Nature of unconventional pairing in the kagome superconductors AV$_3$Sb$_5$

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The recent discovery of AV$_3$Sb$_5$ (A=K,Rb,Cs) has uncovered an intriguing arena for exotic Fermi surface instabilities in a kagome metal. Among them, superconductivity is found in the vicinity of multiple van Hove singularities, exhibiting indications of unconventional pairing. We show that the sublattice interference mechanism is central to understanding the formation of superconductivity in a kagome metal. Starting from an appropriately chosen minimal tight-binding model with multiple van Hove singularities close to the Fermi level for AV$_3$Sb$_5$, we provide a random phase approximation analysis of superconducting instabilities. Non-local Coulomb repulsion, the sublattice profile of the van Hove bands, and the interaction strength turn out to be the crucial parameters to determine the preferred pairing symmetry. Implications for potentially topological surface states are discussed, along with a proposal for additional measurements to pin down the nature of superconductivity in AV$_3$Sb$_5$.

Introduction. The kagome lattice has become a paradigmatic setting for exotic quantum phenomena of electronic matter. This particularly applies to quantum magnetism, where the large geometric spin frustration inherent to the corner-sharing triangles promotes the emergence of extraordinary quantum phases [1]. From an inherent limit, electronic kagome bands are likewise particular, as they feature a flat band, Dirac cones, and van Hove singularities at different fillings. The kagome flat band suggests itself as a natural host for the realization of ferromagnetism [2, 3] or possibly non-trivial topology [4–7], while the kagome Dirac cones have been proposed to be a promising way to accomplish strongly correlated Dirac fermions [8] and turbulent hydrodynamic electronic flow [9]. The kagome lattice at van Hove filling has been shown to be preeminently suited for the emergence of exotic Fermi surface instabilities [10–13]. Among others, this involves charge and spin density-wave orders with finite relative angular momentum [14]. Moreover, the kagome Hubbard model was first predicted to yield degenerate nematic instabilities which can break point-group and time-reversal symmetry simultaneously [13], which has currently regained attention in the context of twisted bilayer graphene [15].

The recent discovery of AV$_3$Sb$_5$ [16] provides an instance of kagome metals tuned to the vicinity of multiple van Hove singularities. What further makes them unique is the combination of metallicity, strong two-dimensional electronic character, and significant electronic correlations derived from the d-orbital structure of the Vanadium kagome net. KV$_3$Sb$_5$ was discovered to be a kagome superconductor with $T_c = 0.93$ K [17], along with RbV$_3$Sb$_5$ ($T_c = 0.92$ K) [18] and CsV$_3$Sb$_5$ ($T_c = 2.5$ K) [19, 20], where the latter was shown to rise up to $T_c = 8$ K under 2 GPa hydrostatic pressure [21–23]. While the wheel of experimental exploration is still in spin, certain tendencies about the superconducting phase are starting to crystallize. The observed charge density wave (CDW) order [24], interpreted as a potential parent state for unconventional superconducting order [13, 25–27], exhibits indications for an electronically driven formation [28]. Specific-heat measurements suggest at least a strongly anisotropic gap [17]. While a significant residual term from thermal conductivity suggests a nodal gap [29], penetration depth measurements claim a nodeless gap [30]. The dome shape suggests unconventional superconductivity along with a large value of $2\Delta/k_B T_c$, hinting at a strong-coupling superconductor [31].

In this Letter, we formulate a theory of unconventional superconductivity in AV$_3$Sb$_5$. In a first step, we develop an effective tight-binding model suitable for the analysis of pairing instabilities. In order to retain the necessary complexity of multiple van Hove singularities in the
vicinity of the Fermi level in AV$_3$Sb$_5$, we distill a six-band minimal model. In a second step, we specify the interaction Hamiltonian. Due to matrix elements implied by the sublattice interference mechanism [11], which we review below, it is essential to take non-local Coulomb repulsion into consideration. Over a large range of coupling strengths, we find dominant $f$-wave triplet superconducting order, succeeded by $d$-wave singlet pairing for stronger coupling. Throughout the phase diagram, the $p$-wave order stays subdominant but competitive. Aside from this general trend, the detailed competition between the different orders is crucially influenced by the location of the Fermi level with respect to the multiple van Hove singularities and the nearest-neighbor (NN) Coulomb repulsion.

**Sublattice decoration of kagome van Hove points.** As opposed to related hexagonal van Hove singularities such as for the bipartite honeycomb lattice, the kagome bands can host two different types of van Hove singularities which we label as sublattice mixing (m-type) and sublattice pure (p-type), characterized by odd and even parity at the M point, respectively. This is illustrated in Fig. 1 for the minimal kagome tight-binding model with three distinct sublattice sites located on the 3f Wyckoff positions of the $P6/mmm$ space group. The upper van Hove singularity ($E = 0$) is of p-type, since the Fermi level eigenstates in the vicinity of the three $M$ points are localized on mutually different sublattices (left inset). By contrast, the lower van Hove filling ($E = -2\tau$) has mixed sublattice character and thus is of m-type, with the eigenstates equally distributed over mutually different sets of two sublattices for each $M$ point (right inset). These distinct sublattice decorations have a strong impact on the nesting properties (see Sec. II of supplementary materials (SM) [32]). Since p-type van Hove points do not couple to each other via local interactions, the inclusion of at least NN Coulomb repulsion is quintessential to adequately model interacting kagome metals close to p-type van Hove filling [11, 33].

**Effective model.** The ab-initio band structure of AV$_3$Sb$_5$ matches well with ARPES measurements below the CDW transition temperature, even though the corresponding density functional theory (DFT) calculations are performed neglecting the star-of-David-type structural distortion [19, 34]. Due to the multiple sublattices and the large number of contributing orbitals from both V and Sb in the vicinity of the Fermi level, a reduction to an effective model is a prerequisite to any analysis of many-body instabilities.

The layered structure of AV$_3$Sb$_5$, together with the large transport anisotropy of $\rho_{c}/\rho_{ab} \approx 600$ [19] (for CsV$_2$Sb$_5$) allows us to constrain ourselves to the two-dimensional V-Sb kagome plane. Analyzing the Fermi level at $k_z = 0$ by means of density functional theory, we find three distinct Fermi surfaces in AV$_3$Sb$_5$: (i) a pocket composed of Vanadium $d_{xy}, d_{xz-zy}, d_{x^2-y^2}$ orbitals in proximity to a $p$-type van Hove singularity, (ii) two additional pockets composed of Vanadium $d_{zx,zy}$ orbitals in proximity to another $p$-type and $m$-type van Hove singularity above and below the Fermi level, respectively (Fig. 2), and (iii) a circular pocket around $\Gamma$ formed by Antimony $p_z$-orbitals. Note that (i) and (ii) do not hybridize due to opposite $M_z$ eigenvalues and the symmetry-wise allowed hybridization of (ii) and (iii) is parametrically weak. These features are not particularly sensitive to spin-orbit coupling, which is hence not further considered in the following.

For the effective model, we restrict ourselves to the Fermi pockets (ii) for three reasons. First, the pockets in (ii) carry the dominant density of states at the Fermi level. Second, we preserve the complexity of multiple van Hove singularities of p-type and m-type in our minimal model. Third, upon comparison to the ab-initio band structure, our minimal model manages to correctly capture all irreducible band representations at the high symmetry points in the Brillouin zone. The constituting $d_{xz/yz}$ orbitals belong to the $B_{2g/3g}$ irreducible representations of the site symmetry group $D_{2h}$ for the 3f Wyckoff positions [Fig. 2(b)], forming a set of bands with opposite mirror eigenvalues along the $\Gamma-M$ line. These bands give rise to a mirror-symmetry-protected Dirac cone on the $\Gamma-M$ line and hence, an upper and lower van-Hove filling with opposite sublattice parity (Fig. 2). Employing the $D_{2h}$ point group symmetry, our corresponding effective six-band Hamiltonian can then be derived as

$$H = \sum_{k\alpha} \epsilon_{k\alpha} c^{\dagger}_{k\alpha} c_{k\alpha} - \sum_{kij} t_{ij} \Phi_{ij}(k) c^{\dagger}_{kj\alpha} c_{ki\alpha}$$

$$-t' \sum_{k,ij} \Phi_{ij}(k) s_{ij} (c^{\dagger}_{kjxz} c_{kiyz} - c^{\dagger}_{kjyz} c_{kixz}),$$

(1)
where \((i = A, B, C\) and \(\alpha = xz, yz\)). The crystal field splitting is denoted by \(\epsilon_\alpha\), where the operator \(c_i^{\dagger \alpha}\) (\(c_i^{\alpha}\)) creates (annihilates) an electron with momentum \(k\) of sublattice \(i\) in orbital \(\alpha\). The lattice structure factors read \(\Phi_{\text{AB}}(k) = 1 + e^{-2i(k-a_1)}\), \(\Phi_{\text{BC}}(k) = 1 + e^{-2i(k-a_2)}\), and \(\Phi_{\text{AC}}(k) = 1 + e^{-2i(k-a_3)}\) obeying the hermiticity condition \(\Phi_{ij}(k) = (1 - \delta_{ij})\Phi_{ji}^*(k)\), where the sublattice-connecting vectors are denoted by \(a_{1,2} = (\sqrt{3}/2, \pm 1/2)^T\) and \(a_3 = (0, -1)^T\). The second term represents the intra-orbital NN hoppings on the kagome lattice with two distinct amplitudes \(t_\alpha\), while the third term describes NN inter-orbital hopping amplitude \(t'\). The non-trivial transformation properties of the \(d_{xz}\) and \(d_{yz}\)-orbitals under the site-symmetry group result in a non-trivial sign structure for the third term, described by \(s_{\text{AC}} = s_{\text{CB}} = -s_{\text{AB}}\) and \(s_{ij} = -s_{ji}\). We approximately fit our model to the ab-initio cRPA estimates for a target manifold comprising V-3d and Sb-5p orbitals [35]. An extensive ab initio study of interactions and their dependence on the effective low-energy model will be presented elsewhere.

The inset of Fig. 3 displays the leading eigenvalue of the bare susceptibility \(\chi_\alpha(q)\) along high-symmetry lines. It is mainly attributed to the \(d_{yz}\) orbital and features three prominent peaks. The largest two are located proximate to the \(\Gamma\) point, while the peak close to \(M\) is suppressed through sublattice interference. Including onsite and NN interactions at the RPA level, these peaks get significantly enhanced in the spin as well as charge channel. Note that, indeed, we find the charge susceptibility at the verge of diverging around the \(M\) point for strong NN repulsion, hinting at an incident CDW instability.

Below the critical interaction, superconductivity emerges, triggered by charge and spin fluctuations [36–38]. The obtained pairing eigenvalues as a function of \(U\) are displayed in Fig. 3. For \(U < 0.54\) eV, pairing on the p-type Fermi sheet from the p-type van Hove band with \(B_{1u}\) \((f_{z^2-3y^2}-\text{wave})\) symmetry is favored and \(E_{1u}\) \((p-\text{wave})\) and \(E_{2g}\) \((d-\text{wave})\) pairings are subdominant. Increasing the coupling results in a rapid increase of the \(B_{2u}\) \((f_{y^2-3z^2}-\text{wave})\) and \(E_{2g}\) pairings on the Fermi sheet from the m-type van Hove band, where the spin-triplet solution still dominates slightly. Upon further increase of the interaction strength, the \(d\)-wave pairing on this Fermi sheet becomes dominant. Meanwhile, the
$E_{1u}$ pairing is subdominant. Varying the ratio of $V/U$ ($0.2 < V/U < 0.35$) does not qualitatively change the above results in the six-band tight-binding model (see Sec. IV of SM). Note that we have also performed calculations with a seven-band tight-binding model including the circular pocket around $\Gamma$ [39] and find that it has negligible effect on the pairing (see Sec. IV of SM).

We further analyze the harmonic fingerprint of the obtained pairings. The $f^{3}_{x^2-y^2}$-wave pairing is dominated by the sublattice-pure $d_{xz}$ Fermi surface and the corresponding gap function in $k$-space is shown in Fig. 4(a), where there are line nodes along $\Gamma-K$ and the superconducting gap changes sign under $60^\circ$ rotation. The corresponding real-space pairing is displayed in Fig. 4(b), which represents a spin-triplet sublattice-triplet pairing between $d_{xz}$ orbitals on the next-nearest-neighbor (NNN) sites. This pairing is promoted by the effective interaction between the NNN sites from the second-order effect of NN repulsion, an effect which is robust to in-plane interaction between the NN sublattices. Furthermore, note that the superconducting gap in the obtained states for our minimal model are either dominant on the $d_{xy}$ or $d_{xz}$ Fermi surface, which can be attributed to the assumed weak inter-orbital hopping.

**Topological properties of the pairing states.** Our minimal-model analysis is dominated by an $f$-wave state for weak coupling. Combined with the observation that the band renormalization in ARPES appears moderate, $f$-wave order could be a favored candidate for the nature of pairing in AV$_3$Sb$_5$. For time-reversal-invariant superconductors, the topological criterion about zero-energy Andreev bound states on edges is determined by winding numbers [42, 43]. For both $f$-wave pairing states emerging in our analysis, each node carries a winding number of $+1$ or $-1$. If we impose open boundary conditions, where the projections of nodes with opposite winding number do not overlap, a zero-energy flat band connecting the projections of nodes is created. For illustration, we present the surface spectrum of the $f^{3}_{x^2-y^2}$-wave state with open boundary conditions along the $x$ direction in Fig. 4(c). The corresponding local density of states features a sharp zero-bias peak, shown in Fig. 4(d) which could be observed at corresponding step edges in STM measurements. A similar analysis can likewise be performed for the $d$-wave and $p$-wave state. Chiral superconductors, which are likely to result from either $d$-wave or $p$-wave instabilities on hexagonal lattices, are potential hosts to Majorana zero modes in their vortex cores [44, 45].

**Experimental signatures.** The observation of a finite $\kappa/T$ for $T \to 0$ in thermal conductance measurements [29] as well as the typical $V$-shaped gap in STM measurements [31] have provided supporting experimental evidence for a nodal gap in AV$_3$Sb$_5$, which would be in line with $f$-wave pairing which we obtain in a large parameter regime for our minimal model. An $f$-wave state will have additional, clear experimental signatures. First, since the $f$-wave state pairs electrons in the spin-triplet channel, we expect the spin susceptibility in the superconducting phase to stay constant upon lowering the temperature, which should be seen in Knight-shift measurements. A further signature of spin-triplet pairing is often a high critical field. However, recent critical field measurements for both in-plane [46] and out-of-plane [29] fields indicate orbital limiting at rather low fields, such that the critical fields cannot distinguish the Cooper-pair spin structure. Finally, many thermodynamic quantities allow to identify the nodal structure through the tem-
FIG. 4. The inter-sublattice triplet $f_{x^3-3y^2}$-wave pairing function in (a) momentum and (b) real space, where the superconducting order parameter changes sign under $60^\circ$ rotation. (c) Edge spectra for open boundary conditions along $x$ direction; zero-energy Andreev bound states appear between the projections of nodal points in the $f$-wave pairing. (d) Density of states for the bulk $f$-wave pairing (black) and local density of states at edges from Andreev bound states (red).

Our work shows the unique principles for unconventional pairing in kagome metals, which promises to unlock a whole new paradigm of electronically-mediated superconductivity.

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