

## Anonymous Report 2 on 2019-7-13 Invited Report

### Strengths

- 1- An interesting proposal is presented for superconductivity in heavy fermions
- 2- This is potentially testable in experiments

### Weaknesses

- 1- Poor writing
- 2- Assumptions need not be discussed/justified a little more strongly
- 3- Possible experimental tests must be fleshed out

### Report

The manuscript discusses a holon-based mechanism for superconductivity in heavy fermion systems. Starting from a Kondo lattice model, the authors introduce a holon-singlon-doublon representations  $n$  to describe the Hilbert space of each impurity. A strong on-site repulsion on the impurity site prohibits doublons. Subsequently, a chemical potential is introduced for the holons in order to preserve the impurity occupancy. The authors argue that these holons can mediate superconductivity by generating an effective attraction between pairs. Notably, each pair is composed of one conduction and one impurity electron.

**We thank the referee for taking the time to carefully read our manuscript and for providing us with encouraging and helpful comments and suggestions. We are glad that the referee finds our proposal interesting and testable in experiments. We carefully address referees all comments and suggestions below in point-by-point details and make appropriate changes in the manuscript.**

The presentation and the writing needs improvement. Spelling errors include “guage” and “Ovbnikov”. There are incorrect usages that make the manuscript difficult to read (e.g., “former materials” after listing three families of materials, using “condensate” as a verb, “primitive CeCu<sub>2</sub>Si<sub>2</sub> compound”, “...incipiently relies on...”, “..intercepting the first dome”, etc.). The authors should revise the manuscript in this regard.

**We apologize for our oversights and typographical errors. We appreciate that the referee has kindly pointed out these errors. We have now proof-read the entire manuscript more carefully to correct the above typos as well as any other typos that we spotted.**

The central idea presented here is interesting and plausibly applicable to CeCu<sub>2</sub>Si<sub>2</sub>. However, there are several points that are not clear to me:

- a. The authors review experiments on CeCu<sub>2</sub>Si<sub>2</sub>. In particular, they point out an apparent contradiction between early indications of nodal unconventional pairing and recent claims of gapped conventional pairing. In their subsequent analysis, they invoke many simplifying assumptions to argue for s-wave gapped pairing. Their stand is not clear here. Do they suggest that CeCu<sub>2</sub>Si<sub>2</sub> is a gapped superconductor? How do they explain earlier observations?

**Referee’s above point is valid, and we explain here this point in details and also made appropriate revision as mentioned below. There were mainly three experimental results**

which could be taken as evidence for ‘unconventional’ pairing. However, they can also be explained within the ‘conventional’ pairing scenario. (Here by ‘unconventional’ we refer to sign-reversal, k-dependent pairing structure with/without a nodal state, while ‘conventional’ means constant-sign, fully gapped pairing symmetry). We discuss these three experiments and their possible explanation within our theory.

- 1)  $T^3$  – dependence of the  $1/T_1$  data of NQR experiment [Refs. 13-15 in our manuscript]. The power law behavior of NQM data has been taken as evidence of nodal SC structure, and hence unconventional pairing. Later papers [Refs. 21, 22] have shown that the  $T^3$  behavior of  $1/T_1$  data can as well be reproduced within a two-band superconductor model with conventional, s-wave pairing symmetry. The same model is also used to describe specific heat and penetration depth data. Note that we also have a two band (conduction and local electrons) model with s-wave pairing and thus our theory can reproduce this data as well.
- 2) Angle dependent  $H_{c2}$  [Ref 16]: There are early reports finding a weak but finite a four-fold angle dependence of the upper critical field ( $H_{c2}$ ). This result suggests that the vortex size is anisotropic. The anisotropy in vortex can come from the SC gap anisotropy (such as d-wave gap), and/or from the Fermi surface anisotropy as well. In fact, one of us showed earlier in a different paper with realistic two-band model [Ref. 17] that a four-fold modulation in  $H_{c2}$  (and in other quantities) can solely come from the four-fold modulation of the Fermi surface even for isotropic s-wave superconductor. In addition, the disorder dependence of  $H_{c2}$  is very weak compared to what one would expect from an unconventional pairing [see Ref. 23, 25]. Therefore, the anisotropy in  $H_{c2}$  cannot be taken as a direct and ambiguous proof of unconventional pairing and can be explained within our theory by including Fermi surface anisotropy. In fact, prior DFT calculations have shown the presence of anisotropic Fermi surface in  $\text{CeCu}_2\text{Si}_2$  [Refs. 29,30].
- 3) Spin Resonance peak: A recent inelastic neutron scattering (INS) experiment observed a peak or hump like feature in the magnetic susceptibility below the superconducting transition temperature ( $T_c$ ). Both unconventional and conventional superconductor theories predict spin resonance in the SC state. For unconventional superconductivity, a resonance is expected at energy  $\omega < 2\Delta$  with very sharp spectral weight. On the other hand, a hump feature in spin susceptibility can be expected from conventional pairing at energy  $\omega > 2\Delta$ . Therefore, whether the observed mode indicates an unconventional or conventional pairing is not yet fully resolved. Our argument is as follows. (a) Given the absence of any direct measure of the SC gap amplitude  $\Delta$  in  $\text{CeCu}_2\text{Si}_2$ , it is yet not clear if the mode energy is below or above  $2\Delta$ . (b) The observed peak is very broad compared to a resonance peak expected for unconventional pairing, (c) Our theory also predicts a novel resonance like peak/hump in the energy scale of  $\frac{2\Delta^2}{\xi_f}$  which can well explain the experimental observation (see Sec. IV B).

Following referee’s comments above, we now expand our discussion section to address these three points in details [page 6, paragraphs 3, 4, 5 in the discussion section].

**Regarding referee's second point above "In their subsequent analysis, they invoke many simplifying assumptions to argue for s-wave gapped pairing. Their stand is not clear here. Do they suggest that CeCu<sub>2</sub>Si<sub>2</sub> is a gapped superconductor?": There are plenty of evidence for fully gapped, fixed sign superconductivity in CeCu<sub>2</sub>Si<sub>2</sub>. It is known from BCS theory that for an attractive potential, s-wave superconductivity is the most favorable channel if the attractive interaction is isotropic. In fact, it can be shown that for attractive pairing interaction, a sign-reversal pairing channel has much lower strength than a fixed sign one, like s-wave. Therefore, s-wave, spin singlet channel is the most natural pairing symmetry for an attractive potential, as we obtained here. Therefore, we aim to explain the increasing evidence for fully-gapped, constant-sign, s-wave pairing symmetry in this material.**

b. The authors refer to the e-particle as a gauge field. In my view, this language is not appropriate. It does not represent a gauge degree of freedom (like photons), even though a gauge structure can possibly be invoked for the representation. It may be better to call it a pseudo-particle in analogy with Schwinger bosons or Abrikosov fermions.

**Referee's above point is valuable. In the quantum field theory, when a gauge field (like electromagnetic potential) is quantized, we write the field in terms of ladder operators (e.g. photons). Since these ladder operators follow commutation relation, they are bosons. Similarly, phonons are the quantized bosonic particles of vibration, i.e., the restoring force – a gauge field. In the present case, we have holons (e-particles) which are bosons, and one can write an associated gauge field in the same fashion as**

$$A(\mathbf{r}) = \sum_q [e_q \exp(i\mathbf{q} \cdot \mathbf{r}) + e_q^\dagger \exp(-i\mathbf{q} \cdot \mathbf{r})]$$

where A is the corresponding gauge potential. In this sense, holons are not a gauge field, but are associated with a gauge field.

We realize it is better off using an easier nomenclature and hence we avoid calling holons as gauge field, and call it a bosonic field or simply holons throughout the entire paper.

c. There are several hidden and perhaps unnecessary assumptions in Eq. 1.  
i. Why do all impurity orbitals (all m's) have the same energy?

**This is an important point. First of all, as we mentioned in the manuscript, we have a "periodic" Anderson model. This means the impurity potential is same in every unit cell, enforced by the discrete translational invariance of the system. This fixes the impurity potential  $\xi_f$  (f-orbitals onsite energy) to be the same in all unit cells.**

The reason for not having any 'm' (pseudospin index) dependence in the onsite energy is the same. We are working in a single-impurity model, which means we only have a single orbital state with two possible spins. We note that even for a single impurity case, the holon onsite energy  $\omega_e$  can have different values. Here also, we are under the assumption that the holons are already condensed to a single state, and we are studying the fluctuations around the condensed energy. This is analogous to the Einstein phonon model. We mention this point in the manuscript [page 2, right column, third paragraph.]

ii. Why is the hybridization,  $v_k$ , independent of conduction electron spin? This seems particularly unreasonable. This assumption goes on to give s-wave character in pairing. This needs strong justification.

$v_k$  is the valence fluctuation potential, which mediates ‘charge’ or valency transfer between  $f$ - and conduction orbitals. The potential, by nature, does not violate the spin-conservation symmetry of each orbital, and thus cannot be spin-dependent. This is the model proposed and used widely in the literature to describe the mixed valence phenomena of heavy-fermions materials.

Although, the spin-dependence of the valence fluctuation potential (hybridization potential)  $v_k$  is itself unphysical, but it is not related to the s-wave pairing channel. The  $k$ -independence of  $v_k$  (isotropic) is rather related to the isotropic nature of the potential.

[A secondary note can be made in the context that: The valence fluctuation in addition to Hubbard interaction gives the Kondo model in which spin-spin interaction (Kondo coupling) arise from the Hubbard interaction, not from the valence fluctuation.]

iii. Later on, the authors assume on-site hybridization to motivate s-wave pairing. They should justify this or atleast provide references to previous studies where this has been argued.

This was also briefly mentioned in the manuscript [page 3, right column, first paragraph]. A  $k$ -dependence in the valence fluctuation potential  $v_k$  would arise if the  $f$ -orbital and the conduction electron orbitals are not sitting on the same site. If they are coming from the same atom, and/or they reside on the same site, the  $k$ -dependence drops out. In  $\text{CeCu}_2\text{Si}_2$ , DFT results showed that the  $f$ -orbitals and the  $d$ -orbitals (conduction electrons) near the Fermi level both come from Ce atom [Ref. 30]. Therefore, the  $k$ -dependence is not present. We kept  $k$ -dependence up to the point which can be done without this assumption. For analytical calculations, we dropped the  $k$ -dependence.

[Note that for attractive potential, the  $k$ -dependence of the potential can give a weak  $k$ -dependence in the gap function, but it still possesses fixed sign gap. Such gaps are often refereed as  $s_{++}$  or extended s-wave etc. [See Alex Aperis and Peter M. Oppeneer, Phys. Rev. B 97, 060501(R) (2018).]

d. The  $e$ ,  $f$  and  $d$  particles are assumed to be fermions. This must be clearly specified.

By the way,  $e$  and  $d$  are bosons (spin = 0), while  $\bar{f}$  states are fermions (spin = 1/2). Their commutation relations also suggest the same. Note also that these Fock states are the fractionalized excitations of the original  $f$ -fermion state. Therefore to have the anticommutation relation of the original  $f$ -state intact,  $\bar{f}$  is fermions,  $e$  and  $d$  have to be bosons. These are consistent with the literature [Refs. 47-49, 51, 53].

e. I do not find the discussion regarding the relation between  $T_K$  and  $v_k$  to be convincing, especially in the light of the  $h$ - $d$ - $e$ - $f$  representation used here. What is the mean value  $n_e$  that is appropriate? When is this justified?

First of all, we do not have any 'h' operator, only f, d, e operators in the main text. Note that  $v_k$  gives Kondo/coherence temperature  $T_K$ , which is well studied in the literature using mean-field theory [see our text in Sec. III. A]. This is traditionally done in the literature [see, e.g., Refs. 45, 54, 55, 57], where d-operators are projected out, e-operators are replaced with its mean-value (scalar). By definition  $n_e = e^\dagger e$  is the number operator for the holons. When we take a mean-value of  $\langle e^\dagger \rangle = \langle e \rangle$  (real number), we get  $\langle e^\dagger \rangle = \langle e \rangle = \sqrt{n_e}$  which is the average number of holons.

With this approximation, the rest of the Hamiltonian can be exactly diagonalized. This opens a band gap between the  $\bar{f}$  and c bands, and  $T_K$  represents the corresponding temperature where the coherence occurs. This is already solved by many groups and we used those results [with appropriate references e.g. Refs. 45, 54, 55, 57] to find a relationship between  $T_K$  and  $T_c$ . Note that  $T_K$  and  $T_c$  are both measured experimentally, therefore, we wanted to point out that our predicted relationship agreed well with the corresponding experimental data.

Referee's comment made us realize that the introduction of  $n_e$  before Eq (12) is rather oddly done and it is not used in the following discussions. Therefore, its better off removing this text, and only pointing out the earlier result of  $T_K$  here with proper references to the corresponding papers.

f. Eq. 14 seems to have a typo with regard to the position of  $\mathbf{a}$ . It should perhaps sit inside the k summation. What is the significance of the primed summation here?

There is not typo here.  $a(\mathbf{q})$  is a function of photon momenta  $\mathbf{q}$ , while  $\mathbf{k}$  gives electron's momentum. Since the summation is over  $\mathbf{k}$ -only, and  $J(\mathbf{q})$  is a function of  $\mathbf{q}$ , there is no summation on  $\mathbf{q}$  in Eq. 14.  $a(\mathbf{q})$  does indeed come out of the  $\mathbf{k}$ -summation [This is shown explicitly in Appendix D (Eqs. D4,D5)]. In any case, we have neglected the photon momentum and set  $\mathbf{q}=0$  only.

"The prime over the summation indicates that the summation is restricted to the first quadrant of the Brillouin zone." as mentioned in Appendix D. We missed to mention the same in the main text and now do so. [This is because we explicitly include the fermions from both  $+\mathbf{k}$  and  $-\mathbf{k}$  in our spinor in Eq. D1 and thus the  $\mathbf{k}$ -summation should be restricted to  $+\mathbf{k}$  only to avoid double counting.]

g. The behaviour shown in Fig. 4 is described as 'exponential decay' in the text. This is not the commonly understood meaning of 'exponential decay'.

We apologize for any confusion here. We meant the exponential behavior at  $T \rightarrow 0$  limit which is indeed the behavior. This is commonly termed so for fully gapped superconductor [see Refs. 23, 24, 43 and also Tinkham book chapter 3 (3.10,3.11)]. We rephrase the 'exponential decay' to 'exponential behavior at  $T \rightarrow 0$ '.

h. Fig. 3 compares experimental data for two materials with calculated  $T_C$  and  $T_K$ . These quantities are calculated using several assumptions that lead to s-wave pairing. Are these assumptions justified in these materials?

As mentioned above, the result of s-wave pairing channel is a natural one due to the fact that the valence fluctuation is isotropic in CeCu<sub>2</sub>Si<sub>2</sub> (onsite valence fluctuations between different orbitals within the same compound). Hence the momentum dependence of the gap naturally drops out. Furthermore, the effective interaction is attractive, and hence the most favorable solution is a fixed-sign pairing symmetry. Therefore, for such a case, s-wave is the natural pairing symmetry.

Note however that for anisotropic potential  $v_k$ , general, low-energy formulas for  $T_c$  and  $T_K$  will remain the same as Eq. 9 and before Eq. 12, respectively. The only change will be in the value of SC coupling constant  $\lambda$ , and  $J_K$ . In these two quantities the  $(v_k)^2$  term will be replaced with a Fermi surface average value of  $\langle v_k \rangle^2$ . Note that,  $\lambda$  and  $J_K$  drops out from Eq. 13 and hence the  $T_c$  vs  $T_K$  relationship remains the same.

i. In my opinion, the discussion section should be expanded to discuss clear experimental signatures of the proposed mechanism.

We thank the referee for the suggestions. We have now expanded the discussion section. We include discussions of two-band SC gap, FFLO superconductivity in the discussion section. We also include now elaborate discussions on how the present theory can also explain the earlier data which predicted unconventional pairing. Finally, we also now include discussion of how our theory compares with other theory of ‘conventional’ pairing in CeCu<sub>2</sub>Si<sub>2</sub>. [Please see newly added paragraphs (2,3,4) in the Discussion section]

i. The discussion of Andreev reflection is not fleshed out. There seem to be strong assumptions about the character of the normal side, e.g., its electrons hybridize with the conduction electrons of the heavy fermion, but not with the local moments.

We thank the referee for the above question. We have made a generic statement about the expected Andreev reflection behavior for the presented superconducting state in a junction with a normal metal. The argument is very simple. When a conduction electron tunnels to the superconductor, it forms pair with a  $f$ -orbital. Therefore, a heavy  $f$ -hole is reflected back to the normal metal. Since the  $f$ -holes are heavy or localized, the Andreev reflection amplitude is strongly suppressed. This serves as a testimony to the theory of pairing between a conduction and local electrons. Otherwise, if the pairing occurs between two conduction electrons, a conduction electron will be reflected back with enhanced reflection amplitude. Otherwise, if the pairing occurs between two localized electrons, there will be no Andreev process when junction is made with a normal metal without  $f$ -orbitals. Given that suppression of Andreev reflection is observed in heavy-fermion compounds [Refs. 62, 19, 20], it serves as a strong proof of local-conduction electron pairing.

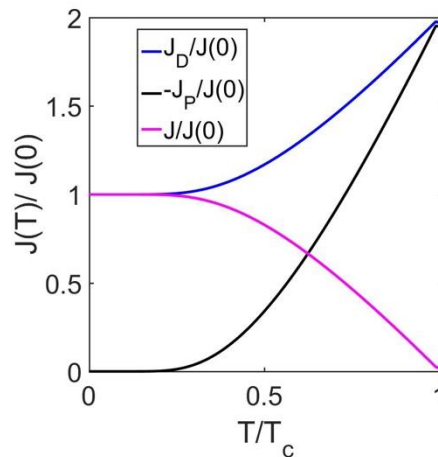
There is no assumption about the hybridization, local moment etc for the normal metal. In fact, the normal metal should not have any local moment or heavy-electrons. It should only have conduction electrons. Any typical metal such as Cu, Al etc should be used for the normal metal.

ii. Are there clear ways to distinguish charge  $e$  vs.  $2e$ ?

This is a good point. People commonly perform flux quantization, noise measurements, or quantum capacitance measurements to estimate the change of Cooper pairs. We note that the proposed Cooper pair here is a bound state of two electrons with total charge  $-2e$ . However, here the  $f$ -electrons do not (orbitally) couple to the magnetic field. Therefore, if the flux quantization and other measurement are done with the magnetic field applied perpendicular to the samples, the Cooper pairs may exhibit single  $-e$  charge. This however comes with the disclaimer that if the  $f$ -orbitals are dispersive then the measurements will show  $-2e$  charge or an average value which lies between  $-e$  to  $-2e$ . This is not an invalidation of the present theory.

iii. The authors state "...we find a complete exclusion of the magnetic field at  $T \rightarrow 0$ ". How is this known? How strongly does this depend on the assumptions made (e.g.,  $s$ -wave pairing)?

We should start with a note that even in other superconductors, the complete exclusion is possible only at  $T=0$  limit where all electrons form Cooper pair and the paramagnetic current is zero. In our case also, we find that [Eq. D5] that the paramagnetic current goes to zero as  $T \rightarrow 0$  and only diamagnetic current is present. This proves that the magnetic field be excluded from the entire sample. We show below the separate  $T$ -dependence of the paramagnetic, diamagnetic and total current. We immediately observe that there is no paramagnetic current and the total current comes from diamagnetic term. Therefore, the system will be fully diamagnetic at  $T \rightarrow 0$  limit.



Finally, we thank the referee again for reading our manuscript and for appreciating its novelty and originality. We are also thankful for all the suggestions and comments. Our answers and revisions indeed helped improve the manuscript for which we are thankful to the referee. Since there is no points remained unaddressed, we hope the referee will be satisfied and recommend for publication of our manuscript.

- Validity: Ok
- Significance: Good

- Originality: Good
- Clarity: Low
- Formatting: Perfect
- Grammar: Below Threshold



# Novel attractive pairing interaction in strongly correlated superconductors

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Conventional and unconventional superconductivity, respectively, arise from attractive (electron-phonon) and repulsive (many-body Coulomb) interactions with fixed-sign and sign-reversal pairing symmetries. Although heavy-fermions, cuprates, and pnictides are widely believed to be unconventional superconductors, recent evidence in former materials [one of the heavy fermion superconductor \(CeCu<sub>2</sub>Si<sub>2</sub>\)](#) indicate the presence of a novel conventional type pairing symmetry beyond the electron-phonon coupling. We present a new mechanism of attractive potential between electrons, mediated by emergent [gauge boson](#) fields (vacuum or holon) in the strongly correlated mixed valence compounds. In the strong coupling limit, localized electron sites are protected from double occupancy, which results in an emergent holon [gauge](#) fields. The holon states can, however, attract conduction electrons through valence fluctuation channel, and the resulting doubly occupied states with local and conduction electrons [condense](#) as Cooper pairs with onsite, fixed-sign, *s*-wave pairing symmetry. We develop the corresponding self-consistent theory of superconductivity, and compare the results with experiments. Our theory provides a new mechanism of superconductivity whose applicability extends to the wider class of intermetallic/mixed-valence materials and other flat-band metals.

## I. INTRODUCTION

Superconductivity arises from the formation of electron-electron pairs, namely, Cooper pairs. Celebrated Bardeen-Cooper-Schrieffer (BCS) theory showed that an effective attractive potential between electrons can emanate from the electron-phonon coupling, resulting in a fully gapped, constant sign superconducting (SC) gap (conventional *s*-wave symmetry).[1] Interestingly, discussions of unconventional superconductivity from repulsive interactions dates back to 1965.[2] It was shown that Cooper pairs can be formed in a repulsive interaction medium, provided the corresponding gap function changes sign in the momentum space[2, 3, 4, 5]. The first heavy-fermion (HF) superconductor CeCu<sub>2</sub>Si<sub>2</sub>[6] was widely believed to be an unconventional superconductor.[7, 8, 9, 10] Subsequently, more HF superconductors,[11] followed by cuprate, and pnictide superconductors are discovered to feature unconventional pairings with either nodal *d*-wave, or nodeless but sign-reversal *s*<sup>±</sup>-pairing symmetry, or their various irreducible combinations.[12]

However, the pairing symmetry, and the pairing mechanism in the [primitive first-discovered heavy-fermion compound CeCu<sub>2</sub>Si<sub>2</sub>](#) are recently called into questions. Earlier reports of nuclear quadrupole resonance (NQR) data revealed a *T*<sup>3</sup> behavior in the relaxation rate without a coherence peak, suggesting the presence of line nodes in the SC gap structure.[13, 14, 15] Observation of four-fold modulation in the upper critical field *H*<sub>c2</sub> in CeCu<sub>2</sub>Si<sub>2</sub> can predict a point-node *d*-wave pairing state[16], provided the Fermi surface (FS) anisotropy is small enough to cause the same modulation.[17] Finally, the observation of a spin resonance in the SC state by inelastic neutron scattering measurement[18] can be interpreted as to arise from sign-reversal of the SC gap [if the resonance peak is very sharp and its energy lies within the SC gap amplitude](#). More recently, counter-evidence of fully gapped

superconductivity are obtained in various measurements including point-contact tunneling spectroscopy,[19, 20] specific heat,[21, 22, 23] magnetic penetration depth,[23, 24] and thermal conductivity[23]. The field-angle dependence of the specific heat data also shows no evidence of gap anisotropy.[22] Furthermore, the observed robustness of superconductivity to disorder supports the absence of sign-reversal in the pairing symmetry scenario.[23, 25] These results collectively signal towards a conventional, fixed-sign, isotropic pairing symmetry in CeCu<sub>2</sub>Si<sub>2</sub>.

CeCu<sub>2</sub>Si<sub>2</sub> has an interesting phase diagram exhibiting two SC domes under pressure, with an antiferromagnetic (AFM) quantum critical point (QCP) [interecepting lying beneath](#) the first SC dome, while a valence fluctuation critical point is possibly present at the second dome.[26, 27, 28] The proximity to the AFM QCP inspires the proposals of spin-fluctuation mediated unconventional, sign-changing pairing symmetry.[24, 29, 30] **The valence fluctuation, which is ubiquitous in HF compounds, can promote superconductivity with unconventional pairing mechanism.**[8, 9, 26, 27, 31, 32] [In particular, it is widely argued by various groups that the vertex correction due to valence-fluctuation exchange can directly mediate a pairing channel,\[9, 31, 32\] or can augment pairing strength arising from other sources\[33, 34\].](#) Kondo coupling can induce various unconventional pairings.[10, 35, 36, 37, 38, 39, 40] Following the overwhelming evidence of conventional pairing symmetry, the electron-phonon coupling problem with strong Coulomb interaction is revisited recently.[33, 41, 42] In general, electron-phonon coupling, if present, can be overturned by the strong onsite Coulomb repulsion in the HF quasiparticles exhibiting effective mass  $\sim 10^3$  times the bare mass.

Our present work is motivated by the question: Can there be other source of attractive potential for superconductivity in general? Here, we provide a new mechanism of attractive potential originating from the interplay between the Coulomb interaction and valence fluctuations. The physical picture is illustrated in Fig. 1. When the Coulomb interaction is strong on the *f*-electron's site, double *f*-electron's occupancy is

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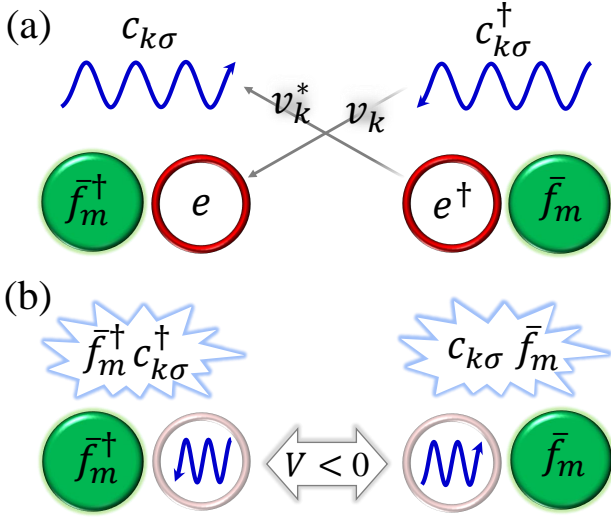


FIG. 1. Illustration of the valence fluctuation mediated attractive potential. (a) The unoccupied state (holon) in each valence fluctuation term can attract another conduction electron through the valence fluctuation channel. The conjugate process also occurs simultaneously. Wavy lines depict conduction electrons ( $c, c^\dagger$ ), while filled ( $\bar{f}, \bar{f}^\dagger$ ) and open ( $e, e^\dagger$ ) circles give singly occupied and unoccupied  $f$ -sites, respectively. Bar symbol over  $f$ -operators emphasizes that they are single- $f$ -electrons occupied states. Arrows dictate valence fluctuation channels. (b) As we integrate out the unoccupied states ( $e, e^\dagger$ ), we obtain an effective interaction  $V < 0$ , forming Cooper pair between the single site  $\bar{f}$ -electron and conduction  $c$  electron.

prohibited. Within the field theory view, a singly occupied  $f$ -electron site is annexed with an unoccupied  $f$ -state – a bosonic holon gauge field – which repels another  $f$ -electrons to occupy the state. However, the unoccupied  $f$ -site can be occupied by a conduction electron since the presence of valence fluctuation channel allows mutation between the  $f$ - and conduction electrons. Remarkably, we show here that the doubly occupied state with  $f$ - and conduction electrons *condense* like a Cooper pair. Mathematically, as we integrate out the gauge boson fields (unoccupied holons), we obtain a robust, new *attractive* potential channel between the conduction electrons and singly occupied  $f$ -sites, naturally commencing on-site, constant sign,  $s$ -wave like superconductivity. Conceptually, this process is somewhat analogous to the theory of meson mediated attractive nuclear force, except here the attraction commences between onsite electrons. We formulate the corresponding theory of superconductivity, and find excellent agreement with the recently observed fully gap, constant sign gap features in CeCu<sub>2</sub>Si<sub>2</sub>, [19, 20, 21, 22, 23, 24, 25] as well as in the Yb-doped CeCoIn<sub>5</sub> superconductors [43]. We predict definite relationship between SC  $T_c$  and valence fluctuation (coherence) temperature  $T_K$ , and other unique properties of the present theory.

## II. THEORY

The low-energy phenomena of HF compounds are well described by the periodic Anderson impurity (PAI) model [44, 45], which has four parts:

$$H = \sum_{\mathbf{k}, \sigma} \xi_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \xi_f \sum_m f_m^\dagger f_m + \sum_{\mathbf{k}, \sigma, m} v_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger f_m + U \sum_m f_m^\dagger f_m f_{-m}^\dagger f_{-m} + \text{h.c.} \quad (1)$$

$c_{\mathbf{k}\sigma}^\dagger$  ( $c_{\mathbf{k}\sigma}$ ) is the creation (annihilation) operator for the conduction electron with spin  $\sigma = \pm 1/2$ . The conduction electron has a dispersion  $\xi_{\mathbf{k}}$ , with  $\mathbf{k}$  being crystal momentum. The strongly correlated  $f$ -electrons are treated as impurity, sitting on each unit cell with an onsite potential  $\xi_f$ . The valence fluctuations between the conduction and correlated electrons lead to a hybridization potential  $v_{\mathbf{k}}$ . Finally,  $f$ -electrons are subjected to a strong Hubbard interaction  $U$ . (The model also holds for narrow ‘band’  $f$ -electrons as long as  $U \gg D_f$ , with  $D_f$  being its bandwidth.) Such a model is well studied in the literature, and can be projected onto the Kondo-lattice model using a Schrieffer-Wolf transformation [46]. Another popular route to solve this problem is the so-called slave-boson approach [47, 48, 49, 50, 51].

The basic phenomenologies of the slave-boson model can be described in two parts. A single  $f$ -orbital on a given site has four Fock states, namely, doubly occupied site ( $d$ ), singly occupied site ( $\bar{f}_m$ ), and unoccupied site ( $e$ ). Clearly,  $d$  and  $e$  operators are bosons, while  $\bar{f}_m$  are fermions, with  $m$  being the spin index (owing to spin-orbit coupling,  $m$  can, in general, have many multiplets). In the  $U \rightarrow \infty$  limit where double occupancy is strictly prohibited, one can project onto the  $d$ -states. [52] The  $f$ -orbitals can be expressed in the remaining three Fock states as  $f_m = e^\dagger \bar{f}_m$  with the constraint  $Q \equiv n_e + n_{\bar{f}} = 1$ , where  $n_e = e^\dagger e$ ,  $n_{\bar{f}} = \sum_m \bar{f}_m^\dagger \bar{f}_m$  are the corresponding number density at every site. [47, 48, 49, 51, 53] Hence we obtain,

$$H = \sum_{\mathbf{k}, \sigma} \xi_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \xi_f \sum_m \bar{f}_m^\dagger \bar{f}_m + \omega_e e^\dagger e + \sum_{\mathbf{k}, \sigma, m} (v_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger e^\dagger \bar{f}_m + v_{\mathbf{k}}^\dagger \bar{f}_m^\dagger e c_{\mathbf{k}\sigma}). \quad (2)$$

We have introduced a gauge onsite potential  $\omega_e > 0$  for the unoccupied state, which arises as a Lagrangian multiplier to conserve the number of  $f$ -electron states to  $Q = 1$  in the  $U \rightarrow \infty$  limit.  $\omega_e$  is considered to be site-independent, respecting the translational invariance, which physically implies that all holons are condensed to the same energy. The renormalized  $\bar{f}$ -electron’s energy is  $\tilde{\xi}_f = \xi_f + \omega_e = Z\xi_f$ , where the corresponding band renormalization factor  $Z$  is defined as  $Z = 1 + \eta$  with  $\eta = \omega_e/\xi_f$ .

Eq. (2) is our starting point in this work. This is not exactly solvable due to the presence of the  $e, e^\dagger$ -states. Popular methods involve hard-core boson model (classical), or mean-field theory around the saddle point of  $\langle e \rangle$  [49, 54, 55]. Here

we include the quantum fluctuations of the holons, and solve Eq. (2) within the quantum field theory approach.

The last term in Eq. (2) implies that each valence fluctuation process generates (or annihilates) a **gauge boson** field  $e^\dagger$  ( $e$ ), whose job is to prohibit double occupancy on the  $f$ -sites. However, the unoccupied states or holons can attract another conduction electron (and vice versa), i.e., they trigger another valence fluctuation process. The two valence fluctuations process can be tied together to generate an effective interaction potential, which turns out to be attractive at low-energy. Mathematically, this is done by integrating out the coherent bosonic  $e$ ,  $e^\dagger$ -operators to obtain an effective interaction potential  $V_{\mathbf{k}\mathbf{k}'}(i\omega_n)$ . Sparing the details to Appendix A, we present the final result of an effective interacting Hamiltonian (in the static limit) as

$$H_{\text{eff}} = \sum_{\mathbf{k},\sigma} \xi_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \bar{\xi}_f \sum_m \bar{f}_m^\dagger \bar{f}_m + \sum_{\mathbf{k}\mathbf{k}',\sigma\sigma',mm'} V_{\mathbf{k}\mathbf{k}'} c_{\mathbf{k}\sigma}^\dagger \bar{f}_m \bar{f}_{m'}^\dagger c_{\mathbf{k}'\sigma'}. \quad (3)$$

Spin conservation leads to  $\sigma+m = \sigma'+m'$ . The most impressive aspect of the above result lies in the form of the effective potential

$$V_{\mathbf{k}\mathbf{k}'}(i\omega_n) = v_{\mathbf{k}} v_{\mathbf{k}'}^\dagger \frac{2\omega_e}{(i\omega_n)^2 - \omega_e^2}, \quad (4)$$

where  $i\omega_n$  is the bosonic Matsubara frequency. In what follows, in the low energy limit  $i\omega_n < \omega_e$  and  $\omega_e > 0$  (since holon's energy is generally positive), Eq. (4) produces an *attractive* potential. This is one of our principle results of this work. As in the case of the BCS theory,[1] we consider here the static limit  $i\omega_n \rightarrow 0$  limit, yielding

$$V_{\mathbf{k}\mathbf{k}'} = -\frac{2v_{\mathbf{k}} v_{\mathbf{k}'}^\dagger}{\omega_e} < 0. \quad (5)$$

For a generic attractive potential, the pair correlation function has a logarithm divergence with temperature (see Appendix C), and we have a SC ground state. Looking at Eq. (3), we find that the Cooper pairs form here between the conduction electron and singly occupied  $\bar{f}_m$ -site with the SC gap parameter defined as

$$\Delta_{\mathbf{k}} = \frac{2v_{\mathbf{k}}}{\omega_e} \sum_{\mathbf{k}'} v_{\mathbf{k}'}^\dagger \langle c_{\mathbf{k}'\sigma} \bar{f}_m \rangle. \quad (6)$$

Here we make few observations. (i) This is an inter-band pairing between the spin- $\frac{1}{2}$  conduction electron and single-site  $f$ -electron with  $m$  multiplet. (ii) The  $\mathbf{k}$ -dependence of the SC gap is solely determined by that of the hybridization term  $v_{\mathbf{k}}$  in Eq. (5). (iii) This is a finite-momentum pairing, but unlike the Fulde-Ferrel-Larkin-Ovchinnikov state (FFLO) or the pair density wave state, here the Cooper pair solely absorbs the conduction electron's momentum. (For dispersive, narrow  $f$ -band, which is often the case in many HF systems, Cooper pairs can have zero center-of-mass momentum.) (iv) The SC state, in general, does not have the particle-hole symmetry, unless at  $\xi_{\mathbf{k}} = \bar{\xi}_f$ . (v) Symmetry of the Cooper pairs ineipently

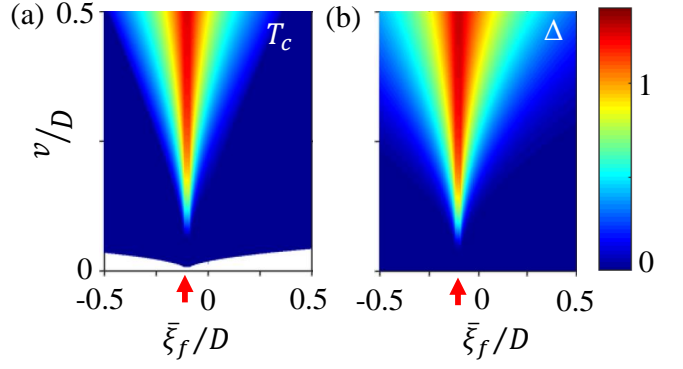


FIG. 2. SC phase diagram with respect to valence fluctuation potential  $v$  and renormalized  $f$ -electron's energy  $\bar{\xi}_f$ . (a), The SC transition temperature  $T_c$  is plotted in the  $(v, \bar{\xi}_f)$  space, scaled with respect to the conduction electron's bandwidth  $D$ . We set  $\xi_f/D = -0.1$ . The white region for small values of  $v$  gives the SC-forbidden region (Eq. (11)). (b), SC gap amplitude  $\Delta$  (at  $T = 0$ ) plotted in the same parameter space. Above the critical value of  $v$ , both  $T_c$  and  $\Delta$  grows with  $v^2$  as in Eq. (9). Interestingly, optimal superconductivity commences at a finite value of  $\bar{\xi}_f$  where all the holon **gauge** fields **condense** to  $\omega_e \rightarrow 0$ , and the pairing potential  $V \rightarrow \infty$ .

relies on is dictated by the values of  $m$ ,  $\sigma$ , and the parity of  $V_{\mathbf{k}\mathbf{k}'}$ . In  $\text{CeCu}_2\text{Si}_2$ , the hybridization occurs between the Ce- $f$  and Ce- $d$  orbitals of the same Ce-atom,[30] and thus the hybridization potential can be considered as onsite, i.e.,  $v_{\mathbf{k}} = v$ . For onsite hybridization, one expects a spin-singlet pair for  $m = \pm 1/2$  (or higher order antisymmetric spin component if  $|m| > 1/2$ ). For an attractive potential, spin-singlet, onsite ( $s$ -wave) pairing state has the highest eigenvalue as obtained in the BCS case as well.[1]

### III. MEAN-FIELD RESULTS AND CRITICAL PHENOMENA

So far, we have obtained all results exactly. We now invoke the mean-field theory for superconductivity. The effective mean-field Hamiltonian reads

$$H_{\text{MF}} = \sum_{\mathbf{k}\sigma} \xi_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \bar{\xi}_f \sum_m \bar{f}_m^\dagger \bar{f}_m + \sum_{\mathbf{k}\sigma m} \Delta_{\mathbf{k}} \bar{f}_m^\dagger c_{\mathbf{k}\sigma}^\dagger + \text{h.c.} \quad (7)$$

The corresponding self-consistent gap equation is (see Appendix B)

$$\Delta_{\mathbf{k}} = \frac{2v_{\mathbf{k}}}{\omega_e} \sum_{\mathbf{k}'} v_{\mathbf{k}'}^* \frac{\Delta_{\mathbf{k}'}}{4E_{0\mathbf{k}'}} \sum_{\nu=\pm} \nu \tanh\left(\frac{\beta E_{\mathbf{k}'\nu}}{2}\right). \quad (8)$$

$\nu = \pm$  are the two quasiparticle bands:  $E_{\mathbf{k}}^\pm = \xi_{\mathbf{k}}^\pm \pm E_{0\mathbf{k}}$ , where  $E_{0\mathbf{k}} = \sqrt{(\xi_{\mathbf{k}}^\pm)^2 + |\Delta_{\mathbf{k}}|^2}$ , and  $\xi_{\mathbf{k}}^\pm = (\xi_{\mathbf{k}} \pm \bar{\xi}_f)/2$ .  $\beta = 1/k_B T$ .

In the case of onsite hybridization  $v_{\mathbf{k}} = v$ , the  $\mathbf{k}$ -dependence of the pairing potential is removed. This gives  $V_{\mathbf{k}\mathbf{k}'} = -\frac{2|v|^2}{\omega_e}$  with  $\omega_e > 0$ , leading to a 'conventional'  $s$ -wave pairing symmetry  $\Delta_{\mathbf{k}} = \Delta$ . Taking advantage of the

onsite attractive potential, and  $s$ -wave pairing channel, we can solve Eq. (8) analytically. Solutions of Eq. (8) in the two asymptotic limits of  $T \rightarrow 0$ , and  $\Delta \rightarrow 0$  yield the gap amplitude  $\Delta$  and  $T_c$  as

$$\begin{aligned} \Delta &= \bar{D}e^{-\frac{1}{2\lambda}} \left[ 1 + re^{-\frac{1}{\lambda}} \right]^{1/2}, \\ k_B T_c &= D_\gamma e^{-\frac{1}{\lambda}} \left[ 1 - \left( \frac{\bar{\xi}_f}{2D_\gamma} \right)^2 e^{\frac{2}{\lambda}} \right]^{1/2}. \end{aligned} \quad (9)$$

Here  $\bar{D} = \sqrt{D^2 - \bar{\xi}_f^2}$ ,  $D_\gamma = 2D\gamma/\pi$  and  $r = (D + \bar{\xi}_f)/(D - \bar{\xi}_f)$ , with  $\gamma$  being the Euler constant, and  $D = 1/2N$ , and  $N$  are bandwidth and density of states of conduction electrons at the Fermi level. The SC coupling constant is defined as

$$\lambda = \frac{2N|v|^2}{\omega_e} = 2|\eta|^{-1}NJ_K, \quad (10)$$

where  $J_K = |v|^2/|\xi_f|$  is the Kondo coupling constant.  $\eta$  is defined below Eq. (2). The first terms before the parenthesis in both  $\Delta$  and  $T_c$  are the usual BCS solutions, while the correction terms within the parenthesis have important consequences. The correction term in Eq. (9) suggests that superconductivity arises above a critical value of the coupling constant

$$\frac{1}{\lambda} < \ln \left( \frac{2D_\gamma}{|\bar{\xi}_f|} \right). \quad (11)$$

This implies that there exists a lower critical value of the hybridization  $v_c$  above which superconductivity is possible. Since  $v$  is related to the coherence temperature  $T_K$ , we show below that the above constraint translates into a lower limit for  $T_K$  to produce superconductivity. This result is in contrast to the BCS result where any infinitesimal electron-phonon coupling is sufficient for finite  $T_c$ . Interestingly, the BCS ratio  $\Delta/k_B T_c$  is not a universal constant here, even in the weak coupling limit. In the limit of  $D \gg \bar{\xi}_f$ , we recover BCS-type behavior of  $\Delta \rightarrow De^{-1/2\lambda}$ , and  $k_B T_c \rightarrow D_\gamma e^{-1/\lambda}$ , with  $\Delta/k_B T_c \rightarrow 1.73e^{1/2\lambda}$ , suggesting a strong coupling limit of the superconductivity.

Plots of  $\Delta$  and  $T_c$  as a function of  $v$  and  $\bar{\xi}_f$  are shown in Fig. 2. Both phase diagrams exhibit funnel like behavior in the  $v - \bar{\xi}_f$  space. We highlight here two key features. (i) In  $T_c$  plot we find a white region for small values  $v$  which marks the forbidden (non-SC) region dictated by the constraint  $1/v^2 > (N/2\omega_e) \ln |2D_\gamma/\bar{\xi}_f|$  (Eq. (11)). In the rest of the regions where both  $\Delta$  and  $T_c$  are finite, we obtain a second order phase transition with the critical exponent of  $1/2$ . (ii) Secondly, superconductivity is optimal at a characteristic value of  $\bar{\xi}_f \neq 0$  (marked by arrows in Fig. 2). At this point  $\omega_e \rightarrow 0$  ( $\xi_f = \bar{\xi}_f$ ) and hence the pairing potential  $V \rightarrow \infty$ , stipulating maximum superconductivity. At the optimal  $T_c$ ,  $f$ -electron's band renormalization  $Z \rightarrow 1$ .

#### A. Connection to coherence temperature $T_K$ .

From Eq. (4), it is evident that  $\omega_e$  is analogous to the Debye frequency of the electron-phonon mechanism. The es-

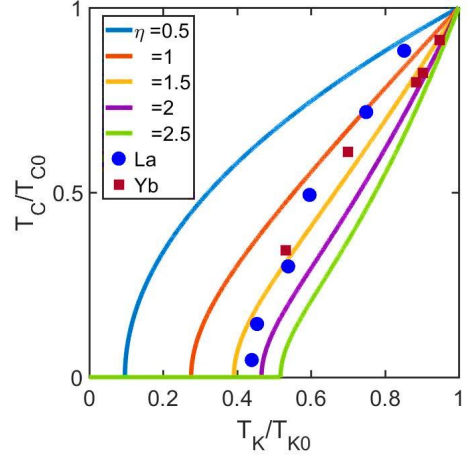


FIG. 3. Relationship between  $T_c$  and  $T_K$ . We demonstrate the relationship between  $T_c$  and  $T_K$  for several values of the exponent  $\eta$  (from Eq. (13)). Interestingly,  $T_c$  vanishes below some critical value of  $T_K$ , where the cutoff value decreases with decreasing  $\eta$ .  $T_c$ ,  $T_K$  are normalized to some highest values of  $T_{c0}$ ,  $T_{K0}$ , respectively, for each values of  $\eta$ . For CeCoIn<sub>5</sub>, Yb and La dopings[56] are known to modulate the valence fluctuation strength  $T_K$ , giving an intriguingly similar  $T_c$  versus  $T_K$  relationship, as predicted by our theory in Eq. (13). Experimental values agree well for  $\eta \sim 1 - 1.5$  for  $\xi_f = 0.7\text{eV}$ .

sential dependence of  $T_c$  and  $\lambda$  on observable parameters such as coherence temperature  $T_K$  can be derived using the saddle point approximation[49, 54, 55]. near a mean value of  $\langle e \rangle = \langle e^\dagger \rangle = \sqrt{n_e}$ . For this case, Eq. (2) can be solved exactly,[57] yielding  $k_B T_K = De^{-1/NJ_K}$ . Therefore, from Eq. (10), we find that the SC coupling constant  $\lambda$  depends on  $T_K$  as

$$\frac{1}{\lambda} = \eta \ln \left( \frac{D}{k_B T_K} \right). \quad (12)$$

This result is consistent with the fact that the Kondo critical point prompts optimal superconductivity as obtained in CeCu<sub>2</sub>Si<sub>2</sub>,[26] as well as in many other HF superconductors.[8, 9, 11, 58, 59, 60] However,  $T_c$  is terminated below a critical  $T_K$  which can be obtained from Eq. (9) as

$$(k_B T_c)^2 = D_\gamma^2 \left( \frac{k_B T_K}{D} \right)^{2|\eta|} - \frac{\bar{\xi}_f^2}{4}, \quad (13)$$

where  $\eta$  is the same as before. Eq. (13) is another important result of our theory, which finds a surprisingly consistent agreement with experimental data (see Fig. 3). We plot  $T_c$  and  $T_K$  for several parameter values in Fig. 3. Both the critical behavior and the power-law dependence between  $T_c$  and  $T_K$  agree remarkably well with the experimental data of La, and Yb doped CeCoIn<sub>5</sub> samples.[56]

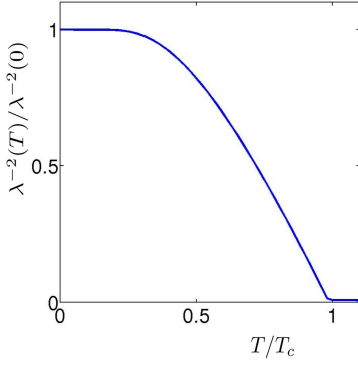


FIG. 4. Computed superfluid density as a function of temperature. The temperature dependence shows a typical exponential behavior at low- $T$  as seen in  $\text{CeCu}_2\text{Si}_2$ .

#### IV. SIGNATURES OF PAIRING STRUCTURE.

##### A. Meissner effect

Unlike the typical Cooper pair of two conduction electrons with opposite momenta in other types of superconductors, here we have a pairing between conduction electron and correlated singly occupied  $f$ -electrons. The conduction electrons directly couple to the gauge field  $\mathbf{A}$  as  $\mathbf{p}' = \hbar\mathbf{k} - \frac{e}{c}\mathbf{A}$ . On the other hand, the  $f$ -states do not couple to the vector potential in its localized limit. Importantly, despite that the magnetic field couples only to the conduction electron, we find a complete exclusion of the magnetic field at  $T \rightarrow 0$ , a hallmark of superfluid state. Interestingly, however, in the strongly localized limit of the  $f$ -orbitals, the Meissner effect experiments will exhibit charge of the Cooper pair to be  $-e$ , instead of  $-2e$  as in other Conventional Cooper pair between two itinerant electrons. Caution to be taken in realistic heavy-fermion systems, where the band structure calculation[29] shows weak dispersion of the  $f$ -electrons, which couple to the external gauge field, and hence may contribute to the Cooper pair charge of  $-2e$  or a value between  $-e$  to  $-2e$  on average.

Here we proceed with computation of the diamagnetic ( $\mathbf{J}_d$ ) and paramagnetic ( $\mathbf{J}_p$ ) current of the conduction electrons only:

$$\mathbf{J}_d = \frac{e^2 \mathbf{a}}{c} \sum_{\mathbf{k}\sigma} \frac{1}{m_{\mathbf{k}}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}, \quad \mathbf{J}_p = e \sum_{\mathbf{k}\sigma} \mathbf{v}_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}. \quad (14)$$

$\mathbf{v}_{\mathbf{k}}$  and  $m_{\mathbf{k}}$  are the velocity and effective mass, respectively, of the conduction electron, and  $\mathbf{a}$  is the Fourier component of the vector potential  $\mathbf{A}$ . Using the mean-field solution of the quasiparticle bands, the superfluid density (inversely proportional to the magnetic penetration depth) is obtained to be

$$\lambda_{ij}^{-2}(T) = \frac{4\pi e^2}{c^2} \sum_{\mathbf{k}} \left[ \frac{1}{m_{ij,\mathbf{k}}} \left( 1 - \sum_{\nu} (\alpha_{\mathbf{k}}^{\nu})^2 \tanh\left(\frac{\beta E_{\mathbf{k}}^{\nu}}{2}\right) \right) - \frac{\beta}{2} v_{i\mathbf{k}} v_{j\mathbf{k}} \sum_{\nu} (\alpha_{\mathbf{k}}^{\nu})^2 \text{sech}^2\left(\frac{\beta E_{\mathbf{k}}^{\nu}}{2}\right) \right], \quad (15)$$

$\nu = \pm$  for two quasiparticle bands. [Prime symbol over the summation indicates that it is restricted within the first quadrant of the Brillouin zone, since both  $+\mathbf{k}$  and  $-\mathbf{k}$  fermions are included exclusively to obtain Eq. (D2), (D3) (see Appendix D).]  $(\alpha_{\mathbf{k}}^{\mp})^2 = \frac{1}{2} \left( 1 \mp \frac{\xi_{\mathbf{k}}^+}{E_{0\mathbf{k}}} \right)$  is the coherence factors of the mean-field solutions. The numerical evaluation of Eq. 15 yields an exponential behavior of superfluid density as  $T \rightarrow 0$ , as shown in Fig. 4. This behavior is also observed experimentally in  $\text{CeCu}_2\text{Si}_2$  [23, 24] as well as in Yb-doped  $\text{CeCoIn}_5$ [43].

##### B. Spin-resonance mode

For unconventional pairing symmetry, the sign-reversal of the SC gap leads to a spin-resonance mode at  $\omega_{\text{res}} \leq 2\Delta$ . [12] Such a mode is rather weak in intensity and may lie above  $2\Delta$  for conventional (fixed sign) pairing symmetry.[61] Experimentally, a resonance is observed in the SC state in  $\text{CeCu}_2\text{Si}_2$  at  $\mathbf{Q} \sim (0.215, 0.215, 1.458)$  in r.l.u. in the energy scale of  $\sim 0.2$  meV which is roughly at  $4k_B T_c$  ( $T_c \sim 0.6$  K).[18]

The present pairing symmetry has few interesting collective spin modes which can explain the above experimental behavior. For the calculation of spin fluctuation to be tractable we consider that the  $f$ -electrons possess spin  $m = \pm 1/2$ . In this case, the total spin operator can be defined as a summation over conduction spin and  $f$ -electrons spin:

$$\mathbf{S}_{\mathbf{q}} = \frac{1}{2} \left( \sum_{\mathbf{k}\alpha\beta} c_{\mathbf{k}\alpha}^\dagger \boldsymbol{\sigma}_{\alpha\beta} c_{\mathbf{k}+\mathbf{q}\beta} + \sum_{\alpha\beta} \bar{f}_{\alpha}^\dagger \boldsymbol{\sigma}_{\alpha\beta} \bar{f}_{\beta} \right). \quad (16)$$

$\alpha, \beta$  are spin indices. The transverse spin susceptibility is defined as  $\chi(\mathbf{q}, \tau) = \langle T_{\tau} S^+(\mathbf{q}, \tau) S^-(\mathbf{-q}, 0) \rangle$ . Solving in the mean-field SC state, we obtain

$$\chi(\mathbf{q}, i\omega_n) = \sum_{\mathbf{k}} \sum_{\mu, \nu = \pm} A_{\mathbf{k}\mathbf{q}}^{\mu\nu} \frac{f(E_{\mathbf{k}+\mathbf{q}}^{\mu}) - f(E_{\mathbf{k}}^{\nu})}{i\omega_n + E_{\mathbf{k}}^{\nu} - E_{\mathbf{k}+\mathbf{q}}^{\mu}}, \quad (17)$$

where

$$A_{\mathbf{k}\mathbf{q}}^{\mu\nu} = \frac{1}{2} \left( 1 \pm \frac{\xi_{\mathbf{k}}^+ \xi_{\mathbf{k}+\mathbf{q}}^+ + \Delta_{\mathbf{k}} \Delta_{\mathbf{k}+\mathbf{q}}}{E_{0\mathbf{k}+\mathbf{q}} E_{0\mathbf{k}}} \right), \quad (18)$$

$\mu, \nu = \pm$  are the band indices, and  $\pm$  in Eq. (18) corresponds to amplitude of the oscillators for  $\mu = \nu$  (intra-) and  $\mu \neq \nu$  (inter-) quasiparticle band transition. Eq. (17) can give various collective excitations, depending on the band structure details. We are here interested in the possible modes inside the SC gap. Indeed, we find the solution of a localized spin-excitation in the SC state at a wavevector which corresponds to the condition  $\xi_{\mathbf{k}}^+ = -\xi_{\mathbf{k}+\mathbf{Q}}^+$ . (Note that this is not the condition of the conduction electron's FS nesting). In this case, we have a resonance at an energy

$$\omega_{\text{res}} = E_{\mathbf{k}+\mathbf{Q}}^+ - E_{\mathbf{k}}^- \sim \frac{2\Delta^2}{|\bar{\xi}_f|}, \quad (19)$$

in the limit of  $\Delta \gg \xi_{\mathbf{k}}^+$ . The corresponding oscillator strength of the resonance mode is  $A_{\mathbf{k}\mathbf{q}}^{\mu, \nu \neq \mu} = (\xi_{\mathbf{k}}^+)^2 / E_{0\mathbf{k}}^2 > 0$ . Since  $\bar{\xi}_f > \Delta$ , the resonance occurs inside the SC gap, as observed experimentally in  $\text{CeCu}_2\text{Si}_2$ [18].

### C. Other measurements

The present theory of valence fluctuation mediated attractive pairing channel can be verified in multiple ways. For example, the present theory predicts a unique Andreev reflection behavior. In a typical normal metal and superconductor interface, as an electron tunnels from the metal into the superconductor side, it reflects back a hole, and vice versa. In our present case, the conduction electron from the normal metal forms a Cooper pair with a  $f$ -state in the SC sample, and thus *reflects a  $f$ -electron to the normal metal*, which can be easily probed. The reflection probably is inversely proportional to the effective mass of the  $f$ -electron. This means in the limit of the localized  $f$ -electron case, the Andreev reflection can be strongly suppressed or absent. **A suppression of Andreev reflection amplitude is observed in CeCoIn<sub>5</sub>,[62] and CeCu<sub>2</sub>Si<sub>2</sub> [19, 20].**

As also mentioned in the above section, in the limit of fully localized  $f$ -orbitals when the coupling to the external gauge field is suppressed, one may find evidence of  $-e$  charge of the Cooper pair in such experiments. However, the band structure effect of the  $f$ -orbitals can help coupling of the  $f$ -orbitals to the gauge field and hence the charge of the Cooper pair on average can be observed to be somewhere between  $-e$  to  $-2e$  in experiments.

## V. DISCUSSIONS AND CONCLUSIONS

Our theory demonstrates the existence of an attractive pairing potential mediated by the interplay between Coulomb interaction and valence fluctuations. The origin of the attractive potential is the emergent **gauge boson** field (holon) associated with single-site  $f$ -states to restrict double occupancy due to strong Coulomb interaction. The effective interaction is a result of multiple valence fluctuations: The holon field generated in a given valence fluctuation is absorbed in the second valence fluctuation, and the resulting two valence fluctuation processes generate an effective interaction between the  $f$ - and conduction electrons. The interaction is attractive at low-frequency and isotropic in the case of onsite valence fluctuation process. The onsite, attractive interaction naturally gives an isotropic, constant sign  $s$ -wave pairing channel between the single-site  $f$ -electrons, and conduction electrons.

Our result of fixed-sign, isotropic  $s$ -wave pairing channel is consistent with numerous experimental data discussed in the introduction.[19, 20, 21, 22, 23, 24, 25] The exponential temperature dependence of point-contact tunneling spectroscopy,[19, 20] specific heat,[21, 22, 23] thermal conductivity[23], and penetration depth[23, 24] are naturally explained within our model. Moreover, there have been several recent evidence of two-band superconductivity in CeCu<sub>2</sub>Si<sub>2</sub>. [21, 22, 24] It was shown that most of the above data, as well as the  $T^3$  dependence of the NQR data[13, 14, 15] can be fitted well with a two-band model with a simple  $s$ -wave pairing symmetry. This is fully consistent with our theory which has a two-band (conduction and local) behavior with  $s$ -wave pairing. Furthermore, the pro-

posed pairing (Eq. (6)) is a finite momentum pairing in the limit of fully localized  $f$ -electrons, and itinerant conduction electrons. Consistently, there have been recent evidence of finite momentum pairing state in CeCu<sub>2</sub>Si<sub>2</sub>. [63] Finally, strong suppression of Andreev reflection amplitude in CeCoIn<sub>5</sub>, [62] and CeCu<sub>2</sub>Si<sub>2</sub> [19, 20] are well known, suggesting the involvement of the localized  $f$ -orbitals in the Cooper pairs.

In addition, the present theory can also explain the other three experimental signatures which were taken earlier as evidence of unconventional, sign-reversal pairing symmetry. (i) The  $T^3$  dependence of the NQR relaxation rate  $1/T_1$  below  $T_c$  in CeCu<sub>2</sub>Si<sub>2</sub> is often considered as evidence of line nodes in the SC gap structure. [13, 14, 15] As mentioned above, a two-band model with purely  $s$ -wave gap, as in the present case, is shown to reproduce the same power-law behavior of  $1/T_1$  without invoking gap nodes. [21, 22] Therefore, we anticipate our theory is equally applicable here. (ii) The four-fold angular modulation of  $H_{c2}$  in CeCu<sub>2</sub>Si<sub>2</sub> [16] can be a signature of the SC gap anisotropy. However, it was shown in a realistic two-band model that a strong anisotropy in  $H_{c2}$  (as well as in other quantities) can well arise solely from the Fermi surface anisotropy even for a purely isotropic  $s$ -wave SC gap. [17] Indeed, the conduction electron's Fermi surface is known to be substantially anisotropic in CeCu<sub>2</sub>Si<sub>2</sub>. [29, 30] (iii) Finally, it is known that a spin-resonance as measured by inelastic neutron scattering experiments can arise either from unconventional, sign-reversal pairing symmetry, or even for a fixed-sign  $s$ -wave pairing. [61] For sign-reversal pairing gap, the spin-resonance is typically very sharp and its energy must follow  $\omega_{\text{res}} < 2\Delta$ , where  $\Delta$  is the SC gap amplitude. On the other hand, for fixed-sign, conventional pairing, the resonance is usually very broad, and its energy lies at  $\omega_{\text{res}} \geq 2\Delta$ . The measured spin-resonance in CeCu<sub>2</sub>Si<sub>2</sub> [18] is indeed quite broad, and the present data cannot discern if the resonance energy lies below or above  $2\Delta$ . Moreover, our theory also predicts a novel resonance mode at an energy (Eq. (19)) determined by  $2\Delta/\xi_f$ .

We compare and contrast the concepts of the present theory with the prior theories of 'conventional' pairing solutions in CeCu<sub>2</sub>Si<sub>2</sub>. Valence fluctuation mediated or assisted pairing mechanism has been a steady theme of discussions in the heavy-fermions community. [8, 9, 26, 27, 31, 32, 33, 34] Miyake and Onishi [31, 32] have proposed a phenomenological pairing vertex formula with the help of an empirical valence fluctuation susceptibility defined near its critical point. Unlike our case, the pairing vertex in Ref. [31] does not invoke electron-electron correlation, however, the pairing interaction is argued to be retarded when correlation is included. On the other hand, in our case, the pairing interaction is microscopically derived from the interplay between correlation and valence fluctuation and has a robust solution of attractive channel at the low-energy limit. Our pairing interaction can be considered as a generalized, dynamical Kondo interaction. If we express the interaction in Eq. (3) in terms of local spin and conduction spin interaction, then  $V_{\mathbf{k}\mathbf{k}'}(\omega)$  can be cast as dynamical Kondo interaction  $J_K(\omega)$  (similar result in the static limit can be obtained within the Schrieffer-Wolf transformation[46]). Starting from Kondo interaction

with  $J_K < 0$ , a composite Cooper pair theory was proposed where conduction electron pairs up the (chargeless) fermionic representation of the local spin.[36, 38] Such composite pairing channel is also  $s$ -wave like in the limit of local Kondo channel. A prior quantum Monte Carlo simulation of periodic Anderson model showed the existence of a  $s$ -wave pairing interaction.[35] This gives a validation of the attractive pairing interaction we derive in Eq. (4). Finally, we propose that a future dynamical mean-field theory (DMFT) calculation will be valuable to further confirm the existence of the attractive pairing solution in such a model.

Finally, we make few remarks about the future extension of the present theory. A full, self-consistent treatment of  $T_c$ ,  $\eta$ , and  $T_K$  requires an Eliashberg-type formalism. Since  $T_c$  is significantly low in HF compounds, the present mean-field treatment is however a good approximation for the estimates of  $T_c$ . The theory also holds for dispersive  $f$ -electrons state as long as the corresponding bandwidth is much lower than  $U$ . For a dispersive  $f$ -state, one can obtain a zero center-of-mass momenta Cooper pair  $\langle c_{\mathbf{k}\sigma}^\dagger \bar{f}_{-\mathbf{k}m}^\dagger \rangle$ . Therefore, the present theory is applicable to the wider class of intermetallic and mixed valence superconductors where a narrow band and a conduction band coexist, and possesses finite interband tunneling (valence fluctuation) strength.[64] Our calculation does not include Coulomb interaction between the conduction and  $f$ -electrons (the Falicov-Kimball type interaction). However, it is obvious that such a Coulomb interaction term will lead to a pair breaking correction (e.g  $\mu^*$ -term), in analogy with the Coulomb interaction correction to the electron-phonon coupling case (the so-called McMillan's formula)[65]. Finally, the vertex correction to the pairing potential can be envisaged, in analogy with the Migdal's theory, to scale as  $m/M$ , where  $m$ , and  $M$  are the mass of the conduction and  $f$ -electrons. Since  $M \sim 10^3$  in these HF systems, we argue that the vertex correction can be negligible.

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### Appendix A: Field theory treatment of the hole states and effective attractive potential

The action of the Hamiltonian in Eq. (2) is broken into four components

$$S = S_c + S_{\bar{f}} + S_e + S_v, \quad (\text{A1})$$

where

$$S_c = \int d\tau \sum_{\mathbf{k}, \sigma} \tilde{c}_{\mathbf{k}\sigma}(\tau) (\partial_\tau + \xi_{\mathbf{k}}) c_{\mathbf{k}\sigma}(\tau), \quad (\text{A2})$$

$$S_{\bar{f}} = \int d\tau \sum_m \tilde{\bar{f}}_m(\tau) (\partial_\tau + \xi_{\bar{f}}) \bar{f}_m(\tau), \quad (\text{A3})$$

$$S_e = \int d\tau \tilde{e}(\tau) (\partial_\tau + \omega_e) e(\tau), \quad (\text{A4})$$

$$S_v = \int d\tau \sum_{\mathbf{k}, \sigma, m} (v_{\mathbf{k}} \tilde{c}_{\mathbf{k}\sigma}(\tau) \tilde{e}(\tau) \bar{f}_m(\tau) + \text{h.c.}). \quad (\text{A5})$$

Here  $\tilde{e}, e$  are bosonic coherent states and  $\tilde{\bar{f}}, \bar{f}, \tilde{c}, c$  are Grassmann variables for singly occupied  $f$ -states, and conduction electrons respectively ('tilde' means conjugation).  $\tau$  is imaginary time axis. Thermodynamic properties of the system can be calculated from the partition function  $\mathcal{Z} = \text{Tr} e^{-\mathcal{S}}$ , where the trace is taken over all degrees of freedom of the system. We obtain an effective action  $\mathcal{S}_{\text{eff}}$  by integrating out the bosonic variables  $\tilde{e}, e$  as

$$\begin{aligned} \mathcal{Z} &= \int \mathcal{D}[\tilde{c}, c] \mathcal{D}[\tilde{\bar{f}}, \bar{f}] \mathcal{D}[\tilde{e}, e] e^{-S_c - S_{\bar{f}} - S_e - S_v}, \\ &= \int \mathcal{D}[\tilde{c}, c] \mathcal{D}[\tilde{\bar{f}}, \bar{f}] e^{-S_c - S_{\bar{f}}} \int \mathcal{D}[\tilde{e}, e] e^{-S_e - S_v}, \\ &= \int \mathcal{D}[\tilde{c}, c] \mathcal{D}[\tilde{\bar{f}}, \bar{f}] e^{-S_{\text{eff}}[\tilde{c}, c, \tilde{\bar{f}}, \bar{f}]}, \end{aligned} \quad (\text{A6})$$

where

$$S_{\text{eff}} = S_c + S_{\bar{f}} - \ln \int \mathcal{D}[\tilde{e}, e] e^{-S_e - S_v}. \quad (\text{A7})$$

It is easier to perform the  $\tau$  integration in the Matsubara frequency space. The Fourier transformation to the Matsubara frequency domain of the  $e(\tau)$  variable gives  $e(\tau) = \frac{1}{\sqrt{\beta}} \sum_n e_n \exp(-i\omega_n \tau)$ , where  $i\omega_n$  is bosonic Matsubara frequency and  $e_n = e(i\omega_n)$ . In the Matsubara space, we get

$$S_e = - \sum_n \tilde{e}_n (\mathcal{G}^e)^{-1}(i\omega_n) e_n, \quad (\text{A8})$$

where  $\mathcal{G}^e$  is the bare Green's function for the  $e_n$ -states:  $(\mathcal{G}^e)^{-1} = i\omega_n - \omega_e$ .

Next we define a bosonic hybridization field  $\rho_{\mathbf{k}\sigma m}$  as

$$\rho_{\mathbf{k}\sigma m}(\tau) = \tilde{c}_{\mathbf{k}\sigma}(\tau) \bar{f}_m(\tau), \quad (\text{A9})$$

whose Fourier component is  $\rho_{\mathbf{k}\sigma m}(\tau) = \frac{1}{\sqrt{\beta}} \sum_n \rho_{\mathbf{k}\sigma m, n} \exp(-i\omega_n \tau)$ , where  $\rho_{\mathbf{k}\sigma m, n} = \rho_{\mathbf{k}\sigma m}(i\omega_n)$  with  $i\omega_n$  being the bosonic Matsubara frequency. Hence we can express the hybridization action as

$$\begin{aligned} S_v &= \int_0^\beta d\tau \sum_{\mathbf{k}, \sigma, m} (v_{\mathbf{k}} \tilde{e}(\tau) \rho_{\mathbf{k}\sigma m}(\tau) + v_{\mathbf{k}}^* \tilde{\rho}_{\mathbf{k}\sigma m}(\tau) e(\tau)), \\ &= \sum_{\mathbf{k}, \sigma, m} \sum_n (v_{\mathbf{k}} \tilde{e}_n \rho_{\mathbf{k}\sigma m, n} + v_{\mathbf{k}}^* \tilde{\rho}_{\mathbf{k}\sigma m, n} e_n). \end{aligned} \quad (\text{A10})$$

Interestingly, now in Eqs. (A8), (A10) the integration over  $\tau$ -variable is replaced with summation over discrete Matsubara frequencies  $n$ . Let us say at a given temperature we

have  $N$  number of Matsubara frequencies. So we define a bosonic spinor  $\mathbf{E} = (e_1, e_2, \dots, e_N)^T$ , and  $\tilde{\mathbf{E}} = (\tilde{e}_1, \tilde{e}_2, \dots, \tilde{e}_N)$ . Similarly, we define a vector for the hybridization field as  $\mathbf{V} = (\mathbf{v}_1, \mathbf{v}_2, \dots, \mathbf{v}_N)^T$ ,  $\tilde{\mathbf{V}} = (\tilde{\mathbf{v}}_1, \tilde{\mathbf{v}}_2, \dots, \tilde{\mathbf{v}}_N)$  where  $\mathbf{v}_n = \sum_{\mathbf{k}\sigma m} v_{\mathbf{k}} \rho_{\mathbf{k}\sigma m, n}$ , and  $\tilde{\mathbf{v}}_n = \sum_{\mathbf{k}\sigma m} v_{\mathbf{k}}^* \tilde{\rho}_{\mathbf{k}\sigma m, n}$ . Finally, we define a diagonal matrix  $\mathbf{G}^{-1}$  for the inverse Green's function  $(\mathcal{G}^e)^{-1}$  in Eq. (A8), whose components are  $\mathbf{G}_{nn}^{-1} = (\mathcal{G}_e)^{-1} = i\omega_n - \omega_e$ . Hence we can express Eqs. (A8),(A10) respectively as

$$\mathcal{S}_e = -\tilde{\mathbf{E}} \cdot \mathbf{G}^{-1} \cdot \mathbf{E}, \quad (\text{A11})$$

$$\mathcal{S}_v = \tilde{\mathbf{E}} \cdot \mathbf{V} + \tilde{\mathbf{V}} \cdot \mathbf{E}. \quad (\text{A12})$$

Therefore, the last term of Eq. (A7) can be evaluated as

$$\int \mathcal{D}[\tilde{\mathbf{E}}, \mathbf{E}] e^{-\mathcal{S}_e - \mathcal{S}_v} = \pi^N \det \mathbf{G}^{-1} e^{-[\tilde{\mathbf{V}} \cdot \mathbf{G}^{-1} \cdot \mathbf{V}]}. \quad (\text{A13})$$

(We ignored some irrelevant constant factors). The factor of the exponent on the right hand side of Eq. (A13) can now be evaluated rigorously. In  $T \rightarrow 0$  limit, the Matsubara frequencies span from  $n = -\infty$  to  $\infty$ . Hence we obtain,

$$\begin{aligned} & \tilde{\mathbf{V}} \cdot \mathbf{G}^{-1} \cdot \mathbf{V} \\ &= - \sum_{\substack{\mathbf{k}, \sigma, m \\ \mathbf{k}', \sigma', m'}} \sum_{n=-\infty}^{\infty} v_{\mathbf{k}}^* \tilde{\rho}_{\mathbf{k}\sigma m, n} \frac{1}{-i\omega_n + \omega_e} v_{\mathbf{k}'} \rho_{\mathbf{k}\sigma' m', n} \\ &= \sum_{\substack{\mathbf{k}, \sigma, m \\ \mathbf{k}', \sigma', m'}} \sum_{n=0}^{\infty} v_{\mathbf{k}}^* v_{\mathbf{k}'} \frac{2\omega_e}{(i\omega_n)^2 - \omega_e^2} \tilde{\rho}_{\mathbf{k}\sigma m, n} \rho_{\mathbf{k}\sigma' m', n} \\ &= \sum_{\substack{\mathbf{k}, \sigma, m \\ \mathbf{k}', \sigma', m'}} \sum_{n=0}^{\infty} V_{\mathbf{k}\mathbf{k}'} \tilde{f}_m(i\omega_n) c_{\mathbf{k}, \sigma}(i\omega_n) \tilde{c}_{\mathbf{k}', \sigma'}(i\omega_n) \bar{f}_{m'}(i\omega_n). \end{aligned} \quad (\text{A14})$$

In the last equation, we have substituted the hybridization field into fermionic field from Eq. (A9). The effective potential is

$$V_{\mathbf{k}\mathbf{k}'} = v_{\mathbf{k}}^* v_{\mathbf{k}'} \frac{2\omega_e}{(i\omega_n)^2 - \omega_e^2}. \quad (\text{A15})$$

## Appendix B: Mean-field solutions

We use the Nambu-Gorkov basis  $\psi_{\mathbf{k}} = (c_{\mathbf{k}\sigma} \quad \bar{f}_m^\dagger)^T$ , in which the mean-field Hamiltonian (Eq. (7)) reads

$$H_{\text{MF}}(\mathbf{k}) = \xi_{\mathbf{k}}^- I_{2 \times 2} + \xi_{\mathbf{k}}^+ \sigma_z - \Delta_{\mathbf{k}} \sigma_x, \quad (\text{B1})$$

where  $\sigma_i$  are the  $2 \times 2$  Pauli matrices and  $I_{2 \times 2}$  is a unit matrix.  $\xi_{\mathbf{k}}^\pm = (\xi_{\mathbf{k}} \pm \bar{\xi}_f)/2$ . The BdG eigenvalues are

$$E_{\mathbf{k}}^\pm = \xi_{\mathbf{k}}^- \pm E_{0\mathbf{k}}, \quad \text{with } E_{0\mathbf{k}} = \sqrt{(\xi_{\mathbf{k}}^+)^2 + |\Delta_{\mathbf{k}}|^2}. \quad (\text{B2})$$

The Bogoliubov operators for the two eigenvalues  $E_{\mathbf{k}}^\pm$  are

$$\begin{pmatrix} \phi_{\mathbf{k}}^+ \\ (\phi_{\mathbf{k}}^-)^\dagger \end{pmatrix} = \begin{pmatrix} \alpha_{\mathbf{k}}^+ & -\alpha_{\mathbf{k}}^- \\ \alpha_{\mathbf{k}}^- & \alpha_{\mathbf{k}}^+ \end{pmatrix} \begin{pmatrix} c_{\mathbf{k}\sigma} \\ \bar{f}_m^\dagger \end{pmatrix}. \quad (\text{B3})$$

where

$$(\alpha_{\mathbf{k}}^\mp)^2 = \frac{1}{2} \left( 1 \mp \frac{\xi_{\mathbf{k}}^+}{E_{0\mathbf{k}}} \right), \quad (\text{B4})$$

Evaluating the self-consistent gap equation from Eq. (6), we get Eq. (8).

### 1. Transition temperature $T_c$

For the attractive potential, onsite pairing is more favorable. Hence we set  $V_{\mathbf{k}\mathbf{k}'} = -2|v|^2/\omega_e$ . In this case, superconducting transition temperature  $T_c$  can be obtained by taking the limits of  $\Delta \rightarrow 0$ , which renders  $E_{\mathbf{k}}^+ \rightarrow \xi_{\mathbf{k}}$ ,  $E_{\mathbf{k}}^- \rightarrow -\bar{\xi}_f$ ,  $E_{0\mathbf{k}} \rightarrow \frac{|\xi_{\mathbf{k}} + \bar{\xi}_f|}{2}$ . From Eq. (8) we obtain

$$1 = \lambda \int_{-D}^D \frac{d\xi}{2(\xi + \bar{\xi}_f)} \left[ \tanh\left(\frac{\beta_c \xi}{2}\right) + \tanh\left(\frac{\beta_c \bar{\xi}_f}{2}\right) \right], \quad (\text{B5})$$

where we have substituted  $\lambda = 2N|v|^2/\omega_e$ .  $\beta_c = 1/k_B T_c$ . The first integral in Eq. (B5) is a tricky one. In the limit of  $D \gg \bar{\xi}_f$ , we can approximately evaluate this integral. The first integral of Eq. (B5) gives

$$I_1 \approx \lambda \ln \left[ \frac{2D\gamma}{\sqrt{\bar{\xi}_f^2 + (2k_B T_c)^2}} \right], \quad (\text{B6})$$

where  $D\gamma = 2D\gamma/\pi$  with  $\gamma = 1.78$  being the Euler constant. The second integral is trivial to evaluate which gives

$$I_2 = \lambda \tanh\left(\frac{\beta_c \bar{\xi}_f}{2}\right) \ln \left| \frac{D + \bar{\xi}_f}{-D + \bar{\xi}_f} \right|. \quad (\text{B7})$$

In the limit of  $D > \bar{\xi}_f$ ,  $I_2 \rightarrow 0$ . Therefore, we are left with  $I_1 = 1$ , which gives,

$$(k_B T_c)^2 = D_\gamma^2 e^{-2/\lambda} - \frac{\bar{\xi}_f^2}{4}, \quad (\text{B8})$$

Eq. (8) in the main text is obtained from the above equation.

### 2. SC gap amplitude

Next we take the  $T \rightarrow 0$  limit in Eq. (8). In this limit, we get  $\tanh(\frac{\beta E_{\mathbf{k}}^\pm}{2}) \rightarrow \pm 1$ . Hence we are left with

$$\begin{aligned} 1 &= \lambda \int_{-D}^D \frac{d\xi}{\sqrt{(\xi + \bar{\xi}_f)^2 + 4\Delta^2}} \\ &= \lambda \ln \left( \frac{\sqrt{(D + \bar{\xi}_f)^2 + 4\Delta^2} + D + \bar{\xi}_f}{\sqrt{(D - \bar{\xi}_f)^2 + 4\Delta^2} - D + \bar{\xi}_f} \right) \\ &\approx \lambda \ln \left( \frac{2(D + \bar{\xi}_f)}{\sqrt{(D - \bar{\xi}_f)^2 + 4\Delta^2} - D + \bar{\xi}_f} \right) \end{aligned} \quad (\text{B9})$$



In the last equation above, we assumed  $D \gg \Delta$ . Solving Eq.(B9)

$$\Delta = \bar{D} e^{-\frac{1}{2\lambda}} \left[ 1 + r e^{-\frac{1}{\lambda}} \right]^{1/2}, \quad (\text{B10})$$

where  $\bar{D} = \sqrt{D^2 - \bar{\xi}_f^2}$ , and  $r = (D + \bar{\xi}_f)/(D - \bar{\xi}_f)$ . In the weak coupling limit  $\lambda \rightarrow 0$ , we get  $\Delta \rightarrow \bar{D} e^{-\frac{1}{2\lambda}}$  (notice the factor of  $2\lambda$  in the exponent) while in the strong coupling limit, we obtain the BCS-type formalism of  $\Delta \rightarrow \sqrt{D^2 + \bar{\xi}_f^2} e^{-\frac{1}{\lambda}} \approx D e^{-\frac{1}{\lambda}}$ .

### Appendix C: Pair susceptibility

To affirm that there exists a pairing instability in Eq. (3) in the main text, we compute the pair-pair correlation function. We consider the pair field

$$b_{\mathbf{k}}(\tau) = \sum_{\sigma, m} c_{\mathbf{k}\sigma}(\tau) \bar{f}_m(\tau), \quad (\text{C1})$$

where  $\tau$  is the imaginary time. The pair susceptibility is defined as

$$\chi_p(\mathbf{q}, i\omega_n) = \int_0^\beta \sum_{\mathbf{k}} \left\langle \mathcal{T}_\tau b_{\mathbf{k}}(\tau) b_{\mathbf{k}+\mathbf{q}}^\dagger(\tau') \right\rangle e^{-i\omega_n(\tau-\tau')} \quad (\text{C2})$$

Where  $\mathcal{T}_\tau$  is the time ordered operator. Using Wick's decomposition, we evaluate the above average as

$$\left\langle \mathcal{T}_\tau b_{\mathbf{k}}(\tau) b_{\mathbf{k}+\mathbf{q}}^\dagger(\tau') \right\rangle = \sum_{\sigma, m} \mathcal{G}_m^f(\tau - \tau') \mathcal{G}_{\mathbf{k}, \sigma}^c(\tau - \tau') \delta_{\mathbf{q}, 0}, \quad (\text{C3})$$

where  $\mathcal{G}_{\mathbf{k}, \sigma}^c(\tau - \tau') = \langle \mathcal{T}_\tau c_{\mathbf{k}\sigma}(\tau) c_{\mathbf{k}\sigma}^\dagger(\tau') \rangle$  is the conduction electron's Green's function, and  $\mathcal{G}_m^f(\tau - \tau') = \langle \mathcal{T}_\tau \bar{f}_m(\tau) f_m^\dagger(\tau') \rangle$  is the Green's function for the single site  $f_m$  states. In the fermionic Matsubara frequency  $ip_n$  space these two Green's functions become  $\mathcal{G}_{\mathbf{k}, \sigma}^c(ip_n) = (ip_n - \xi_{\mathbf{k}})^{-1}$ , and  $\mathcal{G}_m^f(ip_n) = (ip_n - \bar{\xi}_f)^{-1}$ . Substituting the Green's functions in Eq. (C2), and doing the Fourier transformation we get

$$\chi_p(i\omega_n) = \frac{1}{\beta} \sum_{\mathbf{k}, \sigma, m} \sum_{n'} \mathcal{G}_m^f(ip_{n'}) \mathcal{G}_{\mathbf{k}, \sigma}^c(i\omega_n - ip_{n'}). \quad (\text{C4})$$

Substituting the corresponding Green's functions and performing the standard Matsubara frequency summation on  $ip_{n'}$ , we arrive at

$$\chi_p(i\omega_n) = \sum_{\mathbf{k}} \frac{1 - f(\bar{\xi}_f) - f(\xi_{\mathbf{k}})}{\bar{\xi}_f + \xi_{\mathbf{k}} - i\omega_n}, \quad (\text{C5})$$

$f(\xi)$  is the Fermi distribution function. We are interested in the  $\omega \rightarrow 0$ , and  $\mathbf{q} \rightarrow 0$  limits. Taking analytic continuation to

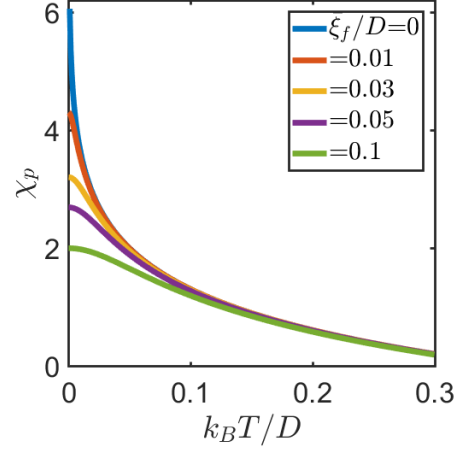


FIG. 5. Static pair susceptibility at  $\mathbf{q} = 0$  as a function of temperature for different values of  $\bar{\xi}_f$ . As expected from Eq. (C7) the pair correlation function diverges at  $T \rightarrow 0$  for  $\bar{\xi}_f \rightarrow 0$ .

the real frequency plane  $i\omega_n \rightarrow \omega + i\delta$ , the pair susceptibility becomes

$$\chi_p(\omega \approx 0) = \frac{N}{2} \int_{-D}^D d\xi \frac{\tanh(\frac{\beta \bar{\xi}_f}{2}) + \tanh(\frac{\beta \xi}{2})}{\bar{\xi}_f + \xi}. \quad (\text{C6})$$

This equation is nothing but the R.H.S. of Eq. (B5), except the constant factor  $V$ . Again in the limit of  $D \gg \bar{\xi}_f$  this integral gives the solution as in Eq. (B6). Hence we get

$$\chi_p(T) = N \ln \left[ \frac{2D\gamma}{\sqrt{\bar{\xi}_f^2 + (2k_B T)^2}} \right]. \quad (\text{C7})$$

Interestingly, unlike the typical BCS case, the pair correlation function does not have a logarithmic divergence as  $T \rightarrow 0$  except in the limit of  $\bar{\xi}_f \rightarrow 0$ . This is the reason superconductivity is limited by a minimum limit of the coupling constant  $\lambda$  and  $T_K$  to overcome the onsite energy  $\bar{\xi}_f$  as discussed in the main text.

### Appendix D: Further details of the Meissner effect

Unlike the typical Cooper pair of two conduction electrons with opposite momenta in other mechanism, here we have a pairing between conduction electron and correlated singly occupied  $f$ -electrons. How do these Cooper pairs couple to the applied magnetic field? It is easy to envisage that conduction electrons directly couple to the gauge field  $\mathbf{A}$  as  $\mathbf{p}' = \hbar\mathbf{k} - \frac{e}{c}\mathbf{A}$ . On the other hand, the  $f$ -states do not couple to the vector potential in its localized limit. Therefore, important changes are expected here, in the Meissner effects, compared to typical BCS case.

First of all, under the magnetic field the BdG states become chiral and thus the Bogolyubov states  $\phi_{\pm\mathbf{k}}^\pm$  and the corresponding eigenvalues  $E_{\pm\mathbf{k}}^\pm$  for  $\pm\mathbf{k}$  are no longer the same.

Hence we treat them explicitly as:

$$\begin{aligned} c_{\mathbf{k}\sigma} &= \alpha_{\mathbf{k}}\phi_{\mathbf{k}}^+ + \beta_{\mathbf{k}}(\phi_{\mathbf{k}}^-)^\dagger \\ c_{-\mathbf{k}\sigma} &= \alpha_{\mathbf{k}}\phi_{-\mathbf{k}}^+ + \beta_{\mathbf{k}}(\phi_{-\mathbf{k}}^-)^\dagger. \end{aligned} \quad (\text{D1})$$

$\alpha_{\mathbf{k}}$ , and  $\beta_{\mathbf{k}}$  are the coherence factors at zero magnetic field. The corresponding change in the eigenvalue are  $E_{\pm\mathbf{k}}^\nu = E_{\mathbf{k}}^\nu \mp \frac{e}{c}\mathbf{a}\cdot\mathbf{v}_{\mathbf{k}}$ , where  $\nu = \pm$ , and  $\mathbf{a}$  is the Fourier component of the vector potential in the momentum space.  $\mathbf{v}_{\mathbf{k}} = \partial\xi_{\mathbf{k}}/(\hbar\partial\mathbf{k})$  is the conduction band velocity with  $\mathbf{v}_{-\mathbf{k}} = -\mathbf{v}_{\mathbf{k}}$ .  $E_{\mathbf{k}}^\nu$  are the eigenvalues without the magnetic field, and hence  $E_{-\mathbf{k}}^\nu = E_{\mathbf{k}}^\nu$ . In the weak magnetic field limit, this corresponds to the change in the Fermi Dirac distribution functions as  $f(E_{\pm\mathbf{k}}^\nu) = f(E_{\mathbf{k}}^\nu) \mp (\frac{e}{c}\mathbf{a}\cdot\mathbf{v}_{\mathbf{k}})\frac{\partial f}{\partial E_{\mathbf{k}}^\nu}$ . The two current operators are

$$\mathbf{J}_d(\mathbf{q}) = \frac{e^2}{c}\mathbf{a}(\mathbf{q})\sum_{\mathbf{k}\sigma}'\frac{1}{m_{\mathbf{k}}}\left[c_{\mathbf{k}-\mathbf{q}\sigma}^\dagger c_{\mathbf{k}\sigma} + c_{-\mathbf{k}+\mathbf{q}\sigma}^\dagger c_{-\mathbf{k}\sigma}\right] \quad (\text{D2})$$

$$\mathbf{J}_p(\mathbf{q}) = e\sum_{\mathbf{k}\sigma}'\mathbf{v}_{\mathbf{k}-\mathbf{q}}\left[c_{\mathbf{k}-\mathbf{q}\sigma}^\dagger c_{\mathbf{k}\sigma} - c_{-\mathbf{k}+\mathbf{q}\sigma}^\dagger c_{-\mathbf{k}\sigma}\right]. \quad (\text{D3})$$

Here  $m_{\mathbf{k}}$  is the effective mass of the conduction electron. In the above two equations we utilized the fact that  $\mathbf{v}_{-\mathbf{k}} = -\mathbf{v}_{\mathbf{k}}$ , and  $m_{-\mathbf{k}} = m_{\mathbf{k}}$ . The prime over the summation indicate

that the summation is restricted to the first quadrant of the Brillouin zone. By substituting Eq. (D1) and after a lengthy and straightforward calculation, we arrive at

$$\begin{aligned} \mathbf{J}_d(0) &= -\frac{e^2\mathbf{a}(0)}{c}\sum_{\mathbf{k}}'\frac{1}{m_{\mathbf{k}}} \\ &\times \left[1 - (\alpha_{\mathbf{k}}^+)^2 \tanh\left(\frac{\beta E_{\mathbf{k}}^+}{2}\right) - (\alpha_{\mathbf{k}}^-) \tanh\left(\frac{\beta E_{\mathbf{k}}^-}{2}\right)\right], \end{aligned} \quad (\text{D4})$$

$$\begin{aligned} \mathbf{J}_p(0) &= \frac{e^2\beta}{2c}\sum_{\mathbf{k}}'(\mathbf{a}\cdot\mathbf{v}_{\mathbf{k}})\mathbf{v}_{\mathbf{k}} \\ &\times \left[(\alpha_{\mathbf{k}}^+) \operatorname{sech}^2\left(\frac{\beta E_{\mathbf{k}}^+}{2}\right) + (\alpha_{\mathbf{k}}^-) \operatorname{sech}^2\left(\frac{\beta E_{\mathbf{k}}^-}{2}\right)\right]. \end{aligned} \quad (\text{D5})$$

Next we take the linear response theory and within the London's equations, we define the penetration depth  $\lambda(T)$  as  $\lambda_{ij}^{-2} = -\frac{4\pi}{c}\frac{J_i(0)}{a_j(0)}$ , where  $\mathbf{J} = \mathbf{J}_p + \mathbf{J}_d$  is the total current.  $i, j$  are the spatial coordinates. This gives the final result given in Eq. (15). This equation reduces to the typical BCS form in the case of  $\xi_{\mathbf{k}} = -\xi_f$ .

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