Triplet Resonating Valence Bond State and Superconductivity in Hund's Metals

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A central idea in strongly correlated systems is that doping a Mott insulator leads to a superconductor by transforming the resonating valence bonds (RVBs) into spin-singlet Cooper pairs. Here, we argue that a spin-triplet RVB (tRVB) state, driven by spatially, or orbitally anisotropic ferromagnetic interactions can provide the parent state for triplet superconductivity. We apply this idea to the iron-based superconductors, arguing that strong on site Hund's interactions develop intra-atomic tRVBs between the t_{2g} orbitals. On doping, the presence of two iron atoms per unit cell allows these interorbital triplets to coherently delocalize onto the Fermi surface, forming a fully gapped triplet superconductor. This mechanism gives rise to a unique staggered structure of on site pair correlations, detectable as an alternating π phase shift in a scanning Josephson tunneling microscope.

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Thirty years ago, Anderson proposed [1] the intriguing idea that the resonating valence bonds (RVBs) of a spin liquid could, on doping, provide the fabric for the development of unconventional superconductivity. A key aspect of the RVB theory is that it departs from weak-coupling approaches to superconductivity, positing that instead of a pairing glue, superconductivity develops from the entangled pairs already present in a spin liquid. RVB theory provides a natural account of the connection between d-wave pairing and antiferromagnetism [2] in almost-localized systems, a connection that has proven invaluable to the understanding of many families of superconductors, from the cuprate superconductors, to their miniature cousins, the 115 heavy-fermion compounds [3].

However, to date, there is no counterpart of RVB theory that applies to ferromagnetically correlated materials. There are a wide variety of unconventional superconductors which, to some extent or another, involve strong ferromagnetic (FM) spin correlations. Examples include uranium-based heavy fermion materials [4,5] that lie close to a FM quantum critical point, candidate low-dimensional triplet superconductors such as the Bechgaard salts [6], twisted double bilayer graphene [7,8], and various transition metal superconductors [9,10], notably the iron-based and ruthenate superconductors, which like Hund's metals involve strong local FM correlations between orbitals. Various papers have speculated that the Hund's interactions might provide the origin of the pairing in these systems [11–16].

Is there a ferromagnetic analog to the RVB pairing mechanism? Here we build on an observation [17] that magnetic anisotropy in a ferromagnet plays an analogous role to frustration in an antiferromagnet (AFM), generating a fluid of triplet resonating valence bonds (tRVBs). We propose that, like their singlet cousins, tRVB states can, on doping, lead to the development of triplet pairing. One of the exciting features of this idea is that tRVBs can form within the interior of Hund's coupled atoms, which under the right symmetry conditions [15,18] can coherently tunnel into the bulk to develop triplet superconductivity (Fig. 1) [19,20].

Consider an easy-plane FM interaction $H_{ij} = -J\vec{S}_i \cdot \vec{S}_j + \Delta J S_i^z S_j^z$, $(J > 0)$ between two spin-1/2 moments $\vec{S}_i$ and $\vec{S}_j$. In the Heisenberg limit ($\Delta J = 0$) and in the presence of a small symmetry breaking Weiss field, the ground state is a product state which lacks entanglement. Suppose the magnetization points in the x direction, the product ground state can then be written in terms of triplets,

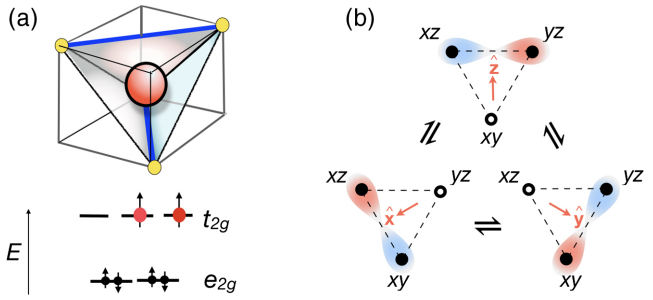

FIG. 1. (a) Isolated tetrahedron in iron-based superconductors, showing the two electrons forming a $S = 1$ triplet in the t_{2g} orbitals. (b) Triplet resonating valence bond (tRVB) as the ground state of a Hund's metal atom. The blue and red colors reflect the odd parity of the triplet pairs, while the red arrows denote the quantization axis (d vector) of the $m = 0$ triplet pair.

$$\frac{|\uparrow_i\rangle+|\downarrow_i\rangle}{\sqrt{2}}\frac{|\uparrow_j\rangle+|\downarrow_j\rangle}{\sqrt{2}}=\frac{|\uparrow_i\uparrow_j\rangle+|\downarrow_i\downarrow_j\rangle}{2}+\frac{|\uparrow_i\downarrow_j\rangle+|\downarrow_i\uparrow_j\rangle}{2}. \quad (1)$$

An easy-plane anisotropy ($\Delta J > 0$) projects out the equal-spin pairs on the right-hand side, stabilizing an entangled spin-1 ground state with $m_z = 0$. In the corresponding easy-plane ferromagnet, with Hamiltonian $H = \sum_{(i,j)} H_{ij}$, the intersite couplings preserve the $m_z = 0$ structure of the valence bonds, and the resulting ground state is a quantum superposition of triplet pairs which retains its ferromagnetic correlations, and may even exhibit long-range order [21,22].

Our interest in a tRVB ground state lies in its potential as a pre-entangled parent state of a triplet superconductor. In classic RVB theory, an antiferromagnetic superexchange interaction, is decoupled in terms of singlet pairs [23]:

$$J\vec{S}_i\cdot\vec{S}_j \equiv -\frac{J}{2}(\psi^\dagger_{i\uparrow}\psi^\dagger_{j\downarrow}-\psi^\dagger_{i\downarrow}\psi^\dagger_{j\uparrow})(\psi_{j\downarrow}\psi_{i\uparrow}-\psi_{j\uparrow}\psi_{i\downarrow}), \quad (2)$$

where we have used a fermionic representation of the spins, $\vec{S}_j = \psi^\dagger_j(\vec{\sigma}/2)\psi_j$. The corresponding relation for triplet valence bonds is obtained by rotating the spin coordinate system at site j through 180° about the z axis, which gives

$$-J_A(S^x_iS^x_j+S^y_iS^y_j-S^z_iS^z_j) \equiv -\frac{J_A}{2}(\psi^\dagger_{i\uparrow}\psi^\dagger_{j\downarrow}+\psi^\dagger_{i\downarrow}\psi^\dagger_{j\uparrow})(\psi_{j\downarrow}\psi_{i\uparrow}+\psi_{j\uparrow}\psi_{i\downarrow}), \quad (3)$$

demonstrating how xy anisotropy stabilizes a triplet pair.

The most direct application of the tRVB idea considers an easy-plane Heisenberg ferromagnet: by analogy with the singlet RVB pairing mechanism, doping with holes drives the formation of a triplet superconductor. On a square lattice, this scenario leads to a $p_x + ip_y$ triplet superconductor, to be presented elsewhere. A more dramatic possibility, in which i and j represent orbitals of a single atom, permits us to apply the tRVB idea to Hund's coupled metals. Here an application of particular current interest is as a theory for iron-based superconductors (FeSC).

The family of FeSC are characterized by high transition temperatures with a fully gapped Fermi surface. The presence of antiferromagnetic correlations and a marked Knight shift has led to the long-held assumption that these materials are spin singlet superconductors [9,24]. The recent observation [25] of a robust ratio $2\Delta/T_c \sim 7.2$ between the gap Δ and the transition temperature T_c across a broad range of FeSC motivates the search for a common pairing mechanism, one that is robust against the wide spectrum of Fermi surface morphologies, and hence most likely rooted in the local electronic structure of the iron atoms. Here, we propose that these systems are tRVB superconductors, with a fully gapped Fermi surface, an anisotropic Knight shift and an alternating pair wave function.

The symmetry properties of a Hund's coupled triplet superconductor were first considered by Anderson [15], who observed that in systems with a center of inversion, the odd-parity wave function of a triplet condensate prevents on site triplet pairing unless the lattice has an even number of atoms per unit cell, related to each other via inversion. In this situation, the odd-parity nature of the condensate means that the on site pair wave function reverses sign when reflected through the center of inversion

$$\langle\psi_{a\sigma}(\mathbf{x})\psi_{b\sigma'}(\mathbf{x})\rangle = -\langle\psi_{a\sigma}(-\mathbf{x})\psi_{b\sigma'}(-\mathbf{x})\rangle, \quad (4)$$

where a and σ are the orbital and spin indices, respectively. The key structural feature of FeSC is an iron atom enclosed in a tetrahedral cage of pnictogen or chalcogen atoms. The tetrahedra are packed in a checker-board arrangement, with a unit cell containing two iron atoms, separated by a common center of inversion, satisfying this requirement. We now show how tRVB predicts a condensate with the above properties.

In the parent compound of the FeSC, each tetrahedron contains two electrons within the three xz, yz, or xy orbitals of the t_{2g} level, Hund's coupled into a $S = 1$, $L = 1$ manifold. Consider the "atomic" limit of an isolated iron tetrahedron. Each pair of t_{2g} orbitals shares a common direction, for instance, the xz and yz orbitals share a common z axis, which in the presence of spin-orbit coupling causes [21] the Hund's interactions to develop an orbitally selective easy-plane anisotropy [Eq. (3)],

$$H_I = -2[(J_H+J_A)\vec{S}_{xz}\cdot\vec{S}_{yz}-2J_AS^z_{xz}S^z_{yz}) + (\text{cyclic permutations})]. \quad (5)$$

Each of the three interaction terms stabilizes a triplet pair with zero spin component along a quantization axis ("d vector") normal to its easy plane [see Fig. 1(c)], thus the xz and xy orbitals have d vector $\hat{d} = \hat{x}$.

With the convention $a \in \{xz, yz, xy\} = \{1, 2, 3\}$, the projected angular momentum operator within the t_{2g} subspace is $(L_a)_{bc} \equiv -ie_{abc}$. Defining the triplet pair creation operators $\Psi^\dagger_{ab} \equiv \psi^\dagger(L_a\sigma_b)\bar{\psi}^\dagger$, $a, b = 1, 2, 3$, where $\bar{\psi}^\dagger \equiv i\sigma_2(\psi^\dagger)^T$, Eq. (5) can be written as $H_I = -\sum_{ab} g_{ab}\Psi^\dagger_{ab}\Psi_{ab}$, with $g_{ab} = \frac{1}{4}(J_H + J_A\delta_{ab})$. In this way, we see that an anisotropy $J_A > 0$ splits off a ground state manifold of triplet pairs in which the orbital angular momenta and the spin quantization axis are aligned, $\Psi^\dagger_{aa}|0\rangle = \psi^\dagger(\sigma_aL_a)\bar{\psi}^\dagger|0\rangle$.

The spin-orbit coupling $H_{SL} = -\lambda\vec{L}\cdot\vec{S}$ causes the triplet valence bonds to resonate between orbitals, giving rise to a tRVB ground state $|\text{tRVB}\rangle = \sum_{ab}\Lambda^{ab}\Psi^\dagger_{ab}|0\rangle$ [see Fig. 1(b)]. Note that within the t_{2g} multiplet, the projected spin orbit interaction has a reversed coupling constant, with $\lambda > 0$, favoring $L + S = 2$ configurations.

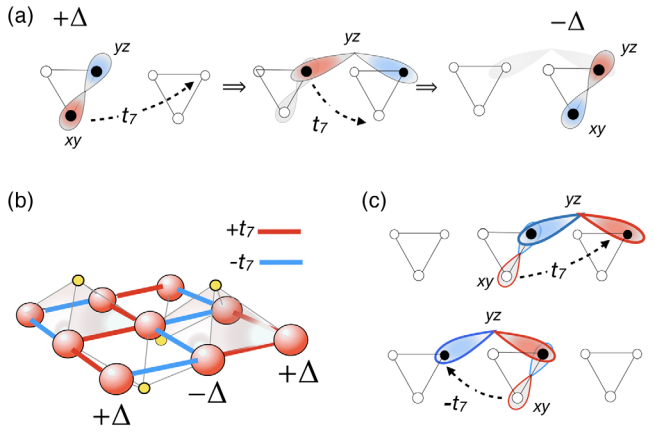

FIG. 2. Schematic showing (a) how tunneling of a triplet valence bond between two iron atoms leads to "tumbling" motion that reverses the on site triplet pair amplitude Δ on neighboring iron atoms, (b) the alternation in the sign of interorbital hopping t_7 and on site triplet pairing, (c) how the asymmetric left and right tunneling permits triplet pairs to align in the same direction between sites, allowing them to coherently condense into a p-wave state on the Fermi surface.

The structure of the resulting energy levels is modeled by a crystal field Hamiltonian given by $H = -\lambda(\vec{L}\cdot\vec{S}) - \alpha(J_x^4 + J_y^4 + J_z^4) + \eta J_z^2$, where $J = S + L$ is the total angular momentum, $\alpha \sim J_A$, while η quantifies the tetragonal anisotropy of the environment. The simplest tRVB ground state, where $\Lambda_{ab} = \delta_{ab}$ is a unit matrix, develops for the wrong sign of the spin-orbit coupling $\lambda < 0$. Two other tRVB states with $\Lambda_{ab} = \mathrm{diag}(1, -1, 0)$ and $\Lambda_{ab} = \mathrm{diag}(1, 1, -2)$ are stabilized for $\lambda > 0$, [21], where the latter becomes the unique ground state in the presence of a tetragonal anisotropy $\eta > 0$, see Fig. 1(b).

When the tetrahedra are brought together to form a conductor, charge fluctuations allow the escape of atomic triplet pairs into the conduction sea. We shall assume that the interactions present in the isolated tetrahedra are preserved in the metallic state that now develops. Imagine a lattice where the xy orbitals are weakly hybridized with the xz/yz orbitals at neighboring sites (we denote this amplitude as t_7). An on site valence bond between an xz and xy orbital can tunnel to the neighboring site in a two step process: an xz electron first hops to a neighboring xy orbital, forming an intersite, intraorbital triplet pair, after which the xy electron follows suit and hops onto the neighboring site to reassemble the intra-atomic triplet bond. In fact, the electrons can tunnel in either order and the resulting tumbling motion of the tRVB causes its amplitude to alternate at neighboring sites. If this process becomes coherent, it leads to a staggered anomalous triplet pairing amplitude [see Eq. (4)] $\Delta(\mathbf{x}) = -\Delta(-\mathbf{x})$ as envisioned in [15] [see Fig. 2(a)]. For this motion to be sustained coherently, there must be two atoms per unit cell. To understand how this works in the FeSC, we note there is an additional nonsymmorphic symmetry [26], under which the lattice is invariant under a glide and mirror reflection through the plane. The opposite parities of the xy and xz/yz orbitals under glide reflection, means that the interorbital tunneling amplitude t_7 *alternates* [see Fig. 2(b)]. When the xz/xy and yz/xy pairs tunnel left or right into the conduction sea, they do so with opposite amplitudes, causing the intersite, intraorbital triplet pairs to coherently condense in the same direction. This permits the phase-alternating tRVB pairs to coherently escape onto the Fermi surface [see Fig. 2(c)], activating a logarithmic Cooper divergence in the pair susceptibility. The nonsymmorphic symmetry of the FeSC allows us to absorb the staggered hopping into a staggered gauge transformation of the xz/yz orbitals [27], $\psi_{xz/yz}(\mathbf{j}) \to (-1)^{j_x+j_y}\psi_{xz/yz}(\mathbf{j})$. This transformation unfolds the Brillouin zone and allows to treat each iron atom on an equal footing.

Following [1] we introduce the simplest tRVB wave function as the Gutzwiller projection of a BCS-like wave function

$$|\mathrm{tRVB}\rangle = \hat{P}_G \prod_{\mathbf{k}} \exp\{\psi_{\mathbf{k}}^\dagger [\vec{\mathcal{L}}(k)\cdot\vec{\sigma}]\bar{\psi}_{-\mathbf{k}}^\dagger\}|0\rangle. \quad (6)$$

Here P_G is the Gutzwiller projector to $n < 2$ electron per site. The functions $\vec{\mathcal{L}} = \sum_g \vec{\Lambda}_g(\mathbf{k})\lambda_g$ with $g = 1, \ldots, 8$ can be expanded in the eightfold space of Gell-Mann matrices which span the t_{2g} multiplet. The triplet character of the condensate means that $\mathcal{L}(-\mathbf{k}) = -\mathcal{L}^T(\mathbf{k})$, so the three antisymmetric $\lambda_g \in \{L_a\}_{a=1}^3$ matrices combine with even parity functions $\Lambda_s(\mathbf{k}) = \Lambda_s(-\mathbf{k})$ to describe the on site, orbitally antisymmetric pairing, while the five symmetric λ_g, combine with odd-parity p-wave functions $\Lambda_a(\mathbf{k}) = -\Lambda_a(-\mathbf{k})$, to describe the tRVBs that have escaped to the Fermi surface.

To calculate the properties of the tRVB wave function, we adopt a Gutzwiller mean field approach, assuming that the action of the microscopic Hamiltonian beneath the projection operator P_G can be modeled by an appropriate renormalization of hopping matrix elements in a mean-field Hamiltonian. A microscopic rationale for these renormalizations can be obtained from a slave boson treatment of the unprojected Hamiltonian, along the lines of RVB theory [23,28]. Here we concentrate on the weak-coupling Cooper instability that arises from the renormalized Hamiltonian. Motivated by our discussion of the isolated tetrahedron, we now rewrite the Hund's interaction, Eq. (5) in the form of a BCS theory

$$H_I = \sum_{\mathbf{x},ab}\left[\frac{1}{g_{ab}}\bar{\Delta}_{ab}\Delta_{ab} + (\Psi_{ab}^\dagger\Delta_{ab} + \mathrm{H.c.})\right]. \quad (7)$$

For t_{2g} materials, the states at the Fermi surface are composed of three component Bloch wave functions $\vec{u}_{n,\mathbf{k}}$ which are eigenstates of the kinetic term $\mathcal{H}(\mathbf{k})\vec{u}_{n,\mathbf{k}} = \epsilon_n(\mathbf{k})\vec{u}_{n,\mathbf{k}}$.

On the Fermi surface, the band-diagonal matrix element of the gap function is given by $\vec{d}_{n\mathbf{k}} \cdot \vec{\sigma}$, where the d vector is $d^a_{n\mathbf{k}} \equiv \Delta_{ab}(\vec{u}^T_{n,-\mathbf{k}} L_b \vec{u}_{n,\mathbf{k}}) = -i\Delta_{ab}(\vec{u}_{n,-\mathbf{k}} \times \vec{u}_{n,\mathbf{k}})_b$. The d vector vanishes if the Bloch wave function $\vec{u}_{n-\mathbf{k}} = \vec{u}_{n\mathbf{k}}$ is symmetric, since $\vec{u}_{n,\mathbf{k}} \times \vec{u}_{n,\mathbf{k}} = 0$. Fortunately, the nonsymmorphic character of the lattice mixes the xy and xz/yz orbitals, so that $\vec{u}_{n\mathbf{k}} \neq \vec{u}_{n-\mathbf{k}}$, which allows the d vector to be finite.

The simplest mean-field theory, corresponding to $\Delta_{ab} = \Delta \mathrm{diag}(1, 1, -2)$, models the iron-based superconductors as a two-dimensional conductor with Hamiltonian

$$H_{\mathrm{MF}} = \sum_{\mathbf{k}} \tilde{\psi}^\dagger_{\mathbf{k}} [\mathcal{H}(\mathbf{k})\tau_3 + \Delta(\sigma_1 L_1 + \sigma_2 L_2 - 2\sigma_3 L_3)\tau_1] \tilde{\psi}_{\mathbf{k}} + \frac{V|\Delta|^2}{g}. \tag{8}$$

Here $\tilde{\psi}_{\mathbf{k}}$ is a Nambu spinor in the space of orbital, spin and charge (isospin) space. The pairing term $(\sigma_1 L_1 + \sigma_2 L_2)\tau_1$ term retains the essential tRVB pairing components that mix the xy and xz/yz orbitals at the Fermi surface and is sufficient to gap out the Fermi surface. In our two-dimensional model the component $\sigma_3 L_3 \tau_1$ has no weak-coupling support on the Fermi surface but induces interband pairing between xz and yz orbitals [13]. The term

$$\mathcal{H}(\mathbf{k}) = \epsilon_{\mathbf{k}} + \vec{e}_{\mathbf{k}} \cdot \vec{\gamma} = \left(\begin{array}{cc|c} a_{\mathbf{k}} & g_{\mathbf{k}} & ip_{k_x} \\ g_{\mathbf{k}} & b_{\mathbf{k}} & ip_{k_y} \\ \hline -ip_{k_y} & -ip_{k_x} & e_{\mathbf{k}} \end{array} \right) \tag{9}$$

describes the band dispersion [27], where $a_{\mathbf{k}} = 2t_1 c_x + 2t_2 c_y + 4t_3 c_x c_y - \mu$, $b_{\mathbf{k}} = 2t_2 c_x + 2t_1 c_y + 4t_3 c_x c_y - \mu$, $g_{\mathbf{k}} = 4t_4 s_x s_y$, $p_{k_x} = 2t_7 s_x + 4t_8 s_x c_y$, $p_{k_y} = 2t_7 s_y + 4t_8 s_y c_x$, and $e_{\mathbf{k}} = 2t_5(c_x + c_y) + 4t_6 c_x c_y - \mu + \delta_{xy}$, and we have employed the short-hand notation $c_l \equiv \cos k_l$ and $s_l = \sin k_l$ $(l = x, y)$.

Although the pairing in this mean-field theory is uniform, if we undo the gauge transformation of the xz/yz states, the on site pairing between the xy and xz/xy states acquires the staggered behavior predicted by Anderson. Remarkably, even though this order parameter is staggered, it induces a uniform gap on the Fermi surface, with a pair susceptibility that is logarithmically divergent at low temperatures.

Figures 3(a) and 3(b) display the spectrum calculated from the mean-field theory, Eq. (8), using the tight binding parameters of Ref. [27] and $t_8 = -t_7/3$. The ground state develops an anisotropic, yet full gap on the Fermi surface, which becomes increasingly isotropic with the introduction of spin-orbit coupling. Historically, the observation of a full gap [29–31] and the presence of a finite Knight shift in all field directions led to an early rejection of the idea of triplet

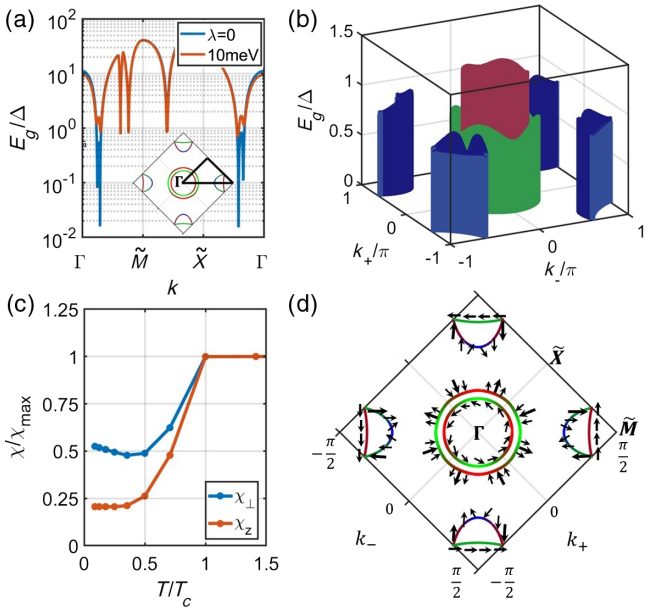

FIG. 3. (a) The size of the gap along a cut passing high-symmetry points in the Fermi surface (FS), for $\Delta = 6.2$ meV for $\lambda_{\mathrm{SO}} = 0$ and $\lambda = 10$ meV. The inset shows the folded Brillouin zone with $k_\pm = (k_x \pm k_y)/2$ and $\tilde{X} = (\pi, 0)$ and $\tilde{M} = (\pi/2, \pi/2)$. (b) The size of the gap on the FS for $\Delta = 6.2$ meV and $\lambda_{\mathrm{SO}} = 10$ meV. (c) The normalized spin-susceptibility at the transition for $\Delta = 6.2$ meV and $\lambda_{\mathrm{SO}} = 10$ meV [32]. (d) The winding of the $\vec{d}(\mathbf{k})$ vector along the FS for $\lambda_{\mathrm{SO}} = 0$ illustrates p-wave (E_u) pairing. Note that $\vec{d}$ vector is entirely in the plane in this case.

pairing in FeSC. However, the calculated Knight shift, obtained by summing both Fermi surface and interband components of the total spin and orbital susceptibility [Fig. 3(c)], shows a marked loss of spin susceptibility for all field directions, in accord with experiment. We note that in a two-dimensional model, the staggered hopping t_7 that delocalizes the pairs is only present in the basal plane. When motion in the c axis is included, the additional staggered hopping along the c axis will now hybridize the xz/yz orbitals, introducing an additional p_z component to the condensate, further reducing the predicted anisotropy.

Various other aspects of the tRVB theory of pairing in FeSC deserve discussion. First, since the Hund's triplet pairing occurs locally on the iron atom (unlike, $s_\pm$ pairing), tRVB accounts for intra-atomic Coulomb repulsion without relying on a cancellation between electron and hole pockets [33]. Second, because this pairing is local, it is expected to be moderately robust against the pair breaking effects of impurity scattering. Microscopically, disorder generates nonzero vertex corrections to the local pair which partially cancel the disorder induced self-energy [21], thereby reducing the pair-breaking effects of disorder. Third, there are multiple sign changes of the triplet d vectors on and in between the various Fermi surfaces [Fig. 3(d)]. The finite winding number of the d vector around each pocket may

lead to interesting topological behavior [34,35]. At the same time the relative sign between d vectors on electron and hole pockets gives rise to quasiparticle coherence factors which closely resemble those of an s_{+-} superconductor with important consequences for quasiparticle interference (QPI) [36–39] and neutron spin resonance measurements. Specifically, the dominant Fermi surface contribution to the antisymmetrized tunneling density of states at wave vector $\mathbf{q}$ is proportional to the Fermi surface (FS) average $\langle 1 - \hat{d}_n(\mathbf{k}+\mathbf{q})\hat{d}_m(\mathbf{k})\rangle_{\mathbf{k}\in \mathrm{FS}}$, with $\hat{d} = \vec{d}/|\vec{d}|$. Features in this observable were previously interpreted as evidence for $s_{\pm}$ pairing, but our calculation suggests that tRVB is also consistent. A more detailed expression and a discussion of a similar feature on the subgap spin resonance [40] are relegated to Ref. [21].

A key feature of tRVB is the prediction that Hund's pairing will give rise to a unique staggered structure of the on site pair correlations. To observe this phenomenon in FeSC and other candidate materials, we propose a novel STM experiment, a "Josephson tunneling microscope" that employs two superconducting tunneling tips of the same tRVB material, one fixed, the other mobile, connected to form a SQUID with the sample. The alternation of the superconducting phase between neighboring iron atoms is predicted to lead to a staggered π-junction behavior as the mobile tip scans across the material [21].

Finally, we mention the possible relevance of tRVB to other superconductors of current interest. The recent discovery of the heavy-fermion UTe_2, which has an even number of uranium atoms per unit cell, with likely triplet superconductivity [41] is one promising example. Another intriguing candidate material is magic angle double bilayer graphene, where the valley degrees of freedom play the role of orbitals, giving rise to Hund's coupled interorbital triplet pairing [42] on a moiré superlattice.

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