase. Define now \( x \equiv \max\{J, K\}/\lambda \), where \( \{J, K\} \) denotes all exchange coupling constants \( J_1, J_2 \) and \( K_1, K_2 \). In the extreme limit \( x \gg 1 \), in which the spin orbit interaction can be neglected, one expects incommensurate magnetically and orbitally ordered ground states. If we assume the spins (pseudo-spins) a coplanar spiral with wavevector \( \mathbf{p} \) and phase shift \( \theta \) between the two fcc sublattices, we obtain 8 conditions for such a configuration to be a classical ground state:

\[
\left\{ \begin{array}{l}
|\Lambda(\mathbf{p})| = \frac{|J_1|}{\sqrt{J_{1,2}}} = \theta = \arg(\Lambda(\mathbf{p})));
|\Lambda(\mathbf{q})| = \frac{K_1}{\sqrt{K_{1,2}}} = \phi = \arg(\Lambda(\mathbf{q}));
|\Lambda(\mathbf{p} + \mathbf{q})| = \frac{K_1}{\sqrt{K_{1,2}}} = \theta + \phi = \arg(\Lambda(\mathbf{p} + \mathbf{q}));
|\Lambda(\mathbf{p} - \mathbf{q})| = \frac{K_1}{\sqrt{K_{1,2}}} = \theta - \phi = \arg(\Lambda(\mathbf{p} - \mathbf{q})).
\end{array} \right. \tag{10}
\]

Here the complex function \( \Lambda(\mathbf{p}) \) is defined as

\[
\Lambda(\mathbf{p}) = \cos \frac{p_x}{4} \cos \frac{p_y}{4} \cos \frac{p_z}{4} - i \sin \frac{p_x}{4} \sin \frac{p_y}{4} \sin \frac{p_z}{4}. \tag{11}
\]

In Eq. (10), after trivially eliminating \( \phi, \theta \), there are actually six independent equations which can completely determine the six real components of \( \mathbf{p} \) and \( \mathbf{q} \). When some coupling constants vanish, a variety of different degenerate classical ground states can be obtained [20]. For example, when \( K_1 = K_2 = 0 \), \( \mathcal{H}_{orb} \) reduces to the familiar \( J_1-J_2 \) model and the degenerate spin spiral momenta \( \mathbf{p}'s \) form the well-known spiral surface in momentum space [7][14][15].

When we turn on the spin-orbital interaction (\( \lambda \neq 0 \)), we expect the spin and orbital ordering to become commensurate with increasing \( x \). This is because, as remarked above, \( \mathcal{H}'_c \) has axial cubic anisotropy, and moreover, it favors orbital order with twice the momentum of the spin spiral. A general spin spiral for which all spins are axially oriented satisfies \( \mathbf{P} = \frac{\pi}{14}(n_1, n_2, n_3) \), with \( n_1, n_2, n_3 \) either all even integers or all odd integers. Assuming \( J_1/(8J_2) \) is not too large (as expected both from comparison with the structurally similar material MnSc\(_2\)S\(_4\), and from the aforementioned neutron data), the states with \( \mathbf{P} = 2\pi(1, 0, 0), 2\pi(1, \frac{1}{2}, 0) \) have low exchange energy, and are favored by the gain from the spin orbit interaction. Therefore we expect the ordered state to become commensurate for \( x_c < x < x_{c1} \) (see Fig. 1). For the \( J_1-J_2-\lambda \) model, by comparing the classical energies given by the incommensurate spiral momenta from the spiral surface \( \Lambda(\mathbf{p}) \) and the commensurate spiral momentum, we find

\[
x_{c1} = J_2/\lambda_{c1} \approx 0.61(J_2/J_1)^2. \tag{12}
\]

Including non-zero \( K_1 \) or \( K_2 \), the critical \( \lambda_{c1} \) is expected to be somewhat smaller than the one found in Eq. (12). Since \( J_2/J_1 \) is expected to be large, we have \( x_{c1} > x_c = 1/16 \).

Excitations in the spin orbital singlet phase — For small \( x \), deep in the disordered phase, one can obtain the excitation...
Theoretically, we expect a double crossover between ac-
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by NMR measurements of the
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is most natural for FeSc
which is consistent with
∆
minima of ω
where the
a
= 25(2
+ K2)
The solid curves are calculations from the small x expansion, with
λ = 25(J2 + K2).
The blue (dark) and green (light) curves have
J1 + K1 = 0 and J1 + K1 = J2 + K2, respectively. The red
dashed curve is a schematic spectrum close to the quantum critical
point, where it shows a Dirac-type structure at low energy.
spectrum as an expansion in the exchange. At the leading or-
der, we find the lowest-lying states form a triplet, with energy

\[ \omega(k) = \lambda + (2J2 + 2K2) \sum A \cos(A \cdot k) \]

\[ -|(2J1 + 2K1) \sum a \exp(iA \cdot k)|, \]  

(13)

where the a and A are summed over the 4 NN and 12 NNN lattice vectors, respectively. Along [100] direction, the energy minima of ω (which are also the global minima) are at \( k_x = \pm 4 \arccos \left( \frac{J1 + K1}{2(J2 + K2)} \right) \) (see Fig. 2). The energy gap (for |J1 + K1| ≤ 8(J2 + K2)) is

\[ \Delta = \lambda - 8(J2 + K2) - \frac{(J1 + K1)^2}{2(J2 + K2)}. \]  

(14)

Comparison with experiments — Perhaps the strongest indi-
cation of proximity to a quantum critical point in FeSc2S4 is in the neutron scattering experiments by Krimmel et al. A magnetic excitation is observed with a bandwidth exceeding 20K, in agreement with the expectations from the Curie-Weiss temperature ΘCW = 45.1K. However, a much smaller gap \( \Delta \approx 2K \) is observed near the minimum, the wavevector of which is consistent with k = 2π(100) which we have argued is most natural for FeSc2S4. A gap \( \Delta \approx 2K \) was also observed by NMR measurements of the 1/T1 relaxation of Sc nuclei. Theoretically, we expect a double crossover between ac-
tivated behavior for \( T \ll \Delta \) to linear behavior \( 1/T1 \sim T \) in the quantum critical regime \( \Delta \ll T \ll T0 \), where \( T0 \) is a cut-
of temperature of order ΘCW, and finally \( 1/T1 \sim \) const. for \( T \gg T0 \). The low and high temperature limits are clearly ob-
served, but the quantum critical behavior is not immediately apparent. The uniform magnetic susceptibility remains large at low temperature despite the gap, which we take as a strong indication of the importance of spin-orbit interaction, i.e. Eq. [4]. The specific heat exhibits approximate power-

law growth \( Cv(T) \approx AT^{2.5} \) for 0.2K < T < 2K, with a linear
term \( Cv(T) \sim \gamma T \) below 0.2K and more complex behavior
above 2K. Due to inversion disorder present in such spinels, we expect the very low temperature \( \gamma \) term is attributable to
two-level system defects, and the \( T^{2.5} \) behavior may represent a
crossover from this to the \( T^3 \) magnetic contribution expected
near the QCP.

This work suggests numerous future directions for theory
and experiment. Theoretically, the effects of fluctuations on
the critical properties, especially with \( J1 \neq 0 \), warrant more
detailed investigation, as do the effects of disorder — a rele-
vant perturbation at the QCP. More theoretical studies that pre-
dict experimental signatures are also warranted, such as possi-
bile signs of the spin-orbital singlet in the Jahn-Teller phonon
spectra. Experimentally, it would be most exciting to attempt
to drive FeSc2S4 past the QCP into an ordered state. This
could perhaps be accomplished by pressure, or with a suffi-
ciently strong applied magnetic field.

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