A silver(II) route to unconventional superconductivity

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The highly unusual divalent silver in silver difluoride (AgF$_2$) features a nearly square lattice of Ag$^{+2}$ bridged by fluorides. As a structural and electronic analogue of cuprates, its superconducting properties are yet to be examined. Our first principles electronic structure calculations reveal a striking resemblance between AgF$_2$ and the cuprates. Computed spin susceptibility shows a magnetic instability consistent with the experimentally observed antiferromagnetic transition. A linearized Eliashberg theory in fluctuation-exchange approximation shows an unconventional singlet d-wave superconducting pairing for bulk AgF$_2$ at an optimal electron doping. The pairing is found to strengthen with a decreasing interlayer coupling, highlighting the importance of quasi-2D nature of the crystal structure. These findings place AgF$_2$ in the category of unconventional high-$T_c$ superconductors, and its chemical uniqueness may help shed new lights on the high-$T_c$ phenomena.

Introduction. Superconducting properties of high $T_c$ cuprates emerge from an intricate interplay between electronic, lattice and spin degrees of freedom [1–2]. The cuprate crystal structure is generally derived from the perovskite type structure, featuring a few universal themes. Structurally they all contain quasi two-dimensional (2D) CuO$_2$ sheets. Their normal state electronic structure near the Fermi energy is dominated by a single band derived from Cu-$d$ orbitals [1–6]. In sharp contrast to conventional superconductors based on electron-phonon coupling assisted Cooper pair formation, superconductivity in cuprates is believed to be driven largely by strong electronic interactions [6]. The quasi-2D nature of crystal structure limits electronic modes in the out-of-plane direction, resulting in reduced screening and enhanced interaction that are essential to high-$T_c$ superconductivity. Understanding obtained from the extensive studies of structural, electronic and superconducting properties of cuprates has led to discoveries of new superconducting materials [6–8]. Insights into the interplay of geometric and electronic structure are key to discovery of novel superconductors. Clearly, it is then attractive to assay materials that resemble cuprates, both structurally and electronically, for potential novel superconductivity.

Materials hosting divalent silver are extremely scarce in comparison with monovalent silver compounds. Silver difluoride (AgF$_2$) has been synthesized from AgNO$_3$, anhydrous hydrogen fluoride treated with K$_2$NiF$_6$ and elemental fluoride, with silver ion Ag(II) in a highly unusual divalent state despite relatively large second ionization potential compares to the first one [9, 10]. More interestingly, AgF$_2$ resembles cuprates’s parent phase La$_2$CuO$_4$ in its geometric, electronic and magnetic structures. An AgF$_2$ sheet of the bulk crystal is structurally similar to a CuO$_2$ sheet, with similar pattern of out-of-plane displacement of anion atoms as shown in Fig. 1(a). The divalent Ag features a 3$d^9$ valence shell, iso-valent to cuprates. The antiferromagnetic ground state of charge neutral AgF$_2$ is a charge-transfer insulator, which again is a familiar scenario in cuprates.

It is then a natural and tempting question whether AgF$_2$ will turn superconducting once metallized upon doping, like the cuprates. It is therefore the purpose of this work to study whether interaction can drive a superconducting transition in AgF$_2$, and what the ensuing pairing symmetry will be. We start with an investigation of crystal and electronic structure of AgF$_2$ and its resemblance to the archetypal cuprate, the orthorhombic La$_2$CuO$_4$ [6]. A comparison of the crystal and electronic structures obtained from first principles calculations establishes a compelling structural and electronic resemblance between these compounds. A multiband Hubbard model is constructed from which the spin susceptibility of AgF$_2$ within the random-phase approximation reveals an antiferromagnetic instability in accordance with experiments. Employing the fluctuation-exchange approximation and solving the linearized Eliashberg equations, we obtain the superconducting pairing strength ($\lambda$) and

![FIG. 1. (a) AgF$_2$ crystal structure. Purple and gray balls are F and Ag, respectively. Blue dashed lines through one of the Ag(II) indicate out-of-plane Ag-F bonds in a AgF$_6$ octahedron. The black arrows labeled $x$, $y$, $z$ indicate the local coordinates used to describe d-orbitals on Ag. In (b), green dash-lined box highlights a AgF$_4$ unit. $+/−$ indicate out-of-plane displacements of fluoride ions. The $d_{x^2−y^2}$ Wannier orbital on the central Ag is shown.](image-url)
symmetry. A phase diagram is obtained by calculating \( \lambda \) at various carrier doping level and Hubbard \( U \) values. The strongest superconducting pairing is obtained at 5% electron doping for bulk AgF_2, with a dominating singlet \( d_{x^2-y^2} \) symmetry. We find that the superconducting pairing strength is gradually noted to increase with decreasing interlayer coupling. We attribute this effect to the renormalization of electron-electron correlations with decreasing out-of-plane coupling.

**Electronic structure.** The structure of AgF_2 can be viewed as a stack of Ag-F square-planar networks resembling the cuprate planes in La_2CuO_4 [6] as shown in Fig. 1(a). Similar to the low-temperature polymorph of La_2CuO_4 [6], AgF_2 has an orthorhombic crystal structure with each Ag(II) in a distorted octahedral crystal field of six nearest-neighbor F\(^-\) ions [11, 12]. However, unlike in a perfect octahedral coordination, the out-of-plane Ag-F bonds are elongated by 24\% relative to the in-plane ones as shown by blue dashed lines on one of the Ag(II) in Fig. 1(a), leaving Ag(II) 4-coordination in a AgF_2 unit. This again resembles La_2CuO_4 in which there is a 27\% elongation of the out-of-plane Cu-O bonds. These four F\(^-\)-coordinated Ag(II) form AgF_4 unit within the square-planar network, as indicated by green dash-lined box in Fig. 1(b). A significant deviation of AgF_2 structure from La_2CuO_4 comes from the tilting of this AgF_4 unit by a large angle \( \sim 25^\circ \), and hence the plane is puckered as shown in Fig. 1. This tilt of CuO_4 in La_2CuO_4 is much gentler (\( \sim 5^\circ \)). The TM-anion-TM (TM = Ag or Cu) angles in the square-planar structure are \( \sim 130^\circ \) for AgF_2, which is \( \sim 173^\circ \) in La_2CuO_4. This distortion from the ideal 180\(^\circ\) angle is expected to manifest itself in the superexchange interaction, and therefore the temperature of magnetic ordering. Indeed, the Ne\'{e}l temperature (\( T_N \)) is 300 K for La_2CuO_4 and 163 K for AgF_2 [13, 14]. Given the striking similarities of structural and magnetic properties of AgF_2 with that of La_2CuO_4 and the subtle difference, investigation of its electronic properties in context of superconductivity is warranted.

As discussed earlier, a single \( d_{x^2-y^2} \) orbital for AgF_2 shown in Fig. 1(b) dominating the low energy space near the Fermi level is one of the most prominent characteristic feature similar to the cuprates. This is schematically shown in Fig. 2(a) where partially filled \( d_{x^2-y^2} \) shown in red, contributes at the Fermi level. In the octahedral crystal field, the \( d \) orbitals are split into a triply degenerate \( t_{2g} \) set and a doubly degenerate \( e_g \) set. Deviation from perfect octahedral symmetry described earlier lifts the degeneracy of \( e_g \) orbitals with \( d_{z^2} \) being lower in energy than the in-plane \( d_{x^2-y^2} \). The occupied anion 2\( p \) orbitals are situated deep below the Fermi level. Non spin-polarized band structure of AgF_2, calculated using the density-functional theory, shown in Fig. 2(b) exhibits features akin to cuprates. For calculational details, refer to the Supplemental Material(SM) [15]. The low-energy excitations are dominated by the half-filled \( d_{x^2-y^2} \) on Ag, and are well separated from all other bands. Thus when constructing a tight-binding model, it is justified to include only one \( d_{x^2-y^2} \)-like Wannier orbital per Ag.

Once the Coulomb interaction is included AgF_2 becomes charge-transfer antiferromagnetic insulator similar to the cuprates in a scenario discussed also in Jakub et al [9].

**Interaction-mediated superconductivity.** To investigate the effect of interaction on the magnetic order and potential superconductivity, we construct a multiband Hubbard model,

\[
H = H_0 + H_U = \sum_{ij\sigma\sigma'} \epsilon_{ij\sigma} c_{ij\sigma}^\dagger c_{ij\sigma'} + U \sum_i n_{i\uparrow} n_{i\downarrow},
\]

where \( i, l \) and \( \sigma \) are lattice, orbital and spin indices, respectively, and \( c \) and \( n \) are Fermion annihilation and number operators, respectively. The low-energy bands are described by tight-binding Hamiltonian \( H_0 \), in which hopping amplitudes are derived from the maximally-localized Wannier function approach [16] (see Table S1 in SM [15]). The resultant band structure is shown in Fig. 2(b), where the four \( d_{x^2-y^2} \) bands (four Ag per unit cell) from the tight-binding model fit the first principles bands well. The intra-orbital Hubbard parameter \( U \) is determined by estimating the Ne\'{e}l temperature in a random-phase approximation (RPA), as described next.

Within the RPA [17–21], the charge (\( \chi^c \)) and spin (\( \chi^s \)) susceptibilities are given by,

\[
\begin{align*}
\chi^c(q) &= \left[ 1 + \chi^0(q) U_{c} \right]^{-1} \chi^0(q), \\
\chi^s(q) &= \left[ 1 - \chi^0(q) U_{s} \right]^{-1} \chi^0(q),
\end{align*}
\]

FIG. 2. (a) Schematic energy level diagram of Ag(II) in the crystal field of F atoms. (b) Non-spin polarized band structures of AgF_2 from first principles calculation (DFT) and Wannier interpolation based tight binding model(TB), near the Fermi energy. (c) \( \eta(q) \) for undoped bulk AgF_2 with isovalue 0.5(cyan), 0.7(yellow) and 0.9(red). Here, \( U = 0.44 \) eV and \( T = 14 \) meV. (d) Magnetic structure of AgF_2 obtained from RPA. Red arrows represent the magnetic moment on Ag atoms.
where $q = (q, \omega)$, $\chi^0$ is the bare susceptibility, and $U^c$ and $U^s$ are the interaction matrices in the charge and spin channels, respectively [15]. The onset of spin instability is detected by the condition $|1 - \chi_0(q, 0)U^c| = 0$, which happens when the maximum eigenvalue of $\chi_0(q, 0)U^c$ (denoted by $\eta(q)$) becomes unity at any $q$. The ensuing divergence of $\chi^0$ leads to a magnetic phase transition. The vector $q^*$ and temperature $T_N$ at which $\eta(q^*) = 1$ are the Néel temperature and propagation vector of the spin pattern, respectively. The spin pattern corresponding to a $q^*$ is determined by diagonal elements of the eigenvector $\xi(q^*)$ corresponding to $\eta(q^*)$. We use a mesh of $48 \times 48 \times 48$ for Brillouin zone sampling in all our calculations on the Hubbard model.

The Hubbard $U$ is estimated to be $0.44 \text{ eV}$, by matching the experimentally observed Néel temperature ($\sim 163 \text{ K}$) of AgF$_2$ (Fig. S1 in SM [15]). Isosurfaces of $\eta(q)$ drawn in Fig. 2(a) for undoped bulk AgF$_2$ at $T = 14 \text{ meV} \ (\sim 163 \text{ K})$ show a strong anisotropy corresponding to a strong intra-layer and a weak inter-layer magnetic exchange interactions. Henceforth, we focus on $q_z = 0$ plane in current analysis. The maximum value of $\eta(q)$ is found to lie along the $q_z$-axis. Thus, for a weak inter-layer coupling when restricted only in the $q_x$-$q_y$ plane, the maximum value of the spin susceptibility is attained at $q = 0$. The computed eigenvectors $\xi_i(q = 0)$ yield an antiferromagnetic order shown in Fig. 2(d), consistent with the experimentally established Néel state in AgF$_2$ [14].

When doped with a carrier concentration that readily suppresses the magnetism, a cuprate goes metallic exhibiting various kinds of instability such as charge and spin fluctuations at low temperatures due to Fermi surface reconstruction. The Hubbard models have been used extensively to explain the superconductivity in doped cuprates [7]. Keeping the striking resemblance of AgF$_2$ with cuprates, similar approach of metallization by carrier doping applies in AgF$_2$ as well. Thus, having a model capable of describing the magnetic instability and order of AgF$_2$, we go on to a scrutiny for potential superconductivity mediated by spin fluctuation. The fluctuation-exchange approximation (FLEX) [22, 23] is employed to describe the effective electron-electron interaction $\Gamma(q)$ given by,

$$\Gamma(q) = \gamma U^c \chi^0(q) U^s - \frac{1}{2} U^c \chi^0(q) U^c + \frac{1}{2} (U^s + U^c) \quad (3)$$

with $\gamma = \frac{3}{4}$ for the singlet channel and $\gamma = -\frac{1}{2}$ for the triplet channel. An effective pairing between the electrons on the Fermi surface arising from spin and/or charge fluctuations can result in the formation of Cooper pairs. To describe the pairing instability of this type, the linearized Eliashberg equation is solved in the weak-coupling regime,

$$\lambda \phi_{mn}(k) = -\frac{1}{N} \sum_{k'} \sum_{\mu \nu} \Gamma_{\mu \nu}^{mn}(k, k') F_{\mu \nu}(k') \phi_{\mu \nu}(k') \quad (4)$$

where $F_{\mu \nu}(k')$ is a factor arising from summing the product of Green’s functions over Matsubara frequencies and $m, n, \mu, \nu$ are band indices. $\phi_{mn}(k)$ is the order parameter of superconducting phase [15]. Eq.(4) then is an eigenvalue equation. The largest eigenvalue $\lambda_{\text{max}}$ becomes unity at superconducting $T_C$ and can be used to gauge the relative pairing strength near the $T_C$.

Eq.(4) is solved for various doping levels and $U$ values at $T = 30 \text{ meV}$. Fig. 3(a) shows the contour plot for $\lambda_{\text{max}}$ in the $U - \delta n$ parameter space. As observed in Fig. 3(a) the superconducting pairing strength increases with increasing $U$ at a given doping, underlining the importance of electronic correlation for potential superconductivity in this compound. The symmetry of pairing can be identified by assigning each solved $\phi_{mn}(k)$ to an irreducible representation of the $D_{2h}$ point group, symmetry group of the bulk AgF$_2$. Corresponding to each irreducible representation $i$ (Table S2 in SM [15]), the largest eigenvalue is denoted by $\lambda_{i\text{max}}$. Fig. 3(b) shows the doping dependence of $\lambda_{i\text{max}}$ for various pairing symmetry at $U = 0.44 \text{ eV}$. One can find that the singlet $d$-wave pairings have significantly higher strength than triplet $p$-wave pairings and the leading pairing symmetry is singlet $d_{xz}$-type wave throughout the $U - \delta n$ parameter space shown in Fig. 3(a). Moreover, the hole doping readily decreases $\lambda$, while the electron doping tends to increase $\lambda$ at first, reaching a peak value at an optimal doping of 5% beyond which further doping tends to reduce $\lambda$.

To circumvent the gauge problem [15] of $\phi_{mn}(k)$ for degenerate bands, we define $\phi(k)$ as

$$\phi(k) = \sum_n \phi_{mn}(k) \delta(\varepsilon_{nk} - \varepsilon_F) \quad (5)$$

which describes the nature of order parameter on Fermi surface. Here $\varepsilon_{nk}$ and $\varepsilon_F$ are band energy and Fermi energy respectively. Energy cutoff of 5 meV is considered for evaluation of $\delta$ function in our calculations. We plot the real part of $\phi(k)$ corresponding to $\lambda_{\text{max}}$ for one of the AgF$_2$ layers with $U = 0.44 \text{ eV}$ and optimal doping of 5%. This is shown in Fig. 3(c) in the 3-dimensional Brillouin zone, and Fig. 3(d) shows a projection onto the $k_x$-$k_z$ plane. One encounters nodes crossing $k_z = \pi$ or $k_z = \pi$ planes indicating a $d_{xz}$-wave pairing. Hence, it can be concluded that the bulk AgF$_2$ crystal becomes unstable to a $d$-wave pairing induced by spin fluctuation.

**Interlayer coupling.** As discussed in the beginning, quasi-2D nature of the crystal structure of cuprates is one of the factors favoring its high $T_C$ [24, 25]. In the case of AgF$_2$, although Ag-F layers resemble the copper oxide sheets, the separation of these planes is 2.91 Å, much smaller than what is observed for La$_2$CuO$_4$ (6.6 Å) as well as other cuprates. Consequently, the effect of interlayer coupling on superconducting properties of AgF$_2$ clearly warrants further study. In other words, AgF$_2$ provides a good platform to investigate the role of
FIG. 3. (a) Contour plot of $\lambda_{\text{max}}$ of the linearized Eliashberg equation at $T = 30$ meV. Blue dash line correspond to $\eta(q)$ reach to unity at any $q$. (b) Doping dependence of $\lambda_{\text{max}}^{i}$ for several pairing symmetry of bulk AgF$_2$ at $U = 0.44$ eV and $T = 30$ meV. Square representing the singlet and triangle representing the triplet channels. (c) Three-dimensional and (d) projection on $k_x-k_z$ plane, of the order parameter $\phi(k)$ at Fermi surface of one of the AgF$_2$ layer with $\delta n/Ag=0.05$ for bulk AgF$_2$.

The quasi-2D nature of crystal structure in superconducting properties. Additionally, monolayer or few-layer samples more prone to doping by techniques such as field or electrolytic gating [26], which is a clear experimental advantage.

To study the effect of interlayer coupling on superconducting properties of AgF$_2$, we interpolate between the bulk and monolayer limits as follow,

$$H_0 = H^{\text{intra}} + \alpha H^{\text{inter}}$$

(6)

Here, $H^{\text{intra}}$ is a tight binding model within a single layer of AgF$_2$, while $H^{\text{inter}}$ is the interlayer hopping term, which is scaled by $\alpha \in [0, 1]$. Bulk AgF$_2$ can be obtained with $\alpha = 1$, and $\alpha = 0$ correspond to a single layer of AgF$_2$ [27].

In Fig. 4(a), $\lambda_{\text{max}}^{d_{xy}}$ and $\lambda_{\text{max}}^{d_{xz}}$ as functions of doping concentrations for different interlayer coupling strength $\alpha$ are shown. It can be seen that the interlayer coupling tends to suppress the superconducting pairing in both hole and electron doping because a stronger interlayer coupling amounts to a weaker electronic correlations as discussed earlier. Evidently the quasi-2D nature of the crystal structure is one of the crucial factors in the favor of high $T_C$ superconductivity, which again confirms the resemblance to cuprates. The optimal electron doping concentration remains unchanged as of bulk AgF$_2$, which indicates that the Fermi surface nesting responsible for divergence of spin susceptibility mainly occur within a single layer without any significant interlayer contribution. In the absence of interlayer coupling, the seperated monolayers have the same leading pairing symmetry of the $d_{xy}$-type. These two degenerate $d_{xy}$ wave then split into $d_{xz}$ and $d_{xy}$ immediately after the interlayer coupling was switched on. The difference of pairing strength between leading $d_{xz}$ wave and competing $d_{xy}$ was also found to increase with increase of $\alpha$.

For single layer AgF$_2$, the symmetry reduces to $C_{2h}$ point group from $D_{2h}$ of bulk AgF$_2$. Performing similar analysis to that for bulk, we show the doping dependence of $\lambda_{\text{max}}$ for several pairing symmetry for single layer in Fig. 4(b). As seen previously in Fig. 4(a), the leading pairing symmetry is $d_{xy}$ followed by $d_{xz}$ and $d_{xy}$ wave. The real part of the $\phi(k)$ corresponding to the $\lambda_{\text{max}}$ at the optimal doping concentration is shown in Fig. 4(c), which clearly reveals $d_{xy}$ pairing symmetry in the monolayer limit.

**Summary and outlook.** Our calculations indicate AgF$_2$ is not only chemically exotic, but also harbors unconventional superconductivity in a way very similar to high-$T_C$ cuprates. Our multiband Hubbard model reveals a magnetic instability in accordance with the experimentally obtained magnetic ground state. In the fluctuation-exchange approximation, we find a superconducting ground state with a singlet $d$-wave pairing for the bulk AgF$_2$ at an optimal electron doping of 5%. By varying the strength of interlayer interaction, we show that the superconducting pairing strength increases with decreasing interlayer coupling, highlighting the crucial role played by quasi-2D crystal structure on superconducting properties of such materials.
Drawing hints from cuprates, metallization of bulk AgF$_2$ can be achieved by synthesizing it with a modified composition as is done in case of La$_{2-x}$Ba$_x$CuO$_4$/La$_{2-x}$Sr$_x$CuO$_4$ [1, 28], leading to doping of extra charge carriers in transition metal-anion plane. Another route to metallization is electric gating [29]. A monolayer of AgF$_2$ may be realized by epitaxial growth [30], metallization of which can be achieved during the deposition process. The idea of liquid-gating induced superconductivity in thin films [31] can also be applied to monolayer AgF$_2$.

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Supplemental Material:
A silver(II) route to unconventional superconductivity

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A. Density-functional theory calculations

To calculate electronic structure of AgF\textsubscript{2}, we have performed density-functional theory based calculations using Vienna \textit{ab initio} simulation package (VASP) [1]. The projector-augmented wave potentials are used in our calculations [2, 3], with a 6×6×8 Gamma-centered mesh of k-points and a plane wave cutoff of 800 eV. The results are found to be well converged with respect to k-point mesh and plane wave numbers. The Perdew-Burke-Ernzerhof functional [4] is used for the exchange-correlation functional, and additional electron-electron correlation is included statically by a $U = 8$ eV within the GGA+U formalism [5]. The total energy is calculated self-consistently till the energy difference between successive steps is below $10^{-5}$ eV. The experimental lattice parameters of orthorhombic AgF\textsubscript{2} with space group \textit{Pbca} (No. 61) are $a = 5.101$ Å, $b = 5.568$ Å and $c = 5.831$ Å [6].

Considering the experimentally observed magnetic ground state and $U = 8$ eV on Ag-\textit{d} states, we perform full optimization of lattice parameters. The value of $U$ on Ag-\textit{d} orbital is chosen to match the experimentally observed Ag magnetic moments of 0.7 $\mu_B$/Ag. Change in a and b lattice constants were found to be less than 1\% while the c vector was enhanced by $\sim 1.7\%$. Since these values are very close to the experimental ones, we use the experimental structure for further study. Non-spin polarized \textit{ab initio} band structure was projected onto a tight-binding model using the maximally-localized Wannier functions for the radial part of the wavefunctions using WANNIER90-VASP interface, implemented within VASP [7]. Mapping is done for the undoped case considering Ag-\textit{d}_{x^2-y^2} orbitals and the hopping amplitudes are listed in Table S1.

B. Random-phase approximation and fluctuation exchange approximation

We start by setting up the multiband Hubbard model, by adding the Hubbard $U$ term to a tight-binding model described in section A,

$$H = H^{\text{TB}} + H^{\text{hub}} = \sum_{ijl'l'} t_{ijl'}^{ll'} c_{il\sigma}^\dagger c_{jl'l'} \sigma + U \sum_{il} n_{il\uparrow} n_{il\downarrow} \quad (1)$$

Here, $c_{il\sigma}$ is annihilation operator of $d_{x^2-y^2}$ orbital on Ag site $l$ in $i$\textsuperscript{th} unit cell with spin $\sigma$ and $t_{ijl'}^{ll'}$ is hopping amplitude. $U$ is the on-site Hubbard parameter.

The random-phase approximation (RPA) [8–12] is employed to study the magnetic insta-
TABLE S1. List of all hopping amplitudes larger than 5 meV in our Wannier function based tight-binding model of bulk AgF$_2$. $R$ and $l/l'$ are the translation vector and orbital index respectively. All other hoppings not listed here can be obtained by applying symmetry operations of $Pbca$, the space group of bulk AgF$_2$.

| $R$         | $l$ | $l'$ | $t$ (meV) |
|-------------|-----|------|-----------|
| (0 , 0 , 0) | 1   | 2    | -173.1    |
| (1 , -1 , 0)| 1   | 1    | 34.4      |
| (0 , 1 , 0) | 1   | 1    | 34.0      |
| (0 , 1 , -1)| 1   | 1    | 28.0      |
| (0 , -1 , 0)| 1   | 3    | 22.8      |
| (0 , 0 , -1)| 1   | 2    | 18.2      |
| (1 , 0 , -1)| 1   | 1    | 18.1      |
| (1 , 1 , 0) | 1   | 1    | 15.1      |
| (0 , 1 , 0) | 1   | 3    | -11.4     |
| (0 , 0 , 0) | 1   | 4    | 7.8       |
| (1 , 0 , 1) | 1   | 1    | 7.1       |
| (0 , 0 , 0) | 1   | 3    | -7.0      |
| (1 , 0 , 0) | 1   | 4    | -6.1      |
| (1 , -1 , -1)| 1   | 1    | -5.6      |
| (1 , 0 , 0) | 1   | 3    | 5.3       |
| (1 , 0 , 0) | 1   | 1    | 5.1       |

bility of the system. The RPA spin and charge susceptibilities are given by

$$\chi^{c}(q) = [1 + \chi^{0}(q)U^{c}]^{-1}\chi^{0}(q)$$

$$\chi^{s}(q) = [1 - \chi^{0}(q)U^{s}]^{-1}\chi^{0}(q)$$

respectively, where

$$(U^{c/s})_{l_{1}l_{2}}^{l_{3}l_{4}} = \begin{cases} U & l_{1} = l_{2} = l_{3} = l_{4} \\ 0 & \text{otherwise} \end{cases}$$

(3)

Here $q = (q,\omega)$. 

3
In above expression the bare susceptibility $\chi^0(q)$ is given by,

$$
(\chi^0)_{l_1l_2}^{l_3l_4}(q,\omega) = -\frac{1}{N} \sum_{k,\mu\nu} a^{l_2}_\nu(k+q) a^{l_3*}_\nu(k+q) a^{l_4}_\mu(k) a^{l_1*}_\mu(k) \frac{f(\varepsilon_{\mu k} - \varepsilon_F) - f(\varepsilon_{\nu k+q} - \varepsilon_F)}{\omega + \varepsilon_{\mu k} - \varepsilon_{\nu k+q} + i0^+},
$$

(4)

where $N$ is the number of $k$-points and $\mu, \nu$ are the band indices. $a^l_\mu(k)$ is the $l$ component of wave function of band $\mu$, obtained from diagonalization of $H^{TB}$ and $\varepsilon_\mu(k)$ is the corresponding eigenvalues of band $\mu$ at $k$-point. $f(E)$ is the Fermi-Dirac distribution and $\varepsilon_F$ is Fermi energy.

Under fluctuation exchange approximation(FLEX) [13, 14], the pairing vertex describing the effective electron-electron interaction is given by,

$$
\Gamma(q) = \gamma U^s \chi^s(q) U^s - \frac{1}{2} U^c \chi^c(q) U^c + \frac{1}{2} (U^s + U^c),
$$

(5)

with $\gamma = \frac{3}{2}$ for the singlet channel and $\gamma = -\frac{1}{2}$ for the triplet channel. The singlet vertex is symmetrized as,

$$
(\Gamma^s)_{l_1l_2}^{l_3l_4}(k,k') = \frac{1}{2} \left( (\Gamma^s)_{l_1l_2}^{l_3l_4}(k,k') + (\Gamma^s)_{l_3l_2}^{l_1l_4}(k,-k') \right),
$$

(6)

and the triplet vertex is anti-symmetrized as,

$$
(\Gamma^t)_{l_1l_2}^{l_3l_4}(k,k') = \frac{1}{2} \left( (\Gamma^t)_{l_1l_2}^{l_3l_4}(k,k') - (\Gamma^t)_{l_3l_2}^{l_1l_4}(k,-k') \right),
$$

(7)

Here, $\Gamma(k,k') = \Gamma(q)$ with $q = k - k'$.

The linearized Eliashberg equation will then be solved to obtain the order parameter and superconducting transition temperature,

$$
\lambda \phi_{l_1l_2}(k) = -\frac{T}{N} \sum_q \sum_{l_3l_4l_5l_6} \Gamma_{l_1l_2}^{l_3l_4}(q) \phi_{l_5l_6}(k-q) G_{l_2l_6}(k-q) G_{l_4l_6}(q-k).
$$

(8)

Here, $\lambda$ is the eigenvalue indicating the pairing strength. The eigenvector $\phi_{l_1l_2}(k)$ is the order parameter written in term of orbital index $l_1$ and $l_2$. $G_{l_1l_2}(k)$ is the Matsubara Green function.

In the weak-coupling regime, the pairing vertex is approximated to be frequency-independent i.e. $\Gamma(q) = \Gamma(q, \omega = 0)$. After summing over the Matsubara frequencies, we obtain the following equation in band basis,

$$
\lambda \phi_{mn}(k) = -\frac{1}{N} \sum_{k'} \sum_{\mu\nu} (\Gamma^t)^{mn}_{\mu\nu}(k,k') F_{\mu\nu}(k') \phi_{\mu\nu}(k'),
$$

(9)
where
\[ F_{\mu\nu}(k) = -\frac{f(\varepsilon_{\mu k} - \varepsilon_F) + f(\varepsilon_{\nu k} - \varepsilon_F) - 1}{\varepsilon_{\mu k} + \varepsilon_{\nu k} - 2\varepsilon_F}. \] (10)

The transformation between orbital basis and band basis for the order parameter is
\[ \phi_{mn}(k) = \sum_{l_1 l_2} \phi_{l_1 l_2}(k) a_{l_1}^{l_1*}(k) a_{l_2}^{l_2*}(-k), \] (11)
and for the vertex,
\[ (\Gamma^\eta)^{mn}_{\mu\nu}(k, k') = \sum_{l_1 l_2 l_3 l_4} a_{m}^{l_1*}(k) a_{n}^{l_2*}(-k) (\Gamma^\eta)^{l_1 l_2}_{l_3 l_4} (k - k') a_{\mu}^{l_3}(k') a_{\nu}^{l_4}(-k'). \] (12)

C. Pairing symmetry

One can see that \( \phi_{mn}(k) \) is not gauge invariant because of the eigenvector \( a_{l\mu}^{l}(k) \) involved in Eq.(11). However, if the following gauge is chosen,
\[ a_{l\mu}^{l}(-k) = a_{n\nu}^{l}(k)^* \] (13)
for the non-magnetic state [15], then the trace of \( \phi_{mn}(k) \)
\[ \phi(k) = \sum_{n} \phi_{mn}(k) \] (14)
or trace taken on Fermi surface
\[ \phi(k) = \sum_{n} \phi_{mn}(k) \delta(\varepsilon_{nk} - \varepsilon_F) \] (15)
is gauge invariant.

For the symmetry group of normal state Hamiltonian \( H \), which are \( D_{2h} \) for bulk AgF\(_2\) and \( C_{2h} \) for single layer AgF\(_2\), \( \phi(k) \) can be identified as one of the irreducible representations of the symmetry group. All the irreducible representation and basis functions of \( D_{2h} \) are listed in Table S2 and \( C_{2h} \) in Table S3. Note that the \( C_2 \) rotation in \( C_{2h} \) is along \( x \)-axis.
TABLE S2. Character table for $D_{2h}$ point group

|    | $E$ | $C_2(z)$ | $C_2(y)$ | $C_2(x)$ | $i$ | $\sigma(xy)$ | $\sigma(xz)$ | $\sigma(yz)$ | Linear, Rotations | Quadratic |
|----|-----|----------|----------|----------|----|--------------|--------------|--------------|------------------|-----------|
| $A_g$ | +1  | +1       | +1       | +1       | +1 | +1           | +1           | +1           |                  | $x^2 - y^2, z^2$ |
| $B_{1g}$ | +1  | +1       | +1       | +1       | -1 | -1           | -1           | -1           | $R_z$            | $xy$      |
| $B_{2g}$ | +1  | -1       | -1       | -1       | +1 | -1           | +1           | -1           | $R_y$            | $xz$      |
| $B_{3g}$ | +1  | -1       | -1       | +1       | +1 | -1           | -1           | -1           | $R_x$            | $yz$      |
| $A_u$ | +1  | +1       | +1       | -1       | -1 | -1           | -1           | -1           |                  |           |
| $B_{1u}$ | +1  | +1       | -1       | -1       | -1 | -1           | +1           | +1           | $z$              |           |
| $B_{2u}$ | +1  | -1       | -1       | -1       | +1 | -1           | +1           | +1           | $y$              |           |
| $B_{3u}$ | +1  | -1       | -1       | +1       | -1 | +1           | +1           | -1           | $x$              |           |

TABLE S3. Character table for $C_{2h}$ point group

|    | $E$ | $C_2(x)$ | $i$ | $\sigma_h$ | Linear, Rotations | Quadratic |
|----|-----|----------|----|------------|------------------|-----------|
| $A_g$ | +1  | +1       | +1 | +1         | $R_x$            | $x^2 - y^2, z^2, yz$ |
| $B_{1g}$ | +1  | -1       | +1 | -1         | $R_y, R_z$       | $xy, xz$   |
| $A_u$ | +1  | +1       | -1 | -1         | $x$              |           |
| $B_{1u}$ | +1  | -1       | -1 | +1         | $y, z$           |           |

FIG. S1. The $U$ dependence of largest $\eta$ at experimental Néel temperature ($\sim 163$ K = 14 meV). $\eta \approx 1$ when $U = 0.44$ eV.
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