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Rev. Mod. Phys. **95**, 031002 — Published 15 September 2023

DOI: 10.1103/RevModPhys.95.031002

Unconventional fully-gapped superconductivity in the heavy-fermion metal $CeCu_2Si_2$

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(Dated: March 7, 2023)

The heavy-fermion metal $CeCu_2Si_2$ was the first discovered unconventional, non-phonon-mediated superconductor, and for a long time was believed to exhibit single-band d-wave superconductivity, as inferred from various measurements hinting at a nodal gap structure. More recently however, measurements using a range of techniques at very low temperatures ($T \lessapprox 0.1$ K) provided evidence for a fully-gapped superconducting order parameter. In this Colloquium, after a brief historical overview we survey the apparently conflicting results of numerous experimental studies on this compound. We then address the different theoretical scenarios which have been applied to understand the particular gap structure, including both isotropic (sign-preserving) and anisotropic two-band s-wave superconductivity, as well as an effective two-band d-wave model, where the latter can explain the currently available experimental data on $CeCu_2Si_2$. The lessons from $CeCu_2Si_2$ are expected to help uncover the Cooper-pair states in other unconventional, fully-gapped superconductors with strongly correlated carriers, and in particular highlight the rich variety of such states enabled by orbital degrees of freedom.

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I. INTRODUCTION

Strongly correlated electron systems are central to contemporary studies of quantum materials. In these materials, electron-electron interactions have a strength that reaches or even exceeds the width of the underlying non-interacting electron bands. This property is to be con-

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trasted with conventional metals such as aluminum or ordinary semiconductors like silicon, where electronic properties can be successfully described in terms of noninteracting electrons with a materials-specific bandstructure. Instead, for strongly correlated electron systems, the interactions lead to rich emergent phenomena and novel electronic phases of matter. Examples of strongly correlated electron systems include cuprate perovskites (Lee *et al.*, 2006; Proust and Taillefer, 2019), iron-based pnictides and chalcogenides (Si *et al.*, 2016; Stewart, 2011), organic charge-transfer salts (Kanoda, 2008; Lang and Müller, 2003; Maple *et al.*, 2004), and the moiré structures of graphene and transition-metal dichalcogenides (Andrei and MacDonald, 2020; Cao *et al.*, 2018).

Among the strongly correlated electron systems, heavy fermion compounds such as $CeCu_2Si_2$ take a special place. The reason is simple. These materials contain partially-filled f-orbitals. For these f-electrons, the interactions are larger than their bandwidth to such an extent that the f-electrons act as localized magnetic moments. Indeed, at the heart of the physics of heavy fermion materials is the Kondo effect, whereby localized magnetic moments situated in a sea of conduction electrons become screened, and below a characteristic temperature scale (the Kondo temperature T_K), the local moments are entirely quenched leaving behind a remanent nonmagnetic Kondo singlet (Hewson, 1997). Such screened moments act as strong elastic scatterers, accounting for the peculiar logarithmic increase of the resistivity upon cooling when small concentrations of certain magnetic impurities are introduced into nonmagnetic metals (Kondo, 1964). As was detected by Triplett and Phillips, 1971 for the dilute magnetic alloys **Cu**Cr and **Cu**Fe, the impurity-derived "incremental" low-temperature specific heat is proportional to temperature, $\Delta C(T) = \gamma T$, with a huge coefficient γ, that exceeds the Sommerfeld coefficient of the host metal Cu by more than a factor of 1000. This indicates the formation of a narrow local Kondo resonance at the Fermi level E_F and could be well described in the framework of a local Fermi liquid theory (Noziéres, 1974).

Heavy fermion metals comprise of two broad classes, lanthanides and actinides. The lanthanide-based variants are commonly considered to be ideal examples of Kondo lattice systems. These materials rather than having a dilute random distribution of local moments instead host a dense, periodic lattice of Kondo ions (Aliev *et al.*, 1983a; Brandt and Moshchalkov, 1984; Fulde *et al.*, 1988; Kuramoto and Kitaoka, 2012; Ott, 1987; Stewart, 1984). The first observation of heavy-fermion phenomena, i.e., the properties of a heavy Fermi liquid, was reported for the hexagonal paramagnetic compound $CeAl_3$ (Andres *et al.*, 1975). Here, the low-temperature specific heat, which is practically identical to the $4f$-electron contribution, was found to be proportional to temperature with a γ coefficient of the same gigantic size as the aforementioned value for **Cu**Fe. In addition, the low-temperature resistivity of $CeAl_3$ was observed to follow a $\Delta\rho(T) = AT^2$ dependence with a huge prefactor A. These early findings were ascribed to a $4f$ virtual bound state at E_F. A very similar large γ coefficient of the low-T specific heat to that of $CeAl_3$ could be estimated for the putative paramagnetic phase of the cubic antiferromagnet $CeAl_2$ (with a similar T_K) by treating the Ce ions as isolated Kondo centers (Schotte and Schotte, 1975). This was taken as striking evidence for the heavy fermion phenomena in these Ce compounds indeed being due to the many-body Kondo effect rather than one-particle physics (Bredl *et al.*, 1978).

The participation of the f-electrons in the electronic structure at sufficiently low temperatures causes the renormalized electronic bands to take on significant 'f-electron' character and the effective mass of the charge carriers exceeds that of ordinary conduction electrons by a factor up to about a thousand (Zwicknagl, 1992). This leads to the aforementioned unusual behaviors of canonical heavy-fermion compounds such as $CeCu_2Si_2$, namely the γ coefficient is of the order of J/K^2mol (Fig. 1(a)), and there is a correspondingly enhanced temperature-independent Pauli spin susceptibility (Grewe and Steglich, 1991; Sales and Viswanathan, 1976) (Fig. 2). As displayed in Fig. 1(b), the electrical resistivity first exhibits an increase upon cooling from high temperatures, reflecting increasing incoherent scattering similar to that which occurs from dilute magnetic impurities. At lower temperatures however, Kondo lattice effects set in whereby coherent scattering of conduction electrons from the Kondo singlets below a characteristic temperature ($T_K \approx 15$ K for $CeCu_2Si_2$ (Stockert *et al.*, 2011)) leads to a pronounced decrease of the resistivity (Coleman, 2007). In several heavy-fermion metals, this decline of the resistivity follows a Fermi-liquid-type AT^2 dependence with a huge A coefficient, whereas $CeCu_2Si_2$ exhibits non-Fermi-liquid behavior, as discussed in the following sections.

Another stark difference between Kondo lattices and the dilute impurity case is that in the former the Kondo effect competes with the indirect Ruderman, Kittel, Kasuya and Yoshida (RKKY) magnetic exchange interaction (Kasuya, 1956; Ruderman and Kittel, 1954; Yosida, 1957) which tends to stabilize the f-electron moments. While predominant Kondo screening results in a paramagnetic heavy-fermion ground state, a dominant RKKY interaction causes magnetic, most frequently antiferromagnetic order. For quite a substantial number of these heavy-fermion metals the Kondo screening turns out to almost exactly cancel the RKKY interaction in the zero-temperature limit, which may give rise to a continuous zero-temperature quantum phase transition or quantum critical point (QCP), that can be easily accessed by adjusting a suitable non-thermal control parameter, e.g., pressure, doping or magnetic fields (Gegenwart *et al.*, 2008; Sachdev, 2011; Si and Steglich, 2010; Stewart, 2001). To get rid of the large residual entropy accumulated at the QCP, symmetry-broken novel phases are often observed, notably 'unconventional' superconductivity which cannot be accounted for by the electron-phonon mediated pairing mechanism of Bardeen, Cooper, Schrieffer theory (Norman, 2013, 2011; Stewart, 2017).

The heavy fermion metal $CeCu_2Si_2$ was also the first unconventional superconductor to be discovered (Steglich *et al.*, 1979) (Table I), and it has recently attracted much research interest again. While it was considered a (single-band) d-wave superconductor for many years (Fujiwara *et al.*, 2008; Ishida *et al.*, 1999), the observation of a fully developed energy gap at very low temperatures (Kittaka *et al.*, 2014; Pang *et al.*, 2018; Takenaka *et al.*, 2017; Yamashita *et al.*, 2017) has led to proposals of $CeCu_2Si_2$ being a two-band s-wave superconductor both with (Ikeda *et al.*, 2015; Li *et al.*, 2018), and without (Takenaka *et al.*, 2017; Tazai and Kontani, 2018, 2019; Yamashita *et al.*, 2017) a sign-change of the order parameter.

In this Colloquium article, after a brief historical overview we discuss the seemingly conflicting results of a large number of experimental studies on this material and address the different theoretical models applied to understand the particular gap structure. These models are divided into two categories. One class builds on a normal state in the presence of Kondo-driven renormalization, and utilizes the multiplicity of orbitals to realize a new kind of pairing state. In the band basis, this takes the form of a band-mixing $d+d$-pairing state (Nica and Si, 2021), in parallel with the proposed pairing state for the iron chalcogenides that are among the highest-T_c Fe-based superconductors (Nica *et al.*, 2017) based on strongly orbital-selective electron correlations. The other class directly works in the band basis, treats the Coulomb repulsive interaction perturbatively, and constructs a pairing state using the standard procedure of finding irreducible representations of the crystalline lattice's point group. This is exemplified by the s_{+-} scenario (Ikeda *et al.*, 2015; Li *et al.*, 2018), by analogy to a similar construction applied to the Fe-based superconductors (Mazin *et al.*, 2008) in which a repulsive interband interaction leads to different signs of the order parameter between hole and electron pockets. We summarize the details of these considerations throughout the body of the article. In addition, we suggest that the insights gained from the analysis of the pairing state in $CeCu_2Si_2$ will have broad implications on strongly correlated superconductivity in multi-orbital systems, and discuss future efforts that may shed further light on this canonical problem in the field of strongly correlated electron systems.

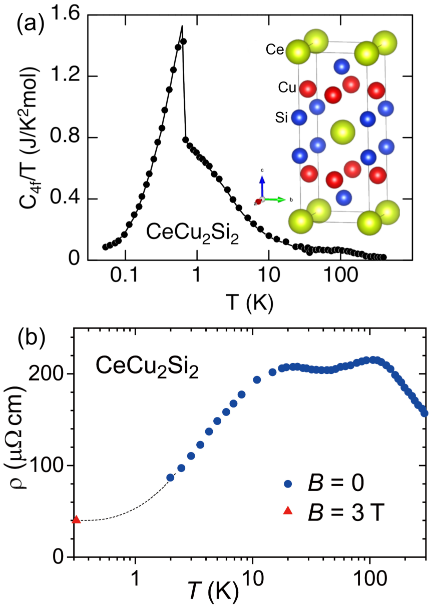

FIG. 1 (a) Contribution of the $4f$-electrons to the specific heat in $CeCu_2Si_2$, plotted as C_{4f}/T vs. T on a logarithmic scale. Replotted from (Steglich, 1990). The solid line is a guide to the eyes. The inset shows the crystal structure of $CeCu_2Si_2$ ($ThCr_2Si_2$-type structure, space group $I4/mmm$), where green, red, and blue spheres correspond to Ce, Cu and Si atoms (see labels), respectively. (b) Temperature dependence of the resistivity of $CeCu_2Si_2$ on a logarithmic temperature scale, reproduced from Shan *et al.*, 2022.

II. THE HISTORY OF HEAVY-FERMION SUPERCONDUCTIVITY

Given the strong pair-breaking effect of diluted localized spins in conventional superconductors (Abrikosov and Gor'kov, 1960; Matthias *et al.*, 1958), the discovery of bulk superconductivity in $CeCu_2Si_2$ (Steglich *et al.*, 1979) came as a big surprise. In a BCS superconductor, a tiny amount of randomly distributed magnetic impurities fully suppresses the superconducting state (Maple, 1968; Maple *et al.*, 1972; Riblet and Winzer, 1971; Steglich and Armbrüster, 1974), but the superconductivity is robust against doping with nonmagnetic impurities (Anderson, 1959; Balatsky *et al.*, 2006). On the other hand, superconductivity in $CeCu_2Si_2$ relies on a periodic array of 100 at% magnetic Ce^{3+} ions, each containing a localized $4f$-shell occupied by one electron in a $J = 5/2$ Hund's rule ground state. Figure 2 displays the first reported evidence for the superconducting transition in $CeCu_2Si_2$ at $T_c \approx 0.5$ K on annealed polycrystalline samples. Upon cooling through T_c, the electrical resistivity falls to zero from a normal state with a non-saturated, nearly linear temperature dependence, while the ac susceptibility undergoes a rapid change from a strongly enhanced Pauli paramagnetic susceptibility to a large diamagnetic value [Fig. 2(a)].

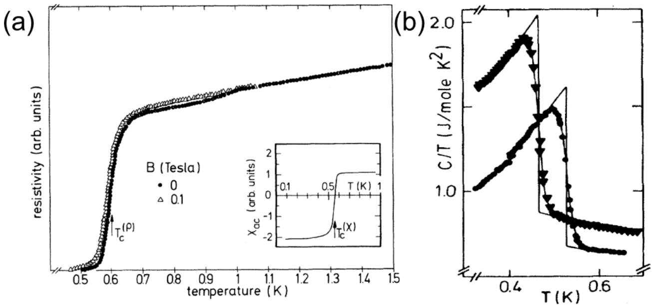

FIG. 2 (a) Resistivity $\rho(T)$, ac-susceptibility $\chi_{\mathrm{ac}}(T)$ and (b) specific heat as C/T vs T for polycrystalline $\mathrm{CeCu_2Si_2}$ indicating bulk superconductivity at $T_{\mathrm{c}} \approx 0.5$ K, after (Steglich *et al.*, 1979). The Pauli susceptibility ($T > T_{\mathrm{c}}$) shown in the inset amounts to $\chi_P = 82 \times 10^{-9}\mathrm{m^3/mol}$ (Aarts, 1984). Note that the normal-state values of both $\rho(T)$ and $C(T)/T$ point to non-Fermi-liquid behavior. In (b) results from two different samples, with the same nominal composition and prepared in the same way, are shown. Reproduced with permission from Steglich *et al.*, 1979.

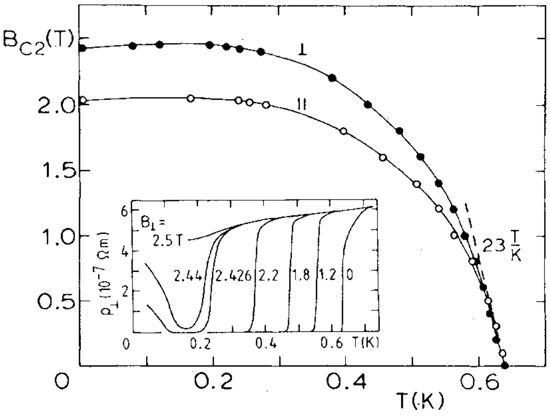

FIG. 3 Upper critical magnetic field B_{c2} vs T of a $\mathrm{CeCu_2Si_2}$ single crystal for fields applied within (∥) and perpendicular to (⊥) the Ce planes as obtained from $\rho(T)$, measured parallel to the respective field. Only a moderate anisotropy, but a giant initial slope at T_{c} is found for B_{c2}(T). Note the shallow maximum of B_{c2}(T) near $T = 0.15$ K as reflected in the inset by the reentrant $\rho(T)$ behavior for $B \geq 2.4$ T. Reproduced with permission from Assmus *et al.*, 1984.

Two early observations have led to the conclusion that $\mathrm{CeCu_2Si_2}$ must be an unconventional bulk superconductor: (i) the nonmagnetic reference compound $\mathrm{LaCu_2Si_2}$ is not a superconductor, at least down to 20 mK (Steglich *et al.*, 1979), and (ii) a tiny amount of nonmagnetic (as well as magnetic) substitution at the level of 1 at % may lead to a complete suppression of superconductivity in $\mathrm{CeCu_2Si_2}$ (Spille *et al.*, 1983), see Sec. III.E. Further evidence for this conclusion could be drawn from the specific-heat results shown in Fig. 2(b). Here the normal-state values of $C(T)/T$ are of the order of several hundreds of mJ/mol K^2; they substantially increase upon lowering the temperature and extrapolate to about 1 J/mol K^2 in the zero-temperature limit. This exceeds the Sommerfeld coefficient of the electronic specific heat of Cu by more than a factor of 1000 and this proves that similar to $\mathrm{CeAl_3}$, the measured specific heat in this low-temperature range is practically identical with the electronic contribution ($\approx C_{4f}$). The corresponding renormalized kinetic energy, $k_{\mathrm{B}}T_{\mathrm{F}}^*$, corresponds to the Kondo screening energy, $k_{\mathrm{B}}T_{\mathrm{K}}$, $T_{\mathrm{K}} \approx 15$ K (Stockert *et al.*, 2011). Therefore, the ratio $T_{\mathrm{c}}/T_{\mathrm{F}}^*$ is of the order of 0.04, compared to $T_{\mathrm{c}}/T_{\mathrm{F}} \approx 10^{-3} - 10^{-4}$ for an ordinary BCS superconductor, highlighting $\mathrm{CeCu_2Si_2}$ as a 'high-T_{c} superconductor' in a normalized sense (Steglich *et al.*, 1979). On the other hand, the ratio $T_{\mathrm{F}}^*/\theta_{\mathrm{D}}$, where θ_{D} is the Debye temperature, also amounts to about 0.05, while in a main group metal or transition metal, $T_{\mathrm{F}}/\theta_{\mathrm{D}}$ is of order 100. The latter warrants the electron-phonon coupling in conventional BCS superconductors to be retarded, such that the Coulomb repulsion between conduction electrons is minimized and isotropic s-wave Cooper pairs may be formed.

For heavy-fermion metals, such phonon-mediated on-site pairing is clearly prohibited because of their low renormalized Fermi velocity which is, at best, of the order of the velocity of sound. Nevertheless, an early proposal was put forward to explain heavy-fermion superconductivity in $\mathrm{CeCu_2Si_2}$ by a coupling of the heavy charge carriers to the breathing mode (Razafimandimby *et al.*, 1984), while recently such a phonon-mediated superconductivity for this compound was expected to be realized near a magnetic instability, thanks to the vertex corrections due to multipole charge fluctuations (Tazai and Kontani, 2018). On the other hand, a broad consensus evolved shortly after the discovery of heavy-fermion superconductivity that here an electronic pairing mechanism must be operating (Machida, 1983; Tachiki and Maekawa, 1984). Therefore, $\mathrm{CeCu_2Si_2}$ was soon regarded generally as an unconventional, i.e., non-phonon-driven, superconductor. Because of the phenomenological similarity of heavy-fermion superconductivity in $\mathrm{CeCu_2Si_2}$ with the superfluidity in ^{3}He (Osheroff *et al.*, 1972a,b), a magnetic coupling mechanism appeared to be most natural (Anderson, 1984).

The jump height at the superconducting transition, $\Delta C/T_{\mathrm{c}}$, is comparable to the Sommerfeld coefficient, extrapolated to $T = 0$, $\gamma_0 = C(T \to 0)/T \approx 1$ J/mol K^2 (Fig. 2(b)). This not only proved bulk superconductivity, but also led to the conclusion that the Cooper pairs are formed by heavy-mass quasiparticles (Steglich *et al.*, 1979) and to the term 'heavy-fermion superconductivity' (Rauchschwalbe *et al.*, 1982). In fact, if the superconductivity were solely carried by the coexisting light conduction electrons, the jump in the electronic specific-heat coefficient at T_{c} would have been so tiny that within the scatter of the data it would not be resolvable in Fig. 2(b). It should be noted that recent theoretical considerations have shown that in order to form Cooper pairs in $CeCu_2Si_2$, a very large kinetic energy cost, exceeding the binding energy by a factor as high as 20, is necessary to overcompensate the similarly large exchange energy between the paired heavy quasiparticles (Stockert *et al.*, 2011). The large kinetic-energy cost has been interpreted in terms of a transfer of single-electron spectral weight to energies above a Kondo-destruction energy scale at the QCP, T^*, that is nonzero but small compared to the bare Kondo scale (Stockert *et al.*, 2011), see Sec. III.C.

The two polycrystalline samples exploited in Fig. 2(b) were prepared and annealed in the same way. Nevertheless, their specific-heat values were found to be significantly different. These variations of physical properties from one sample to the other added to the severe skepticism (Hull *et al.*, 1981; Schneider *et al.*, 1983) which existed throughout the first few years after the report of bulk superconductivity in $CeCu_2Si_2$ (Steglich *et al.*, 1979), subsequently confirmed by other groups (Aliev *et al.*, 1983b, 1984; Ishikawa *et al.*, 1983; Ōnuki *et al.*, 1984). The cause for these 'sample dependences', see also (Aliev *et al.*, 1983b; Stewart *et al.*, 1983), was resolved only many years later by a thorough study of the chemical phase diagram (Müller-Reisener, 1995; Steglich *et al.*, 2001), and the observation of a quantum critical point (QCP) that is located inside the narrow homogeneity range (Lengyel *et al.*, 2011; Steglich *et al.*, 2001), see Sec. III.C. The above-mentioned skepticism could be overcome after a few years, when high-quality single crystals of $CeCu_2Si_2$ were prepared (Assmus *et al.*, 1984; Ōnuki *et al.*, 1984) and found to show even more pronounced superconducting phase transition anomalies compared to polycrystals. The upper critical field curve $B_{c2}(T)$ of such a single crystal is displayed in Fig. 3. It reveals:

(i) Only a small anisotropy for the field being applied either parallel or perpendicular to the basal tetragonal plane [inset of Fig. 1], contrasting a pronounced anisotropy in the electrical resistivity (Schneider *et al.*, 1983).

(ii) A shallow maximum around $T = 0.15$ K (inset) which seems to correspond to a low-temperature hump in $C(T)/T$ (Bredl *et al.*, 1984), also observed for $CeAl_3$ (Bredl *et al.*, 1984; Flouquet *et al.*, 1982; Steglich *et al.*, 1985b), which was ascribed to the opening of a partial coherence gap in the 4f-quasiparticle density of states at the Fermi level, see Table I. Later this hump was ascribed as being related to antiferromagnetic correlations (Steglich *et al.*, 1996; Stockert *et al.*, 2004). For UBe_{13} too, a broad peak in $C(T)/T$ at $T_L \approx 0.6$ K had been detected (Rauchschwalbe *et al.*, 1987a,b) and subsequently identified (Kromer *et al.*, 1998, 2000) as the precursor of an anomaly indicating a continuous phase transition at T_{c2} below the superconducting T_{c1} discovered for $(U_{1-x}Th_x)Be_{13}$ in the critical concentration range $0.019 \leq x \leq 0.045$ (Ott *et al.*, 1985). The nature of this lower-lying phase transition has yet to be resolved (Steglich and Wirth, 2016). While ultrasound-attenuation results (Batlogg *et al.*, 1985) hint at a SDW transition, pressure studies (Lambert *et al.*, 1986) and results of the lower critical field (Rauchschwalbe *et al.*, 1987a) highlight a superconducting nature of the transition at T_{c2}.

(iii) A giant initial slope at T_c which supports the massive nature of the Cooper pairs as inferred from the giant jump anomaly $\Delta C/T_c$.

(iv) Quite a strong Pauli limiting effect in the low-temperature regime for both field configurations. This discards odd-parity (spin-triplet) pairing as observed in superfluid ^{3}He (Leggett, 1975) and originally assumed for heavy-fermion superconductors (Anderson, 1984). A spatially modulated superconducting state in $CeCu_2Si_2$ at very low temperature close to the upper critical field was recently proposed based on Cu-NMR results (Kitagawa *et al.*, 2018).

A dc Josephson effect with a critical pair current of ordinary size was observed on a weak link between polycrystalline $CeCu_2Si_2$ and Al (Steglich *et al.*, 1985b). This as well as Knight shift results from ^{29}Si NMR (Ueda *et al.*, 1987) lent further support to even-parity (spin-singlet) pairing in $CeCu_2Si_2$.

At around the same time, theorists proposed d-wave superconductivity mediated by antiferromagnetic spin fluctuations (Miyake *et al.*, 1986; Scalapino *et al.*, 1986). These theoretical studies extend the theory of ferromagnetic paramagnons developed in the ^{3}He context to the antiferromagnetic case, but the Kondo effect responsible for the heavy mass was not addressed. In more recent years, the Kondo effect has been incorporated into the study of heavy-fermion quantum criticality (Gegenwart *et al.*, 2008), with an emphasis on the notion of Kondo destruction (Coleman *et al.*, 2001; Si *et al.*, 2001). A corresponding theory for quantum-criticality-driven superconductivity in Kondo-lattice models has been advanced (Hu *et al.*, 2021a).

The discovery of heavy-fermion superconductivity in the cubic compound UBe_{13} (Ott *et al.*, 1983) proved this phenomenon to be general and not restricted to a single material. Thereafter, UPt_3 (Stewart *et al.*, 1984), URu_2Si_2 (Maple *et al.*, 1986; Palstra *et al.*, 1985; Schlabitz *et al.*, 1984, 1986), U_2PtC_2 (Meisner *et al.*, 1984), UNi_2Al_3 (Geibel *et al.*, 1991b), and UPd_2Al_3 (Geibel *et al.*, 1991a) were found to be heavy-fermion superconductors, too. They were followed by the pressure-induced Ce-based heavy-fermion superconductors $CeCu_2Ge_2$ (Jaccard *et al.*, 1992), $CeRh_2Si_2$ (Movshovich *et al.*, 1996), $CeIn_3$ and $CePd_2Si_2$ (Mathur *et al.*, 1998). In the years after 2000, many of the Ce-based tetragonal, so-called 115 materials, which are obtained by increasing the c/a ratio of cubic $CeIn_3$ by inserting an additional layer of $T\mathrm{In}_2$ (T: Co, Rh or Ir), as well as the related 218 and 127 compounds were shown to be heavy-fermion superconductors (Sarrao and Thompson, 2007; Thompson and Fisk, 2012). One of the Pu-based isostructural compounds, PuCoGa_5, exhibits the record $T_\mathrm{c} = 18.5$ K for this class of superconductors (Sarrao *et al.*, 2002). Its Rh homologue PuRhGa_5 (Wastin *et al.*, 2003) as well as $\mathrm{NpPd}_5\mathrm{Al}_2$ (Aoki *et al.*, 2009) also show enhanced T_c values of 8.7 K and 4.9 K, respectively. The discovery of heavy-fermion superconductivity in the noncentrosymmetric compound $\mathrm{CePt}_3\mathrm{Si}$ (Bauer *et al.*, 2004) stimulated the search for noncentrosymmetric heavy-fermion as well as weakly-correlated superconductors (Bauer *et al.*, 2012; Smidman *et al.*, 2017), and resulted in several Ce-based counterparts. Such a lack of inversion symmetry allows for a mixing between even- and odd-parity pairing states (Gor'kov and Rashba, 2001). In the case of $\mathrm{CeRh}_2\mathrm{As}_2$ which has a locally noncentrosymmmetric crystal structure, two-phase superconductivity has been reported very recently, along with a proposal for a field-induced transition between an even parity phase at low fields, and an odd parity phase at elevated fields (Khim *et al.*, 2021). Two different superconducting phases in the presence of weak antiferromagnetic order have already been established earlier for UPt_3 (Joynt and Taillefer, 2002), and multifaceted behavior has been reported for thoriated UBe_{13} (Heffner *et al.*, 1990; Oeschler *et al.*, 2003; Ott *et al.*, 1985) as well as $\mathrm{URu}_2\mathrm{Si}_2$, exhibiting a hidden-order phase (Mydosh *et al.*, 2020). All of the three latter materials show a superconducting state with broken time-reversal symmetry (Heffner *et al.*, 1990; Luke *et al.*, 1993; Schemm *et al.*, 2015, 2014).

There are only two Yb-based heavy-fermion superconductors known so far. β-YbAlB_4 with $T_\mathrm{c} = 80$ mK (Nakatsuji *et al.*, 2008) is an intermediate-valence compound showing quantum criticality without tuning (Matsumoto *et al.*, 2011). $\mathrm{YbRh}_2\mathrm{Si}_2$ (Nguyen *et al.*, 2021; Schuberth *et al.*, 2016, 2022; Shan *et al.*, 2022) exhibits an antiferromagnetic QCP at $B \approx 0$ which is induced by nuclear spin order (below $T_\mathrm{A} = 2.3$ mK). The latter strongly competes with the primary $4f$-electronic order ($T_\mathrm{N} = 70$ mK) and causes the emergence of heavy-fermion superconductivity at ultra-low temperatures, $T_\mathrm{c} = 2$ mK. As shown by (Schuberth *et al.*, 2022), measurements of the Meissner effect point to the existence of bulk superconductivity up to magnetic fields of the order of $B = 40$ mT (about two-thirds of B_N, the critical field designating the Kondo-destruction QCP). Furthermore recent resistivity investigations suggest that at such elevated fields superconductivity may be of the spin-triplet variety (Nguyen *et al.*, 2021), which is theoretically supported based on unconventional superconductivity driven by Kondo destruction at magnetic-field-induced quantum criticality in the presence of an effective Ising spin anisotropy (Hu *et al.*, 2021b). Correlated Pr-based superconductors were also found. $\mathrm{PrOs}_4\mathrm{Sb}_{12}$ shows a heavy-fermion normal-state and superconducting properties due to dominant quadrupolar rather than dipolar fluctuations (Maple *et al.*, 2002; Rotundu *et al.*, 2004), while $\mathrm{PrTi}_2\mathrm{Al}_{20}$, $\mathrm{PrV}_2\mathrm{Al}_{20}$ and $\mathrm{PrIr}_2\mathrm{Zn}_{20}$ are quadrupolar Kondo-lattice systems exhibiting superconductivity and quadrupolar order (Onimaru *et al.*, 2011; Sakai *et al.*, 2012; Tsujimoto *et al.*, 2014).

A few heavy fermion superconductors are prime candidates for odd-parity pairing, i.e., the ferromagnetic compounds UGe_2 (Saxena *et al.*, 2000), UCoGe (Lévy *et al.*, 2005) and URhGe (Huy *et al.*, 2007) as well as UPt_3 (Tou *et al.*, 1998) and $\mathrm{UNi}_2\mathrm{Al}_3$ (Ishida *et al.*, 2002). Also being discussed is UTe_2 (Aoki *et al.*, 2019; Ran *et al.*, 2019), which has been suggested to be a chiral topological superconductor (Jiao *et al.*, 2020), for which the role of Kondo and RKKY interactions in the magnetic correlations and superconductivity has been discussed (Chen *et al.*, 2021; Duan *et al.*, 2021, 2020; Knafo *et al.*, 2021; Thomas *et al.*, 2020).

In concluding this survey, we can state that currently about fifty heavy-fermion superconductors are known. Most of these materials are discussed in (Pfleiderer, 2009). They are complemented by the already mentioned compounds β-YbAlB_4, $\mathrm{Pr(Ti,V)}_2\mathrm{Al}_{20}$, $\mathrm{PrIr}_2\mathrm{Zn}_{20}$ $\mathrm{YbRh}_2\mathrm{Si}_2$, UTe_2, and $\mathrm{CeRh}_2\mathrm{As}_2$. The majority of heavy-fermion superconductors are believed to have anisotropic even-parity Cooper pairing. In the following section, we present the early evidence for single-band d-wave superconductivity in $\mathrm{CeCu}_2\mathrm{Si}_2$ down to about $T = 0.1$ K, see also (Stockert *et al.*, 2012).

III. EVIDENCE FOR d-WAVE SUPERCONDUCTIVITY IN $\mathrm{CeCu}_2\mathrm{Si}_2$ ABOVE 0.1 K

A. Phase diagram

One of the major distinguishing features which sets $\mathrm{CeCu}_2\mathrm{Si}_2$ apart from previously-known BCS superconductors is the close proximity between magnetism and superconductivity in the phase diagram, where both are due to the same localized $4f$-electrons. This is reflected in the observation that slight tuning of the Cu:Si ratio within the homogeneity range can lead to crystals with ground states which are entirely antiferromagnetic (A-type), superconducting (S-type) or exhibit both superconductivity and magnetism (A/S-type) (Seiro *et al.*, 2010; Steglich *et al.*, 1996). While within the context of BCS theory, magnetism and superconductivity are generally considered antagonistic, superconductivity on the border of magnetism is a common feature of broad classes of unconventional superconductors (Norman, 2013, 2011; Stewart, 2017) including heavy-fermion superconductors (Pfleiderer, 2009; Steglich and Wirth, 2016), cuprates (Lee *et al.*, 2006; Proust and Taillefer,

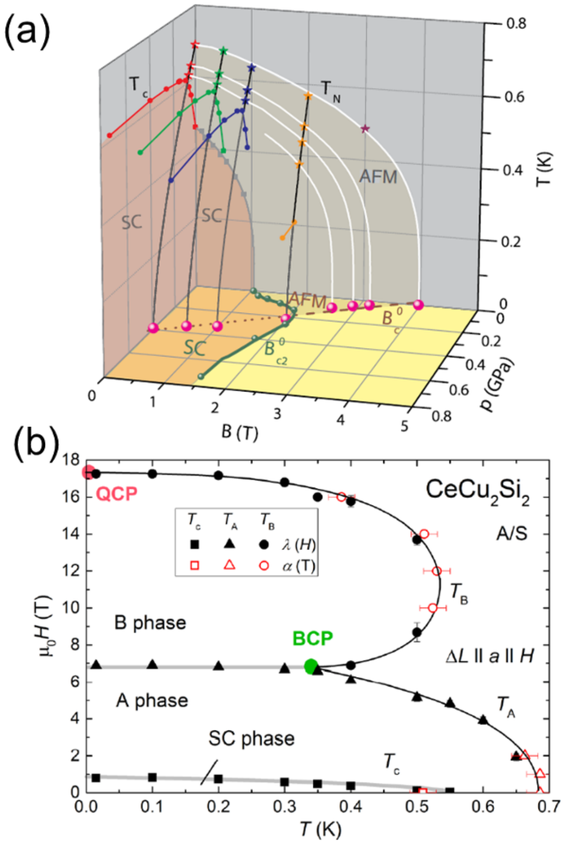

FIG. 4 (a) Temperature-pressure-magnetic field phase diagram of a single crystal of A/S-type CeCu$_2$Si$_2$. Reproduced with permission from Lengyel *et al.*, 2011. (b) Magnetic field - temperature diagram of single crystalline CeCu$_2$Si$_2$, where positions of the field-induced bicritical point (BCP) and QCP are also displayed. Reproduced with permission from Weickert *et al.*, 2018.

2019), iron-based pnictides and chalcogenides (Si *et al.*, 2016; Stewart, 2011), organic superconductors (Kanoda, 2008; Lang and Müller, 2003; Maple *et al.*, 2004) and twisted graphene superlattices (Andrei and MacDonald, 2020; Cao *et al.*, 2018), and may be related to the occurrence of Cooper pairs with a magnetically driven pairing interaction (Scalapino, 2012), rather than the conventional electron-phonon pairing mechanism.

The temperature-pressure-magnetic field diagram of an A/S-type single crystal is displayed in Fig. 4(a) (Lengyel *et al.*, 2011). At ambient pressure, two zero-field phase transitions can be detected in specific heat measurements corresponding to an antiferromagnetic transition at $T_{\rm N} = 0.69$ K and a subsequent superconducting transition at $T_{\rm c} = 0.46$ K. The application of moderate pressure rapidly suppresses $T_{\rm N}$, while $T_{\rm c}$ shows a slight increase, and once $T_{\rm N}$ is suppressed below $T_{\rm c}$, no antiferromagnetic transition is observed. When a magnetic field is applied, both $T_{\rm N}$ and $T_{\rm c}$ are suppressed, but the more rapid decrease of $T_{\rm c}$ with field allows for $T_{\rm N}$ (extrapolated

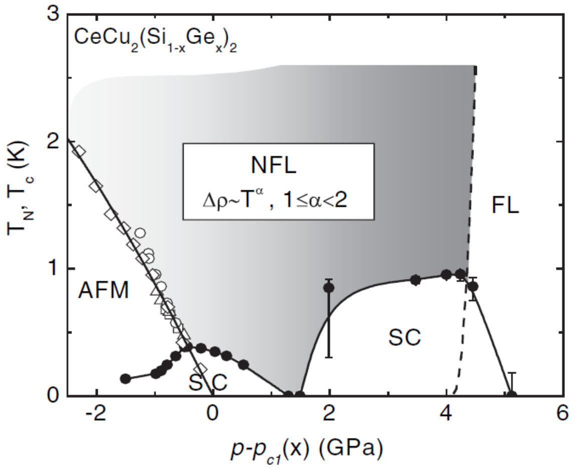

FIG. 5 Temperature - pressure phase diagram of CeCu$_2$(Si$_{1-x}$Ge$_2$)$_2$, which exhibits two superconducting domes, one centered around a lower pressure $p_{\rm c1}$ associated with an antiferromagnetic QCP, while the dome at higher pressures is near a possible valence transition. The diamonds, circles, triangles and squares correspond to compositions with $x = 0.25, 0.1, 0.05$, and 0.01, respectively. The dashed line displays the anticipated line of first-order valence transitions, ending in a critical point somewhere between 10 and 20 K. Solid lines are guides to the eye. Reproduced with permission from Yuan *et al.*, 2006.

to $B = 0$) to be tracked as a function of pressure to lower temperatures. From extrapolating the positions of B_c^0 (where $T_{\rm N}$ vanishes) for fixed values of pressure, a line of QCPs is inferred to lie in the zero-temperature pressure-field phase diagram shown in Fig. 4(a). $B_c^0(p) = 0$ at $p_c = 0.39$ GPa, which is almost twice as large as the pressure where B_{c2}^0 vs p exhibits a local maximum (see Sec. V). p_c can be forced to vanish, if the composition of the (homogeneous) sample becomes slightly more enriched by Cu (i.e., by reducing the average unit-cell volume). Although the ambient-pressure, zero-field QCP is masked by superconductivty, its nature can be well explored by studying the low-temperature normal state of such an S-type sample induced by applying a small external magnetic field, see Sec. III.C.

Detailed measurements of the elastic constants, thermal expansion, and magnetostriction revealed the presence of a field-induced 'B' phase, in addition to the magnetic 'A' phase found at low fields (Bruls *et al.*, 1994; Weickert *et al.*, 2018). The field-temperature phase diagram is displayed in Fig. 4(b), where there are second-order lines between the paramagnetic state and both the 'A' and 'B' phases, while going from the 'A' to 'B' phase corresponds to a first-order transition, leading to a bicritical point in the phase diagram between these phases (Weickert *et al.*, 2018). Measurements to very low temperatures and high fields show the suppression of the 'B' phase to zero temperature in applied fields of around 17 T, giving rise to a field-induced QCP. The nature of the transition from 'A' to 'B' phase is still to be determined, where the small change in magnetization between the two phases suggests that 'B' (like 'A', see below) also corresponds to a spin-density-wave (SDW) phase (Tayama *et al.*, 2003; Weickert *et al.*, 2018).

The shape of the superconducting region in the temperature-pressure phase diagram of S-type $CeCu_2Si_2$ is quite unusual compared to other heavy-fermion superconductors (Knebel *et al.*, 2006; Mathur *et al.*, 1998), namely at low and moderate pressures T_c does not change rapidly with pressure, while at higher pressures it reaches a maximum at around 4 GPa, well away from the point where magnetism is suppressed (Bellarbi *et al.*, 1984; Thomas *et al.*, 1993; Yuan *et al.*, 2003, 2006). Remarkably, upon substituting 10 at% of Si by Ge, which substantially reduces T_c, it is found that this actually results in two superconducting domes in the phase diagram, as shown in Fig. 5, where one is centered around the antiferromagnetic QCP, and another with a higher maximum T_c occuring at higher pressures (Yuan *et al.*, 2003, 2006). It has been suggested that these two domes correspond to superconductivity with different unconventional pairing mechanisms, with the low pressure dome corresponding to magnetically driven superconductivity and the high pressure dome driven by charge (valence) fluctuations (Holmes *et al.*, 2004; Yuan *et al.*, 2003). A similar phase diagram with two superconducting domes was reported for the (Pu,Co)-based 115 systems by (Bauer *et al.*, 2012). Here, the higher T_c of $PuCoGa_5$ (18.5 K) compared to $PuCoIn_5$ (2.5 K) was ascribed to the superconductivity of the former arising from a valence instability, while that of the latter was associated with a magnetic quantum critical point.

B. Origin of the A-phase in $CeCu_2Si_2$

Although the relative increase of the electrical resistivity below the ordering temperature T_N suggested the opening of an excitation gap in $CeCu_2Si_2$ due to a SDW-type of magnetic order (Gegenwart *et al.*, 1998), direct evidence for such a scenario was lacking for a long time. The first indications for antiferromagnetic order as the characteristic of the A-phase came from NMR (Nakamura *et al.*, 1988) and μSR measurements (Uemura *et al.*, 1988, 1989) in the late 1980s, both detecting a static magnetic field (at the muon site or the nuclear site, respectively) in the ordered state. In these measurements even an incommensurate type of magnetic order in $CeCu_2Si_2$ was proposed because of the distribution of local magnetic fields detected. Interestingly, while pronounced phase transition anomalies at T_N were found in both elastic-constant and thermal-expansion measurements (Bruls *et al.*, 1994), no corresponding feature was seen in the magnetic susceptibility for a long time, until a cusp-like anomaly could eventually be resolved in the susceptibility when monitored with the aid of a high-resolution Faraday magnetometer (Tayama *et al.*, 2003).

In 1997, antiferromagnetic order was observed in the reference compound $CeCu_2Ge_2$ using single crystal neutron diffraction (Krimmel *et al.*, 1997) which later could be related to the nesting properties of the Fermi surface (Zwicknagl, 2007). In order to unravel the nature of the A-phase in pure $CeCu_2Si_2$, an approach to study the magnetic order in the Ge-substituted system $CeCu_2(Si_{1-x}Ge_x)_2$ was chosen. Starting from pure $CeCu_2Ge_2$ the antiferromagnetic order was followed in $CeCu_2(Si_{1-x}Ge_x)_2$ with decreasing Ge content. Initially the incommensurate order in $CeCu_2(Si_{1-x}Ge_x)_2$ was detected only for $x \geq 0.6$ in neutron powder diffraction (Knebel *et al.*, 1996; Krimmel and Loidl, 1997). However, measurements in powder samples with lower Ge concentrations were unsuccessful since the ordering temperature as well as the magnetically ordered moment are largely reduced for samples with low Ge content. Until the early 2000s only very small single crystals were available just enabling thermodynamic and transport measurements. Then, with substantially improved crystal growth techniques (Cao *et al.*, 2011; Seiro *et al.*, 2010), quite large single crystals of $CeCu_2Si_2$ and $CeCu_2(Si_{1-x}Ge_x)_2$ (up to $\sim \mathrm{cm}^3$ size) could be synthesized. Now performing single crystal neutron diffraction on $CeCu_2(Si_{1-x}Ge_x)_2$ the antiferromagnetic order could be followed to much lower Ge concentrations (Stockert *et al.*, 2005, 2003). Finally, incommensurate antiferromagnetic order was even detected in pure A-type $CeCu_2Si_2$ with a small ordered magnetic moment $\approx 0.1\,\mu_B/\mathrm{Ce}$ (Stockert *et al.*, 2004) as shown in Fig. 6(a). The propagation wave vector $\mathbf{k} = \mathbf{Q_{AF}} = (0.215\ 0.215\ 0.53)$ at $T = 50\,\mathrm{mK}$ agrees well with theoretical calculations of the Fermi surface using a renormalized band method (Stockert *et al.*, 2004; Zwicknagl, 1992; Zwicknagl and Pulst, 1993). They indicate nesting properties in the corrugated part of the cylindrical Fermi surface of the heavy quasiparticles at the X point of the bulk Brillouin zone [see Fig. 6(b) and Sec. IV.B, below]. Hence, the magnetic order in $CeCu_2Si_2$ is an incommensurate SDW. This is further supported by the temperature dependence of the propagation wave vector below the ordering temperature. It is worth noting that the propagation vectors in $CeCu_2(Si_{1-x}Ge_x)_2$ are quite similar with the largest difference being the $a*, b*$ component changing from 0.215 in pure $CeCu_2Si_2$ to 0.282 in $CeCu_2Ge_2$ and almost no change in the $c*$ component remaining close to 0.5 (Stockert *et al.*, 2005).

The interplay between antiferromagnetism and superconductivity has been studied on small A/S-type $CeCu_2Si_2$ single crystals, where μSR measurements indicated a competition of both phenomena with a full repulsion of antiferromagnetism in the superconducting state (Feyerherm *et al.*, 1997; Luke *et al.*, 1994; Stockert *et al.*, 2006), in contrast to earlier reports on polycrystalline

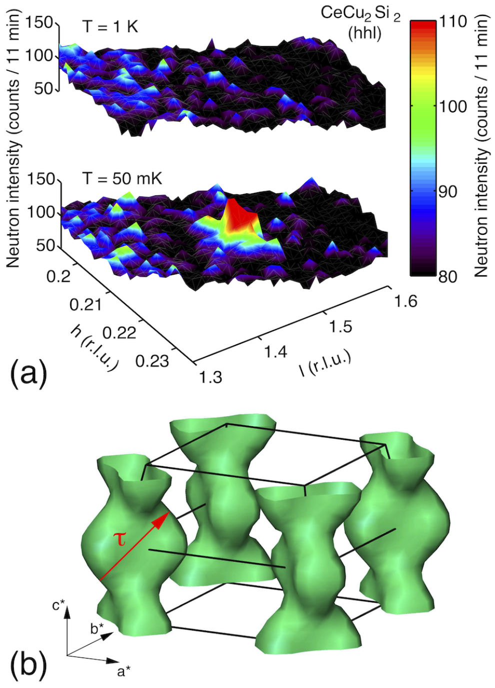

FIG. 6 (a) Neutron-diffraction intensity map of the reciprocal (h h l) plane around $\mathbf{Q} = (0.21\ 0.21\ 1.45)$ in A-phase CeCu$_2$Si$_2$ at $T = 50\,$mK and 1 K. (b) Main heavy Fermi surface sheet in CeCu$_2$Si$_2$ indicating the columnar nesting with wave vector $\boldsymbol{\tau}$. Reproduced with permission from Stockert *et al.*, 2004.

samples (Uemura *et al.*, 1988). Neutron diffraction on quite large A/S-type CeCu$_2$Si$_2$ single crystals also revealed that magnetic order and superconductivity do not coexist in CeCu$_2$Si$_2$ on a microscopic scale (Arndt *et al.*, 2010; Thalmeier *et al.*, 2005).

C. Quantum criticality

Common to many magnetically ordered Ce-based heavy-fermion systems, the application of pressure tunes the relative strengths of the magnetic exchange interactions (Ruderman-Kittel-Kasuya-Yosida interaction) and Kondo coupling, and for sufficiently large pressures the Kondo interaction dominates, suppressing magnetic order. In several cases this allows for the tuning of a second-order antiferromagnetic transition continuously to zero temperature at a QCP, leading to the breakdown of Fermi liquid behavior at finite temperatures (Löhneysen *et al.*, 2007; Sachdev, 2011; Stewart, 2001, 2006). The RKKY interaction leads to antiferromagnetic correlations between the local moments, which reduce the amplitude of the Kondo singlet in the ground state.

Two classes of QCPs have been advanced in recent years, depending on whether this static Kondo-singlet amplitude is destroyed (Coleman *et al.*, 2001; Senthil *et al.*, 2004; Si *et al.*, 2001) or remains nonzero at the antiferromagnetic QCP (Coleman and Schofield, 2005; Si and Steglich, 2010). Prototype examples of the former case of Kondo-destruction quantum criticality include Au-doped CeCu$_6$ (Schröder *et al.*, 2000), YbRh$_2$Si$_2$ (Friedemann *et al.*, 2010; Gegenwart *et al.*, 2007; Paschen *et al.*, 2004) and CeRhIn$_5$ (Park *et al.*, 2006; Shishido *et al.*, 2005). In the latter case, the quantum critical behavior at energies below a certain crossover scale $E^* = k_B T^*$ is of the SDW-type, however the critical fluctuations of the Kondo effect, i.e. partial Mott physics, sets in above the crossover energy scale. CeCu$_2$Si$_2$ shows evidence for a line of QCPs as a function of magnetic field under pressure, in the vicinity of the disappearance of magnetic order, see Fig. 4(a). For an S-type polycrystalline sample in the low temperature normal state, the signatures of a 3D SDW-type QCP are found from a $T^{\frac{3}{2}}$ dependence of the resistivity, as well as a $-T^{\frac{1}{2}}$ dependence of the specific heat coefficient (Gegenwart *et al.*, 1998). In addition, the spin-excitation spectrum at the nesting wave vector $\boldsymbol{\tau} \approx \mathbf{Q}_{AF}$ in the normal state of superconducting (S-type) CeCu$_2$Si$_2$ displays an almost critical slowing down when superconductivity is suppressed by a magnetic field (Arndt *et al.*, 2011), as expected for a compound located very close to a QCP. Moreover, an $E/T^{3/2}$ scaling of the normal state magnetic response [Fig. 7(b)] and a $T^{3/2}$ dependence of the inverse lifetime of the spin fluctuations [Fig. 7(d)] indicate that in CeCu$_2$Si$_2$ a 3D SDW-type QCP seems to be realized, in line with the aforementioned thermodynamic and transport measurements (Arndt *et al.*, 2011; Stockert *et al.*, 2011). Measurement of the damping rate from inelastic neutron scattering (INS) has provided evidence that the Kondo-destruction temperature scale, T^*, is nonzero but small (Smidman *et al.*, 2018) compared to the bare Kondo scale of 15 K. Changes in $C(T)/T$ from a square-root to logarithmic dependence and in the quasielastic neutron-scattering damping rate from $T^{3/2}$ to T-linear behavior are observed between 1 and 2 K, suggesting that T^* is of a similar size.

It is to be noted that $\mathbf{Q}_{AF}$ is not a singular point in ($\mathbf{Q}$,ω) space, but paramagnons are emerging out of $\mathbf{Q}_{AF}$ with an initial linear dispersion (Arndt *et al.*, 2011; Stockert *et al.*, 2011). Comparing the magnetic response in the normal state of superconducting (S-type) CeCu$_2$Si$_2$ and the antiferromagnetic state in A-type CeCu$_2$Si$_2$, the dispersion of the (para)magnons in both states was found to be very similar, with just higher intensity for the A-type sample (Huesges *et al.*, 2018). Upon entering the superconducting state, the dispersion of the paramagnons remains (almost) unchanged with deviations only occurring at low energy transfers below 0.5 meV due to the formation of a spin gap (Stockert *et al.*, 2011), see Sec. III.D. Very recently, INS on S-type CeCu$_2$Si$_2$ have been extended to higher energy transfers up to several meV (Song *et al.*, 2021). These mea-

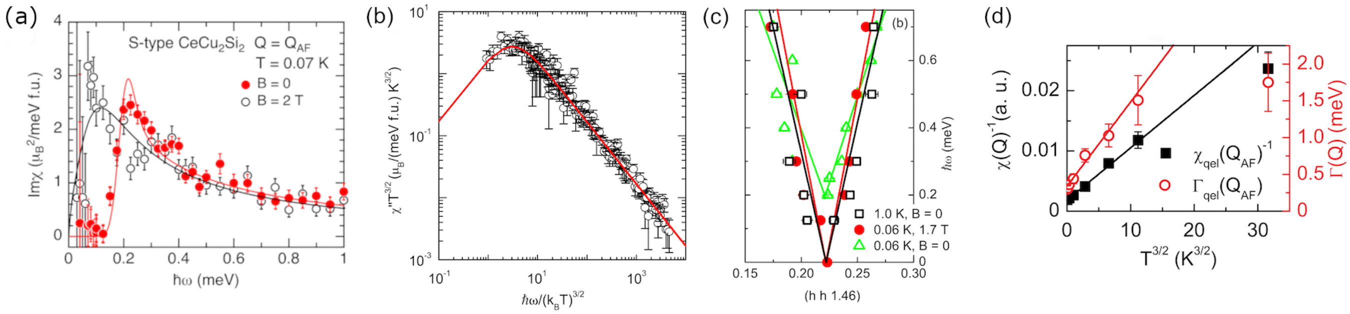

FIG. 7 Spin dynamics in CeCu$_2$Si$_2$. (a) Low-energy spin excitations in S-type CeCu$_2$Si$_2$ at $\mathbf{Q}_{AF}$ and $T = 0.07$ K in the superconducting ($B = 0$) and the normal state ($B = 2\,$T). Reproduced from Stockert *et al.*, 2011. (b) Scaling of the normal-state quasielastic response in S-type CeCu$_2$Si$_2$ at $\mathbf{Q}_{AF}$ and at $B = B_{c2} = 1.7\,$T indicating universal scaling of the dynamical susceptibility $\chi'' T^{3/2}$ vs. $\omega/T^{3/2}$. (c) Dispersion of the spin excitations in the normal and superconducting states of S-type CeCu$_2$Si$_2$. (d) Relaxation rate Γ and inverse susceptibility $\chi(\mathbf{Q})^{-1}$ of the normal-state magnetic response at $\mathbf{Q} = \mathbf{Q}_{AF}$in S-type CeCu$_2Si_2$ versus $T^{3/2}$. (b)-(d) are reproduced with permission from Arndt *et al.*, 2011.

surements fully confirm the previous experiments at low energies, i.e., the spin gap in the superconducting state (Stockert *et al.*, 2011) and the dispersive paramagnons (Arndt *et al.*, 2011; Huesges *et al.*, 2018; Stockert *et al.*, 2011). However, in addition, the dispersive spin excitations are now found to change to a dispersionless column in energy above $\approx 1.5\,$meV (Song *et al.*, 2021). The transition from dispersive to dispersionless magnetic excitations occurs around $k_{\mathrm{B}}T_{\mathrm{K}}$, i.e., the characteristic local energy scale in CeCu$_2$Si$_2$. Currently it is an open question, if and how these high-energy spin excitations are related to the unconventional heavy-fermion superconductivity in CeCu$_2$Si$_2$.

Another issue that has to be clarified by future work concerns the difference in the quantum critical exponent α of the temperature dependence in the low-T resistivity of undoped CeCu$_2$Si$_2$, $\Delta\rho(T) = A'T^{\alpha}$. As already mentioned, this was found to be $\alpha = \frac{3}{2}$ in (Gegenwart *et al.*, 1998), whereas $\alpha = 1$ was reported in (Yuan *et al.*, 2003, 2006). In both cases, the samples had been prepared with some Cu excess pointing to S-type samples. As shown by neutron diffraction (Stockert *et al.*, 2004) as well as earlier μSR results (Feyerherm *et al.*, 1997; Luke *et al.*, 1994), these samples contain a minority phase of A-type that is microscopically separated from the S-type majority phase. It may be possible that, depending on the content and spatial distribution of this minority phase, the volume-integrated response in resistivity experiments is eventually responsible for the differing T-dependences observed.

D. Spin dynamics in superconducting CeCu$_2$Si$_2$

Due to the small magnetic moment and the low transition temperatures, INS experiments were performed on superconducting CeCu$_2$Si$_2$ using cold-neutron triple-axis spectroscopy. While the INS spectra in the normal state yield a quasielastic magnetic response at $\mathbf{Q}_{AF}$ with slowing down and scaling behavior as already mentioned, the spin dynamics in the superconducting state well below $T_c = 0.6\,$K show a clear spin excitation gap at $\mathbf{Q}_{AF}$(Stockert *et al.*, 2011, 2008) followed by a well defined maximum, often called 'spin resonance' [Figs.7(a) and (b)]. It should be noted that this maximum clearly exceeds the magnetic response in the normal state, in contrast to a simple s-wave superconductor where no obvious enhancement of the superconducting response over the normal state response is expected at energies above the spin gap. Its intensity depends on the Fermi surface topology and the paramagnon dispersion (Eremin *et al.*, 2008) and might therefore be less pronounced than in other unconventional superconductors. With a spin-gap size of about 0.2 meV, the maximum is located at $4k_{\mathrm{B}}T_c$ and its position is therefore smaller than $2\Delta = 5k_{\mathrm{B}}T_c$ of the (large) charge gap (Fujiwara *et al.*, 2008). We note that this necessary condition for a 'spin resonance' to be located inside 2Δ was also fulfilled by the acoustic magnon peak in UPd$_2$Al$_3$ (in which the U^{3+} ion has two localized and one more hybridized $5f$-electron), where heavy-fermion superconductivity coexists with local-moment antiferromagnetic order (Sato *et al.*, 2001). Like in the latter case as well as in CeCoIn$_5$ (Song *et al.*, 2016, 2020), the peak in CeCu$_2$Si$_2$ develops in the one-particle channel, i.e. out of the aforementioned quasielastic line that persists to way above T_c for the two Ce-based compounds, and to well above T_N ($> T_c$) for UPd$_2$Al$_3$. This is different from the cuprates where it manifests a singlet-triplet excitation of the d-wave condensate (Sidis *et al.*, 2004).

Though this distinct maximum in the INS data at the edge of the spin-excitation gap should not be called a 'spin resonance' for the reasons given above, it nevertheless highlights a sign-changing superconducting order parameter. Namely, if one considers coupling between a magnetic mode (e.g. magnon/magnetic exciton) and the itinerant quasiparticles/Cooper pairs (Bernhoeft *et al.*, 2006, 1998), the observation in $\mathrm{CeCu_2Si_2}$ of a significant low energy enhancement of the INS intensity along the propagation vector $\mathbf{Q}_{\mathrm{AF}}$ in the superconducting state over that of the normal state (Stockert *et al.*, 2011), implies a large coherence factor , which necessarily requires a sign change of the superconducting order parameter along this wave vector. Alternatively, for $\mathrm{CeCoIn_5}$ and Fe-based superconductors it has been proposed that the low energy INS peak arises from reduced quasiparticle damping in the superconducting state, allowing for the observation of an otherwise overdamped magnon mode (Chubukov and Gor'kov, 2008; Onari *et al.*, 2010). For two reasons, we do not consider this scenario to be viable. First, the ratio of the energy of the INS maximum to 2Δ is comparable to the universal value observed in a variety of correlated superconductors (Duan *et al.*, 2021; Yu *et al.*, 2009). Second (and related to the first), for this scenario to occur, the universality of the INS peak in the superconducting state which occurs in a variety of systems requires some degree of commonality in the behavior of the low-energy spin excitations in their normal states. This expectation is to be contrasted with disparate behavior of the low-energy spin excitations that have been observed in the normal state of these systems. In particular, in the case of $\mathrm{CeCu_2Si_2}$, even in the normal state the paramagnons do not appear to be overdamped as suggested by their well visible dispersion at low energies [cf. Fig. 7(c)], even up to $k_B T_K \approx 1.5$ meV (Song *et al.*, 2021).

The experimentally determined propagation vector $\mathbf{Q}_{\mathrm{AF}}$ agrees very well with the theoretically obtained nesting wave vector $\boldsymbol{\tau}$ shown in Fig. 6(b) (Stockert *et al.*, 2004; Zwicknagl, 1992; Zwicknagl and Pulst, 1993), which connects nested parts of the heavy quasiparticle band, highlighting *intraband* nesting. Importantly, $\mathbf{Q}_{\mathrm{AF}}$ does not connect extended regions of different bands (interband nesting) of e.g. electron and hole bands (Sec. IV.B) as required by the s_{+-} pairing model that has been considered for some Fe-based superconductors (Mazin *et al.*, 2008).

E. Effects of potential scattering

Historically, the effect of nonmagnetic impurities has been an important test for unconventional superconductivity. This is because, while the T_{c} of a conventional BCS superconductor is very sensitive to magnetic impurities, the non-magnetic case has little effect (Anderson, 1959). On the other hand, for superconductors with unconventional sign-changing states, the effect of nonmagnetic impurities may become similar to magnetic impurities in a conventional material (Balatsky *et al.*, 2006). Indeed the high sensitivity of $\mathrm{CeCu_2Si_2}$ to a very small amount of atomic substitution of nonmagnetic impurities was one of the key pieces of early evidence allowing for the identification of an unconventional superconducting state. This is particularly the case for substitutions on the Cu site, where as shown in Fig. 8, doping around 1% of Rh, Pd or Mn completely suppresses T_{c}, while similarly only 0.5% of smaller Sc^{3+} on the Ce site is needed (Spille *et al.*, 1983). A striking difference for the Ce site is the size dependence of the dopant, where T_{c} becomes increasingly insensitive for larger substituents, i.e., with critical concentrations of 6% for Y^{3+}, 10% for La^{3+} (Spille *et al.*, 1983), culminating in 20% for Th^{4+} (Ahlheim *et al.*, 1990). This trend with chemical pressure is analoguous to that found when applying hydrostatic pressure to $\mathrm{CeCu_2Si_2}$ doped with 10 at% of Ge, where T_{c} is suppressed on the high pressure side of the low-pressure dome centered around the antiferromagnetic QCP [Fig. 5]. While this size effect appears to be in line with the strength of the Kondo interaction in the dependence of the volume available to the Ce^{3+} ions, the reason for the distinct site dependence in the atomic substitution experiments is yet to be unraveled.

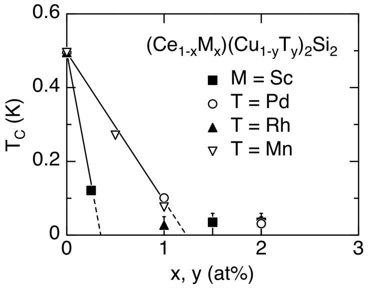

FIG. 8 Dependence of the superconducting transition temperature of $\mathrm{CeCu_2Si_2}$ polycrystals on substitutions for Ce and Cu. Replotted from (Spille *et al.*, 1983).

Such small critical substitutions on the transition-metal side being needed to suppress T_{c} proves that this cannot be simply due to a significant tuning of the Kondo state. Indeed, while Ge doping expands the lattice acting as a negative pressure effect and causes a slight decrease of T_K, it is found that tuning a Ge-doped sample using pressure, which causes an increase of T_K, still yields a greatly suppressed T_{c} (Yuan *et al.*, 2003, 2006). Such a reduction of T_{c} upon 10% Ge-doping allowed for the revelation that there are two separate superconducting domes in the temperature-pressure phase diagram, one sitting near a magnetic QCP, while the higher pressure dome potentially lies near a valence transition (Fig. 5) (Holmes *et al.*, 2004; Yuan *et al.*, 2003). Meanwhile for a more disordered sample with 25% Ge-doping no superconductivity is recovered even after the suppression of magnetism by pressure (Yuan *et al.*, 2004).

On the basis of recent studies of electron irradiated samples, it was proposed that the order parameter of $CeCu_2Si_2$ does not change sign across the Fermi surface, much like a conventional BCS superconductor. Namely it is reported that the suppression of T_c upon the introduction of disorder by electron irradiation is not as rapid as expected for sign-changing pairing states such as those in the cuprates or iron pnictides, but instead is similar to some materials believed to have a conventional pairing mechanism (Yamashita *et al.*, 2017). Moreover the lack of change in the low-temperature penetration depth of electron irradiated samples is taken as evidence for a lack of low-energy impurity-induced bound states, as also expected for sign-preserving order parameters (Takenaka *et al.*, 2017). Since the effect of electron irradiation is likely to correspond to the displacement of Ce atoms from the lattice to interstitial sites, the resulting disorder may be compared to that manifested by the strong (factor of four) variation in the residual resistivity ρ_0 going from a nearly stoichiometric A/S-type to an S-type single crystal with a small amount of Cu excess, where no depression of T_c is observed (Pang *et al.*, 2018). Similar results are well known from the cuprate high-T_c superconductors where substantial variations in ρ_0 are not reflected by any significant changes in T_c, cf. results on YBCO polycrystals (Cava *et al.*, 1987) and single crystals (Liang *et al.*, 1992). As discussed in Sec. IV.C, there are a number of theoretical works underlining the robustness of unconventional superconductivity against certain kinds of ordinary potential scattering (Anderson, 1997; Si *et al.*, 2016). However, from the cuprates it is also known that atomic substitution can be quite hostile for high-T_c superconductivity (Alloul *et al.*, 2009). For example, partial substitution of Cu on the CuO_2 planes by Zn causes a strong depression of T_c (Xiao *et al.*, 1988). Obviously, this is quite similar to the results of the aforementioned substitution experiments on $CeCu_2Si_2$ (Ahlheim *et al.*, 1988; Spille *et al.*, 1983; Yuan *et al.*, 2003), which are at odds with a non-sign changing superconducting state.

In summary, the aforementioned studies on $CeCu_2Si_2$ reveal that "impurity doping", i.e., substitutional disorder, is strongly pairbreaking, while certain kinds of lattice rearrangements, induced, e.g., by electron irradiation or small changes in the Cu/Si occupation, are harmless to superconductivity. This dichotomy of harmful and harmless disorder in unconventional heavy fermion and cuprate high-T_c conductors still needs to be uncovered.

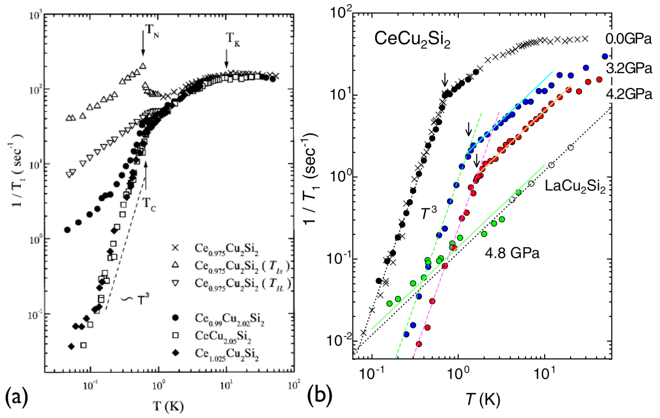

FIG. 9 Temperature dependence of the spin lattice relaxation rate $1/T_1(T)$ of $CeCu_2Si_2$, obtained from Cu-NQR measurements (left) for various superconducting and non-superconducting polycrystals, and (right) single crystals of superconducting $CeCu_2Si_2$ under hydrostatic pressure, as well as $LaCu_2Si_2$. No Hebel-Slichter peak at T_c and a T^3 dependence is found in the superconducting samples down to around 0.1 K. The left panel is reproduced with permission from Ishida *et al.*, 1999. The right panel is reproduced from Fujiwara *et al.*, 2008, copyright 2008 The Physical Society of Japan.

F. Evidence for d-wave pairing

For a long time, the pairing state of $CeCu_2Si_2$ was generally believed to correspond to d-wave superconductivity, in line with other Ce-based heavy-fermion superconductors (Thompson and Fisk, 2012), cuprate materials (Lee *et al.*, 2006; Scalapino, 1995), and organic superconductors (Kanoda, 2008; Lang and Müller, 2003). A decrease of the Knight shift below T_c, the ordinary size of the dc Josephson effect between polycrystalline $CeCu_2Si_2$ and Al as well as evidence for Pauli limiting of the upper critical field (Fig. 3), confirmed quite early that the Cooper pairs correspond to a singlet pairing state (Assmus *et al.*, 1984; Ueda *et al.*, 1987). Meanwhile the clearest evidence for the superconducting gap structure came from Cu-NQR measurements, where the spin lattice relaxation rate $(1/T_1(T))$ displayed in Fig. 9 shows a T^3 dependence down to around 0.1 K, which is characteristic of line nodes in the superconducting gap (Fujiwara *et al.*, 2008; Ishida *et al.*, 1999), although it should be noted that the $1/T_1(T)$ data of Ishida *et al.* (Ishida *et al.*, 1999) also show some deviation from T^3 behavior at the lowest temperatures, as clearly demonstrated in recent NQR experiments extended to somewhat lower temperature (Kitagawa *et al.*, 2017), see below. Evidence for nodal superconductivity was also inferred from measurements of other thermodynamic quantities, including a T^2 dependence of the magnetic penetration depth (Gross *et al.*, 1988), which is consistent with d-wave superconductivity in the presence of strong impurity scattering (Hirschfeld and Goldenfeld, 1993). The requirement that the order parameter is (i) spin singlet, (ii) with gap nodes, and (iii) changes sign on the regions of the renormalized Fermi surface connected by the nesting wave vector $\boldsymbol{\tau} \approx \mathbf{Q}_{AF}$, is most readily satisfied by a $d_{x^2-y^2}$ pairing state, similar to that generally believed to apply to the cuprate high-T_c superconductors (Scalapino, 1995). On the other hand, in isothermal magnetoresistance measurements the angu-

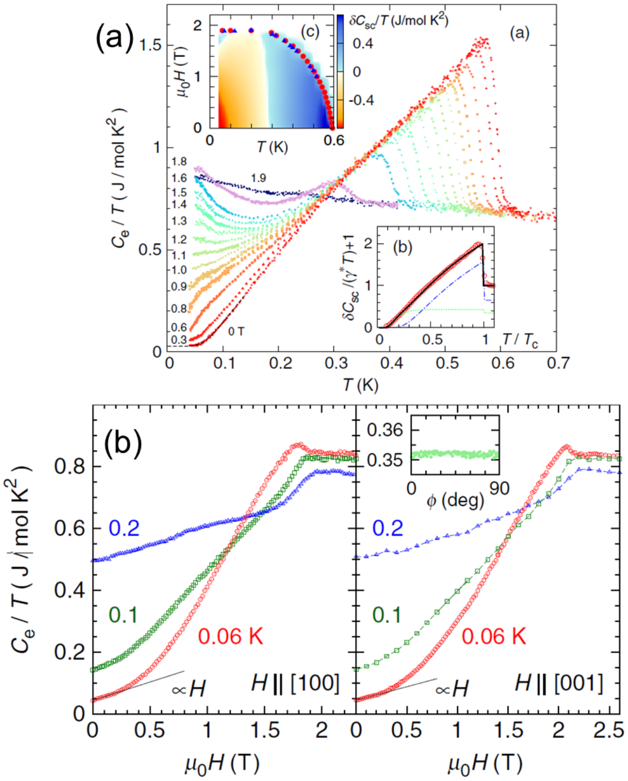

FIG. 10 (a) Temperature dependence of the electronic contribution to the specific heat as C_e/T of CeCu$_2$Si$_2$ down to temperatures of 0.04 K. The lower inset displays the analysis of the data using a nodeless two-gap model, while the upper panel shows the data as a contour plot. (b) The field dependence of the electronic specific heat coefficient at various temperatures for fields along the [100] (left) and [001] directions. At 0.06 K, linear behavior is observed at low fields for both field orientations. The inset in the right panel shows C_e/T as a function of the in-plane azimuthal field-angle ϕ, which remains constant. Reproduced with permission from Kittaka *et al.*, 2014.

lar dependence of the upper critical field in the *ab*-plane at 40 mK was found to be most compatible with a d_{xy} state, although the small amplitude of this modulation made it difficult for firm conclusions to be drawn (Vieyra *et al.*, 2011). Nevertheless, a *d*-wave pairing state of some form with line nodes was long considered to be the most likely candidate pairing state.

IV. FULLY GAPPED UNCONVENTIONAL SUPERCONDUCTIVITY IN CeCu$_2$Si$_2$

A. Evidence for a nodeless gap structure

The understanding of the superconducting state of CeCu$_2$Si$_2$ underwent a radical overhaul following the results from low-temperature specific measurements of Kittaka *et al.* (Kittaka *et al.*, 2014, 2016), which revealed that the superconducting gap is fully open over the whole Fermi surface. Here the temperature dependence of the electronic contribution to the specific heat C_e ($\approx C_{4f}$) of S-type single crystals measured down to 0.04 K begins to flatten upon approaching the lowest measured temperature, and was best described by an exponentially activated temperature dependence, rather than following the $C_e \sim T^2$ behavior of a superconductor with line nodes, as shown in Fig. 10(a). This analysis suggested nodeless superconductivity with a gap $\Delta_0 = 0.39k_BT_c$. Since this is considerably less than the value of $1.76k_BT_c$ derived from weak-coupling BCS theory, in order to demonstrate the presence of a fully open gap in thermodynamic quantities such as the specific heat and penetration depth, measurements across a wide temperature range down to at least 0.05 K are required. A further advantage of this study is the very small residual $\gamma_0 = 0.028$ J mol^{-1} K^{-2}, which again allows for the inference of a lack of low-energy excitations. After subtracting an estimate of the phonon contribution, the data up to T_c could not be described by a model with a single gap, but were instead accounted for by a model with two nodeless isotropic gaps.

These conclusions were supported by specific heat measurements in applied magnetic fields, displayed in Fig. 10(b). Here the isothermal C_e/T at the lowest temperature of 0.06 K exhibits a linear field dependence, as opposed to the $H^{0.5}$ behavior of a *d*-wave superconductor. The range of this low-field linear region is relatively narrow, and at higher fields there is a pronounced increase of C_e/T. Just below B_{c2}, C_e/T even overshoots the normal state value, and the origin of this strong enhancement needs still to be clarified by future work. Upon rotating the field within the *ab*-plane, no modulation of the specific heat is observed, whereas in the single band *d*-wave scenario a four-fold oscillation is predicted theoretically (Boyd *et al.*, 2009; Vorontsov and Vekhter, 2007), and observed experimentally in the CeTIn$_5$ series of heavy-fermion superconductors (An *et al.*, 2010; Aoki *et al.*, 2004; Lu *et al.*, 2012). Furthermore, measurements as a function of the polar angle θ reveal simply the two-fold oscillations arising naturally from the tetragonal symmetry (Kittaka *et al.*, 2016).

It should be noted that early evidence for a potentially exponential temperature dependence of the low-temperature specific heat was provided by measurements of CeCu$_2$Si$_2$ polycrystals, which revealed power-law behaviour with an exponent of two near T_c, but close to three at $T =$ 0.05 K (Steglich *et al.*, 1985a). Further early evidence for fully gapped superconductivity was reported from a point contact study of CeCu$_2$Si$_2$ measured at 0.03 K (De Wilde *et al.*, 1994). They found that the differential resistance curves are flat around zero bias, which is characteristic of a fully open gap, in stark contrast to that observed in UPt$_3$ where the curves have a triangular shape around zero voltage suggesting the presence of gap nodes.

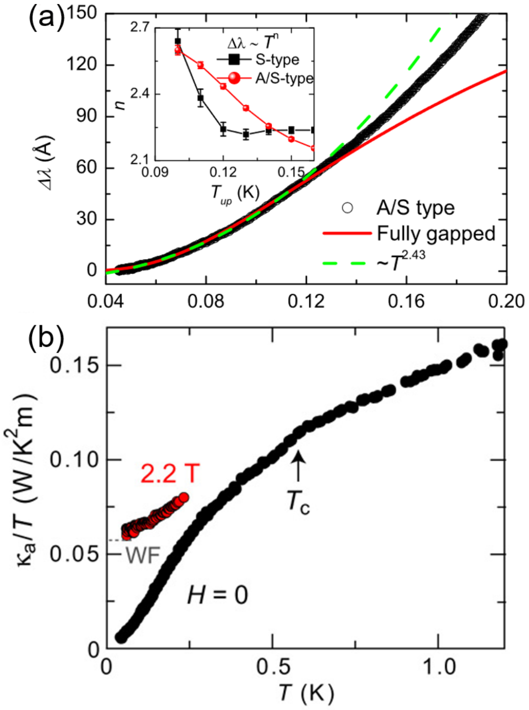

FIG. 11 (a) Temperature dependence of the magnetic penetration depth shift of an A/S-type $CeCu_2Si_2$ single crystal, measured using the tunnel-diode-oscillator-based method. The solid line shows the fit to the low temperature data with a model for a fully gapped superconductor, where the lack of nodes in the gap is corroborated by an exponent n that is consistently larger than two (inset). Reproduced from Pang *et al.*, 2018 (b) Temperature dependence of the in-plane thermal conductivity as κ_a/T of $CeCu_2Si_2$, which in the superconducting state ($H = 0$) extrapolates to zero at $T = 0$, clearly demonstrating fully gapped superconductivity. Also shown are the data in the normal state, which demonstrates the validity of the Wiedemann-Franz law. Reproduced from Yamashita *et al.*, 2017, available under a Creative Commons NonCommercial 4.0 International Public License.

Penetration depth measurements performed down to $T \approx 0.05$ K also demonstrate a fully open gap (Pang *et al.*, 2018; Takenaka *et al.*, 2017; Yamashita *et al.*, 2017). As shown in Fig. 11(a), the low temperature penetration depth shift $\Delta\lambda(T)$ is well described by the expression for a fully gapped material, with gap values well below that of BCS theory (in the range of $0.5 - 1k_BT_c$). Moreover, when analyzed using a power law dependence $\Delta\lambda(T) \sim T^n$ in temperature intervals with decreasing upper limits, the low temperature exponents are consistently found to increase with $n > 2$, exceeding the bounds expected for a line nodal superconductor of $n = 1$ and $n = 2$ in the clean and dirty limits, respectively.

Fully gapped superconductivity was also deduced from recent thermal conductivity measurements, where the coefficient of the in-plane thermal conductivity κ_a/T extrapolates to zero at zero temperature [Fig. 11(b)], again showing evidence for the lack of low-energy excitations expected for nodeless superconductivity (Yamashita *et al.*, 2017). This is further supported by measurements of the magnetic-field dependence of κ_a/T , where there is little change with applied field in the low field region. It is noted that more ambiguous results were found from earlier thermal conductivity studies of $CeCu_2Si_2$ (Vieyra, 2012), while the recent measurements reported by (Yamashita *et al.*, 2017) benefited from samples with lower non-superconducting fractions, as well as better contacts between the heater and the sample. Whereas earlier NQR measurements were found to exhibit a T^3 dependence of $1/T_1(T)$ down to around 0.1 K (Fujiwara *et al.*, 2008; Ishida *et al.*, 1999), more recent results show a deviation from this behavior at very low temperatures which can be accounted for by a small but nodeless gap (Kitagawa *et al.*, 2017).

While most recent low-temperature measurements have indicated that the superconducting gap is fully open, including also small-angle neutron scattering measurements (Campillo *et al.*, 2021), results from a low-temperature scanning spectroscopy study were less conclusive, which may be related to the fact that no good cleaves have been achieved in $CeCu_2Si_2$ until very recently (see Sec. IV.B). The tunneling spectra measured at low temperatures show two clear features at different voltage bias, providing clear evidence for multiple gaps or an anisotropic gap structure (Enayat *et al.*, 2016). However, the data were best accounted for by a model where the large gap is fully open, but the small gap is nodal. The reason for this discrepancy is not clear, but it should be noted that the density of states of the $d+d$ band mixing pairing state (Sec. IV.C) is linear for energies just above the small gap parameter, much like a line nodal superconductor, and therefore this could reconcile these results with other recent findings of nodeless superconductivity.

B. Fermi surface and quasiparticle dispersion

In order to unravel the electronic correlations and superconductivity in $CeCu_2Si_2$, the Fermi surface and quasiparticle dispersions close to E_F are crucial. While the Fermi surface of $CeCu_2Si_2$ has been predicted by a number of theoretical studies (Ikeda *et al.*, 2015; Li *et al.*, 2018; Luo *et al.*, 2020; Pourovskii *et al.*, 2014; Zwicknagl, 2016; Zwicknagl and Pulst, 1993), direct momentum-resolved measurements from angle-resolved photoemission spectroscopy (ARPES) are challenging due to the difficulty of sample cleavage (Reinert *et al.*, 2001). Recently, such experimental obstacles have been overcome due to an improved sample preparation method and a newly developed ARPES technique with a small beam spot (Wu *et al.*, 2021b). Figure 12 summarizes the ARPES results from a typical S-type single crystal. The

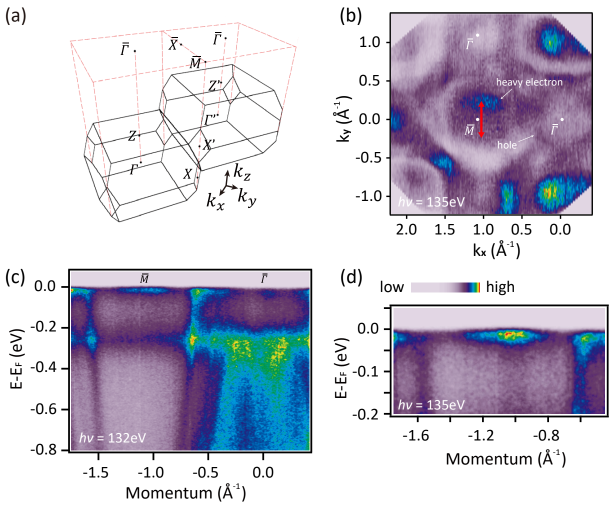

FIG. 12 The Fermi surface and quasiparticle dispersion of S-type CeCu$_2$Si$_2$ from ARPES measurements. (a) Three-dimensional bulk Brillouin zone (black solid lines) and the projected surface Brillouin zone (red dashed lines) of CeCu$_2$Si$_2$. (b) The experimental k_x-k_y map at 10 K taken with 135 eV photons. The red arrow indicates the in-plane component of the SDW ordering wave vector $\mathbf{Q_{AF}}$ observed by neutron diffraction (Sec. III.B). (c) Band dispersion along $\bar{\Gamma}$-$\bar{M}$ at 10 K. (d) Zoom-in view of the heavy electron band near E_F. Reproduced with permission from Wu *et al.*, 2021b.

experimental Fermi surface of CeCu$_2$Si$_2$ consists of three-dimensional hole bands centered at the bulk Z point [projecting onto the $\bar{\Gamma}$ point of the surface Brillouin zone] and a quasi-2D electron band at the X-point ($\bar{M}$ point at the surface Brillouin zone corner), see Figs. 12(a) and (b). Measurements of the energy-momentum dispersion show that the quasi-2D electron band is of predominant $4f$ character and possesses a large effective mass [Figs. 12(c) and (d)], while the hole bands near the $\bar{\Gamma}$ point are mainly derived from the lighter conduction bands.

The heavy electron band observed near the $\bar{M}$ point makes an important contribution to the Fermi surface and is crucial for the heavy-fermion superconductivity (Zwicknagl and Pulst, 1993). Photon-energy dependent scans and detailed analysis reveal that this heavy electron band is cylindrical in momentum space and has an effective mass of $\approx 120 m_e$. Here the effective mass is estimated by first dividing the experimental ARPES spectra by the (resolution-convoluted) Fermi-Dirac distribution function and then fitting the extracted quasiparticle dispersion with a parabola. Due to the limited energy resolution in ARPES, the effective mass estimation can have relatively large uncertainty. Note that the (zero temperature) effective mass used in the renormalized band calculation is $\approx 500 m_e$ (Zwicknagl, 2016; Zwicknagl and Pulst, 1993). Given that the ARPES was performed down to 10 K, at which temperature C_{4f}/T (≈ 0.125 J·mol^{-1}·K^{-2} [Fig. 1]) is approximately seven times smaller than in the low temperature limit (Steglich, 1990), the estimated effective masses indicate a very good correspondence between ARPES and the specific heat. As illustrated in Fig. 6(a), renormalized band calculations (Stockert *et al.*, 2004; Zwicknagl and Pulst, 1993) reveal that this heavy electron band has a warped part with flat parallel sides connected by a nesting vector $\boldsymbol{\tau}$, in excellent agreement with the SDW ordering wave vector $\mathbf{Q}_{\mathrm{AF}}$ observed in neutron diffraction [Figs. 6(a) and (b)] (Stockert *et al.*, 2004). The experimental contour of this heavy band shown in Fig. 12(b) is in fairly good agreement with these calculations. Another interesting observation is that the outer hole band near the $\bar{\Gamma}$ point contains appreciable $4f$ weight and bends slightly near E_F, which is the hallmark of hybridization between conduction and $4f$ electrons (Chen *et al.*, 2017; Im *et al.*, 2008; Jang *et al.*, 2020; Wu *et al.*, 2021a). Its enclosed area is close to the values obtained from quantum oscillation measurements (Hunt *et al.*, 1990; Tayama *et al.*, 2003), which however, could not detect the heavy electron band at the $\bar{M}$ point. Note that the detection of heavy bands can be particularly challenging in quantum oscillation experiments, due to the rapid decay of the quantum oscillation amplitudes with temperature for heavy orbits (Shoenberg, 2009).

C. $d + d$ matrix-pairing state

As discussed in Sec. III.F, the majority of the experiments in superconducting CeCu$_2$Si$_2$ have been interpreted in terms of a single-band, d-wave Cooper pairing. The new results presented in Sec. IV.A point toward the emergence of a full gap. Although a single-band d-wave pairing state is at odds with the more recent results, the underlying sign-changing nature of the pairing state under a C_{4z} rotation continues to play an important role. This is best illustrated by the large peak in the INS intensity in the superconducting state [Fig. 7(a)] associated with a pairing state which changes sign *within* the heavy, cylindrical bands near the edge of the Brillouin zone, as illustrated in Fig. 13.

The robustness of the sign-changing nature of the pairing states suggests that new pairing candidates have to reconcile this feature with the emergence of a full gap. An important requirement is that the sign-changing but also gapped pairing state must belong to a single irreducible representation of the point group. Indeed, unlike systems such as UPt$_3$ (Fisher *et al.*, 1989; Schemm *et al.*, 2014), there have been no reports of multiple superconducting transitions in CeCu$_2$Si$_2$ which further break symmetry with decreasing temperature. Similarly, the lack of evidence for time-reversal symmetry breaking in the superconducting state makes $d + id$ or $s + id$ pairing states unlikely, since these are gapped and sign-changing but only at the price of breaking both point-group and

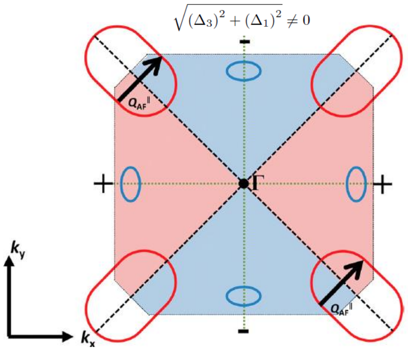

FIG. 13 Projection of the renormalized heavy Fermi surface and ordering wave vector $\mathbf{Q}_{\mathrm{AF}} = (0.215, 0.215, 0.53)$ onto the $k_z = 0$ plane of the 3D Brillouin zone at zero-temperature. The dashed lines indicate the nodes of the individual components of $d + d$ pairing. The diagonal black and vertical/horizontal green dashed lines denote the nodes of $\Delta_3(\mathbf{k}) \propto d_{x^2-y^2}$ and $\Delta_1(\mathbf{k}) \propto d_{xy}$, respectively (see Eq. 1). The effective gap is determined by the addition in quadrature of the two components. Since the nodes of the Δ_3 and Δ_1 components do not overlap except at isolated points of the Brillouin zone, $d + d$ pairing is always gapped on the Fermi surface. The wave vector for the peak of the observed antiferromagnetic fluctuations projected onto the (k_x, k_y) plane, $\mathbf{Q}^{||}_{AF}$, connects parts of the cylindrical Fermi surface near the edges (red pill-shapes) where $\Delta_3 \propto d_{x^2-y^2}$ has opposite signs, leading to the emergence of a pronounced peak inside the superconducting gap in INS experiments. Reproduced from Pang *et al.*, 2018.

time-reversal symmetries. We instead consider a pairing state that can reconcile the features of superconducting $\mathrm{CeCu_2Si_2}$ while preserving the symmetries already mentioned. This is a multi-band $d + d$ pairing of concurrent intra-band $d_{x^2-y^2}$- and inter-band d_{xy}-waves (Nica and Si, 2021; Nica *et al.*, 2017). In it's most general form, $d + d$ pairing is

$$\Delta_{d+d} = \begin{pmatrix} \Delta_3(\mathbf{k}) & \Delta_1(\mathbf{k}) \\ \Delta_1(\mathbf{k}) & -\Delta_3(\mathbf{k}) \end{pmatrix}, \tag{1}$$

where the intra- and inter-band components Δ_3 and Δ_1 transform as $d_{x^2-y^2}$ and d_{xy}, respectively. This *matrix-pairing* state, which is intrinsically multi-band, has additional structure due to the band space on which it is defined. The intra-band $d_{x^2-y^2}$ component naturally satisfies the required sign-change, much like a single-band d-wave pairing. In contrast to the latter, the matrix structure of $d + d$ pairing, due to the anti-commuting Pauli matrices, also ensures that the gap is determined by the addition in quadrature of the two distinct d-wave components. Consequently, the Bogoliubov-de Gennes (BdG) quasiparticle spectrum shows a full gap everywhere on the Fermi surface. As recently discussed in (Nica and Si, 2021), $d + d$ pairing is a natural d-wave analogue to the spin-triplet pairing states of ^{3}He-B, with the bands playing a role similar to the spin as far as the matrix structure is concerned in the former and latter cases, respectively. The $d + d$ pairing yields good fits to penetration depth, specific heat, and NQR data well below and closer to T_c alike (Pang *et al.*, 2018; Smidman *et al.*, 2018), as discussed in the following subsection.

While $d + d$ pairing defined in the band basis provides a direct interpretation of the experimental results, its stability is more naturally addressed using microscopic matrix-pairing candidates defined in the orbital/spin space of the paired electrons. Matrix-pairing states which transform according to the irreducible representations of the point group can be constructed from the decomposition of the products of two-orbital, or more generally, spin-orbit coupled multiplets of definite symmetry. This approach was illustrated in the alkaline Fe-selenides, which are also strongly-correlated multi-band superconductors. [The properties of other Fe-selenide superconductors with a similar or higher T_c as the alkaline Fe-selenides, including the Li-intercalated iron selenides (Lu *et al.*, 2015) and even the single-layer FeSe, the T_c record holder of the iron-based superconductors (Wang *et al.*, 2012a), are similar (Si *et al.*, 2016).] In spite of the difference in the nature of their basic constituents, these Fe-based superconductors remarkably share some of the experimental signatures that are similar to those in $\mathrm{CeCu_2Si_2}$, namely, fully-gapped superconductivity, as indicated by ARPES experiments (Mou *et al.*, 2011; Wang *et al.*, 2011, 2012b; Xu *et al.*, 2012), which supports a spin resonance in the INS spectrum (Friemel *et al.*, 2012; Park *et al.*, 2011). The wave vector of the resonance at $\mathbf{Q}_{\text{Alkaline FeSe}} = (0.5, 0.25, 0.5)$ (Friemel *et al.*, 2012; Park *et al.*, 2011) is distinct from what one could expect from the sign-changing s-wave pairing which, in any case, is unlikely given the absence of hole pockets at the center of the Brillouin zone. Nica *et al.*, 2017 introduced an $s\tau_3$ matrix-pairing state, which consists of an s-wave form factor multiplied by a τ_3 Pauli matrix in the space of the Fe $d_{xz/yz}$ orbitals. Because the $s\tau_3$ matrix does not commute with the symmetry-dictated kinetic part, the multi-orbital $s\tau_3$ pairing is equivalent to $d + d$ pairing in the band basis (Nica and Si, 2021; Nica *et al.*, 2017). On the other hand, $s\tau_3$ transforms as a single B_{1g} irreducible representation of the D_{4h} point group. This implies that $d + d$ pairing also belongs to the same representation and that it preserves both point-group and time-reversal symmetries. When the normal-state band splitting near the Fermi level is small, the BdG quasiparticle spectrum shows a full gap everywhere in the Brillouin zone. Generically, the BdG spectrum is always nodeless everywhere on the Fermi surface. Away from the Fermi surface, nodes can occur in the BdG spectrum when the band splitting exceeds a certain threshold. However, in strongly correlated systems only nodal excitations on the Fermi surface are long lived and, correspondingly, sharply defined; any putative nodal excitations away from the Fermi surface involve a large correlation-induced damping in the normal state, and the distinction between nodal and gapped excitations is obviated (Nica and Si, 2021; Nica *et al.*, 2017). Finally, we note that the $s\tau_3$ and the equivalent $d+d$ pairings are energetically favored: they are stabilized in a multi-orbital $t-J_1-J_2$ model (Nica *et al.*, 2017) in the regime where A_{1g} and B_{1g} pairing channels are quasi-degenerate.

Following the important precedent of the alkaline Fe-selenides, (Nica and Si, 2021) constructed a microscopic candidate for even-parity, spin-singlet $d+d$ pairing which incorporates the nature of the electronic states in $CeCu_2Si_2$. Matrix-pairing candidates can be constructed within the quasi-localized f electron sector, corresponding to $f-f$ pairing, but also in the $f-c$ and $c-c$ sectors, where c stands for a conduction electron. As indicated by several experiments (Amorese *et al.*, 2020; Goremychkin and Osborn, 1993; Rueff *et al.*, 2015) and by LDA+DMFT studies (Pourovskii *et al.*, 2014), the $^2F_{5/2}$ electron states split under the influence of the crystalline-electric-field into a ground-state Γ_7 Kramers doublet and excited Γ_6 and Γ_7 doublets. Within the $f-f$ pairing sector, the product of two ground-state Γ_7 doublets decomposes into Γ_1, Γ_2 and Γ_5 irreducible representations. As previously discussed, $CeCu_2Si_2$ does not show signs of multiple superconducting transitions, implying that two-component pairing states belonging to Γ_5 are unlikely to occur. From the remaining two representations, the matrix associated with Γ_2 is symmetric and thus incompatible with the even-parity, spin-singlet nature of the pairing candidate. The only possible pairing candidate within the $f-f$ sector is a matrix belonging to the identity Γ_1 representation. Because this matrix transforms trivially under the point group, the symmetry of $f-f$ pairing states are determined entirely by the form factor. This implies that $f-f$ pairing is not likely to support $d+d$ pairing. Nica and Si, 2021 considered an alternative in the $f-c$ pairing sector.

Conduction electron states which belong to the Γ_6 irreducible representation can be constructed by first taking linear combinations of the Cu $d_{x^2-y^2}$ orbitals which transform as (p_x, p_y) within each unit cell:

$$p_x = d^{(4)}_{x^2-y^2} - d^{(2)}_{x^2-y^2} \tag{2}$$

$$p_y = d^{(1)}_{x^2-y^2} - d^{(3)}_{x^2-y^2}, \tag{3}$$

as illustrated in Fig. 14. The spin-orbit coupling can be incorporated to obtain the Γ_6 states

$$\Psi_{\Gamma_6;1/2} = \frac{i}{2}\left[p_x + \mathrm{i}p_y\right]\phi_{-1/2} \tag{4}$$

$$\Psi_{\Gamma_6;-1/2} = \frac{i}{2}\left[p_x - \mathrm{i}p_y\right]\phi_{1/2}, \tag{5}$$

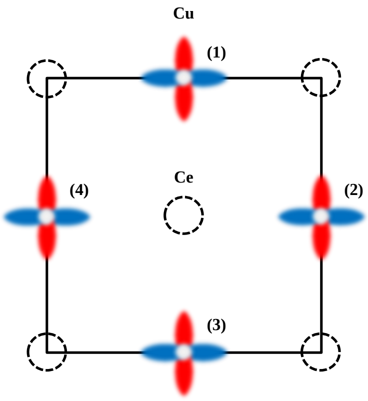

FIG. 14 Single Cu plane in the unit cell of $CeCu_2Si_2$. The four sites labeled $(1)-(4)$ correspond to Cu $d_{x^2-y^2}$ orbitals in the plane. The dashed-line circles represent the Ce sites projected onto the Cu-plane. Reproduced from Nica and Si, 2021, under a Creative Commons Attribution 4.0 International License.

where the ϕ's denote spin-1/2 states. Note that the four d orbitals are localized on distinct sites in the unit cell. The Ψ states are examples of a Zhang-Rice construction (Zhang and Rice, 1988). The decomposition of the products of f-electron Γ_7 doublets, belonging to the ground-state multiplet, and Γ_6 conduction electron doublets includes a sign-changing Γ_3 irreducible representation. When multiplied by a featureless s-wave form factor, the matrix associated with Γ_3 $f-c$ pairing is the analogue of the $s\tau_3$ pairing introduced in the context of the alkaline Fe-selenides. $s\Gamma_3$ thus provides a microscopic candidate for $d+d$ pairing in $CeCu_2Si_2$. Evidence supporting this type of pairing was provided by x-ray absorption spectroscopy experiments (Amorese *et al.*, 2020) which indicated a finite admixture of the f-electron Γ_6 in the ground-state of $CeCu_2Si_2$. It is important to recall that the microscopic candidate for $d+d$ pairing introduced in (Nica and Si, 2021) was constructed using only the point-group symmetry and a minimal input provided by the nature of the lowest-energy $4f$ Kramers doublet. In spite of its simplicity, this construction (i) demonstrates how $d+d$ pairing can emerge in principle, and (ii) provides a well-defined microscopic candidate for any future detailed theoretical studies of the pairing symmetry in $CeCu_2Si_2$ that also incorporate the complex bandstructure of the normal state.

Sign-changing s_{+-} pairing states were also advanced to explain the gapped, sign-changing superconductivity in $CeCu_2Si_2$ (Ikeda *et al.*, 2015; Li *et al.*, 2018). We briefly summarize two of the most important differences between $d+d$ and s_{+-} pairing states. Firstly, although both candidates are sign-changing and therefore conducive to a large peak in the INS intensity inside the superconducting gap, they also imply very different ways of involving the states on the Fermi surface. Li *et al.*, 2018 (see also Ikeda *et al.*, 2015) carried out DFT+U calculations, which capture neither the Kondo effect nor the associated renormalization towards heavy single-electron excitations. Physically, the proposed s_{+-} picture invokes a wave vector that spans the distance between the heavy cylindrical Fermi surface (red pockets in Fig. 13) and the hole pocket near the Z-point (bulk Brillouin zone) projected from light bands (not shown), which does not generate enough spin spectral weight for either the observed antiferromagnetic order or the observed INS spectrum in the superconducting state. A lack of such extended nesting between these different surfaces can also be inferred experimentally from the ARPES results (Sec. IV.B) due to the electron and hole pockets being observed to have very different shapes and effective masses. In contrast, the $d+d$ pairing implies a wave vector spanning within the same cylindrical heavy Fermi surface (red pockets in Fig. 13). The latter picture is naturally associated with a realistic heavy-fermion SDW instability, due to the enhanced density-of-states on these pockets. Secondly, $d+d$ and s_{+-} pairing states have distinct nodal structures. As already discussed, the $d+d$ pairing state has no nodes on the Fermi surface (see Fig. 13). By contrast, the s_{+-} pairing state has gap zeroes that would generally be expected to intersect the extended hole Fermi surface of $\mathrm{CeCu_2Si_2}$, leading to nodal excitations. This is different from the case of Fe-based superconductors, which typically have disconnected hole and electron pockets at the zone center and edges. These points imply that the s_{+-} picture is not viable.

We conclude this section by briefly revisiting the effects of disorder on the paired states in $\mathrm{CeCu_2Si_2}$. As mentioned in Sec. III.E, the weak suppression of T_c in electron-irradiated samples was argued to point towards a more conventional order-parameter that does not change sign (Takenaka *et al.*, 2017; Yamashita *et al.*, 2017), an interpretation which usually relies on the perturbative Abrikosov-Gor'kov theory. However, d-wave pairing in strongly correlated settings is expected to be much less sensitive to disorder introduced via non-magnetic potential scattering (Anderson, 1997). Studies in models with strong, short-range exchange interactions are consistent with this expectation (Chakraborty *et al.*, 2017; Garg *et al.*, 2008). This implies that $d+d$ pairing states are also robust against this type of disorder in a broader class of materials with similar strong exchange interactions. These include for instance the alkaline Fe-selenides, where the $d+d$ state in the form of $s\tau_3$ pairing was stabilized in a multi-orbital $t-J_1-J_2$ model (Nica and Si, 2021). We expect that strong correlations also protect the $d+d$ pairing state in $\mathrm{CeCu_2Si_2}$. In contrast, as already mentioned above, T_c can be sharply suppressed in $\mathrm{CeCu_2Si_2}$ via atomic substitution, as is the case for instance in high-T_c superconductors with Zn substituted for Cu on the $\mathrm{CuO_2}$ planes (Loram *et al.*, 1990).

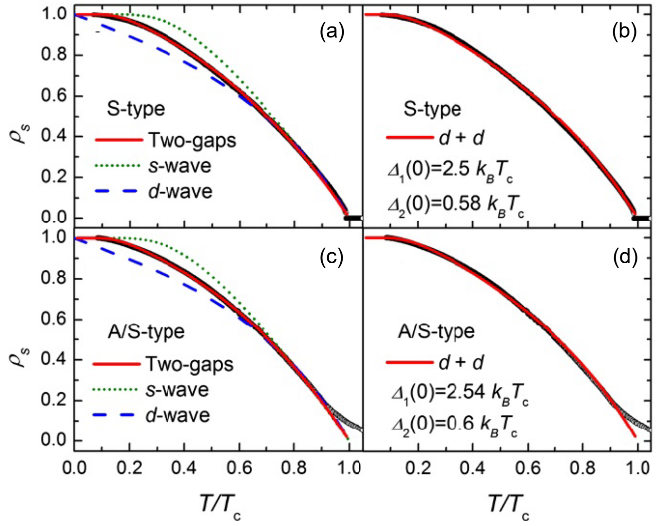

FIG. 15 Temperature dependence of the superfluid density derived from penetration depth measurements using the tunnel-diode-oscillator-based method from. Panels (a) and (b) display fits to the superfluid density of an S-type sample with an isotropic two gap model and $d+d$ band-mixing pairing model, respectively, while (c) and (d) show the corresponding results for the A/S-type sample. Reproduced from Pang *et al.*, 2018.

D. Analysis of experimental results with the $d+d$ model

Upon converting the penetration depth data measured using the tunnel-diode-oscillator-based method to the superfluid density, Pang *et al.* found that the temperature dependence could be described both by an isotropic two-gap model, as well as one for the $d+d$ band mixing pairing state (Pang *et al.*, 2018; Smidman *et al.*, 2018), which is displayed in Fig. 15. For the latter case, a simple model of the gap function is given by $\Delta(T,\phi) = [(\Delta_1(T)\cos2\phi)^2 + (\Delta_2(T)\sin2\phi)^2]^{\frac{1}{2}}$, which has a four-fold oscillatory component where one of the gap parameters corresponds to the gap minimum and the other to the maximum. The basis for applying this model is explained in the previous subsection, and it is found that this describes the data across the whole temperature range well. The fitted values of the gap parameters for measurements of the S-type sample were $2.5k_\mathrm{B}T_\mathrm{c}$ and $0.58k_\mathrm{B}T_\mathrm{c}$, where the small but finite gap minimum ensures a nodeless gap across the Fermi surface, and is close to the magnitude obtained from the low temperature analysis of $\Delta\lambda(T)$ (Sec. IV.A). This model can also fit the temperature dependence of the specific heat (Pang *et al.*, 2018; Smidman *et al.*, 2018), including the data previously reported by Kittaka *et al.* (Kittaka *et al.*, 2014).

In the case of the recent NQR measurements, this $d+d$ band mixing pairing model can well account for $1/T_1(T)$ across the whole temperature range, including the deviation from T^3 behavior resolved at the lowest temperatures from recent measurements (Kitagawa *et al.*, 2017; Smidman *et al.*, 2018). Although the simple two-band BCS model can also describe the low temperature $1/T_1(T)$ results, it is less accurate at elevated temperatures, where it deviates from the data, culminating in the prediction of a pronounced Hebel-Slichter coherence peak below T_c. Such an enhancement, which is a hallmark of conventional BCS superconductivity, is absent from the data (Fig. 9), which is in-line with a sign-changing order parameter. In the s_{+-} scenario this peak is somewhat suppressed, but still present in the model. In analogy with the Fe-based superconductors, effects such as quasiparticle damping and impurity-induced bound states (in the case of s_{+-} pairing) could potentially account for the deviations from these two models (Bang and Stewart, 2017). On the other hand, for a $d + d$ band mixing pairing state the coherence peak is naturally avoided due to the sign change of the intraband pairing component (Kitagawa *et al.*, 2017; Smidman *et al.*, 2018).

V. PERSPECTIVES

Despite the progress made on this prototypical heavy-fermion superconductor, a number of points are worthy of further investigations.

Although the band-mixing $d + d$-pairing state can account for all the experimental results, more direct experimental evidence for such a scenario is still lacking. To unambiguously discriminate between different fully gapped models likely requires high-resolution momentum resolved experimental probes of the superconducting gap at very low temperatures, which is very challenging. In addition, while recent proposals have given a microscopic basis to the $d + d$-pairing state (Nica and Si, 2021), a fully developed microscopic theory for $CeCu_2Si_2$ is still necessary. Indeed, developing fully microscopic theories for strongly correlated superconductors remains a grand challenge of condensed matter physics.

The effect of nonmagnetic potential scattering on $CeCu_2Si_2$ still lacks a complete theoretical and experimental understanding. This is especially so concerning the variable sensitivity of the superconductivity to substitutional disorder, which appears to be site-dependent, as well as to various types of lattice rearrangement, such as that induced by electron irradiation. There is a pronounced size dependence for substitutions on the Ce-site, where the magnitude of the T_c-depression is found to be *anticorrelated* to the volume of the so obtained “Kondo hole”, while Ge atoms exchanged for Si are less strong pairbreakers. The origin of this nonuniversal impact of substitutional disorder on the superconductivity of $CeCu_2Si_2$, as well as the dichotomy between “harmful” and “harmless” (e.g., electron-irradiation-induced) disorder are interesting open questions to be unraveled by future work.

We also note that $CeCu_2Si_2$ has often been regarded as a prototypical example of both heavy-fermion superconductivity, as well as SDW-type quantum criticality, but the extent to which the findings extend to other heavy-fermion systems is currently unclear. In particular, the nodeless superconducting gap structure of $CeCu_2Si_2$ is distinct from the clearly evidenced nodal $d_{x^2-y^2}$ superconductivity in Ce(Co,Ir)In_5 (Allan *et al.*, 2013; An *et al.*, 2010; Izawa *et al.*, 2001; Kasahara *et al.*, 2008; Lu *et al.*, 2012; Park *et al.*, 2008; Zhou *et al.*, 2013).

The spin-excitation spectrum of $CeCu_2Si_2$ consists of both long-wavelength SDW-type fluctuations (paramagnons), as well as high-frequency Mott-like fluctuations of $4f$ electron spins. It is of special interest to understand the role played by these different types of spin fluctuations in either promoting or breaking apart the Cooper pairs. In this context, it is interesting that the $B_{c2}(p)$ curve in the $T = 0$ plane of Fig. 4(a) exhibits its maximum at a pressure which is only about half the value of the critical pressure p_c at $B = 0$. When increasing the pressure far away from this QCP, in the absence of quantum-critical SDW fluctuations, $B_{c2}(p)$ is found to increase which apparently means that superconductivity becomes strengthened. However, when further approaching the QCP (at $p_c/2 < p < p_c$), under increasingly dominant SDW-type quantum critical fluctuations, $B_{c2}(p)$ turns out to decrease and superconductivity deteriorates. A similar conclusion can be drawn from the evolution of $T_c(p)$ for the low-pressure dome displayed in Fig. 5 and may also apply to other correlated metals showing a superconducting dome centered at an SDW- or putative SDW- type QCP, such as $Ba(Fe_{1-x}Co_x)_2As_2$ (Chu *et al.*, 2009) or $CePd_2Si_2$ (Mathur *et al.*, 1998). This non-monotonic evolution suggests that the Mott-type critical excitations are pair-promoting, while the ultra-low-temperature (below $T^* = 1$ K) SDW-type critical excitations in $CeCu_2Si_2$ are pair-breaking. Interestingly, the theoretical work of Hu *et al.*, 2021a, for an SDW-type quantum criticality of Kondo-lattice systems, reached a similar conclusion that the Mott-type quantum critical fluctuations at energies above T^* are primarily instrumental for the Cooper-pair formation. Together, these considerations suggest that the heavy-fermion superconductivity in $CeCu_2Si_2$ should be compared with those of systems with local (Kondo-destroying) rather than itinerant (SDW-type) QCPs (Nguyen *et al.*, 2021; Park *et al.*, 2006; Schuberth *et al.*, 2016, 2022; Shan *et al.*, 2022; Shishido *et al.*, 2005).

VI. SUMMARY

$CeCu_2Si_2$ was originally considered a prototypical intermediate-valence metal (Sales and Viswanathan, 1976). The discovery of heavy-fermion behavior (Steglich *et al.*, 1979) in this compound led to the notion that it belongs to the family of Ce-based Kondo-lattice systems (Bredl *et al.*, 1984, 1978) and, most importantly, $CeCu_2Si_2$ is the first discovered unconventional superconductor (Steglich *et al.*, 1979). Over 40 years of intense research on this system have posed several severe challenges and surprising solutions most of which are covered in this article. In the following, we briefly summarize our current knowledge on $CeCu_2Si_2$.

Its Kondo-lattice ground state, implying a local $J = 5/2$ spin-orbit split Hund's rule multiplet of trivalent Ce, which is further split by the tetragonal crystalline-electric field into two Γ_7 and a Γ_6 Kramers doublets (Amorese *et al.*, 2020), could be recently verified by ARPES experiments performed at 10 K (Wu *et al.*, 2021b), well below the lattice Kondo temperature of 15 K. These investigations revealed a 'large (renormalized) Fermi surface' to which the Ce-$4f$ electrons substantially contribute, i.e., a heavy electron band near the X-point of the bulk Brillouin zone. For this heavy band the effective charge-carrier mass m^* estimated from ARPES of $m^* \approx 120\,m_{\rm e}$ is in very good agreement with that obtained from specific-heat results at the same temperature (Fig. 1) (Steglich, 1990). In addition, ARPES revealed a hole band with small, but significant $4f$-contribution near the bulk Z-point which corresponds to the distinct Fermi surface pocket with moderately enhanced $m^* (\approx 5\,m_{\rm e})$ that had been detected by magnetic quantum oscillation measurements (Hunt *et al.*, 1990; Tayama *et al.*, 2003). In contrast to the aforementioned ground-state and thermodynamic properties which probe the large Fermi surface of the Kondo-lattice state of $CeCu_2Si_2$ at finite temperatures, transport measurements (Shan *et al.*, 2022; Sun and Steglich, 2013) appear to be dominated down to very low temperatures by the fundamental local scattering process underlying the Kondo screening, i.e., scattering of ordinary conduction electrons from the Ce-derived localized $4f$-spins, see also (Coleman *et al.*, 1985). Upon volume compression, Ce-based Kondo-lattice systems commonly show a strengthening of the Kondo interaction and eventually a transition into an intermediate-valence state. This has been observed for $CeCu_2Si_2$ as well (Holmes *et al.*, 2004; Yuan *et al.*, 2003, 2006).

One of the characteristics of these types of materials is their closeness to magnetism. Many of them exhibit a magnetically ordered low-temperature phase in the vicinity of a QCP. While the discovery of superconductivity in $CeCu_2Si_2$ with a finite magnetic moment in each unit cell came as a big surprise for most researchers in the field of superconductivity, this might indeed have been expected for researchers working on superfluid ^{3}He (Vollhardt and Wölfle, 1990). With the discovery of a heavy-fermion low-temperature phase in $CeAl_3$ (Andres *et al.*, 1975), which resembles the renormalized normal phase of (charge-neutral) liquid ^{3}He at sufficiently low temperatures, the question might have arisen: Is there a superconducting analogue in a heavy-fermion metal like $CeAl_3$ to the superfluid phases in ^{3}He? Not surprisingly, magnetically-driven superconductivity in heavy-fermion metals was proposed quite early by theorists (Anderson, 1984; Miyake *et al.*, 1986; Scalapino *et al.*, 1986) and was then gradually verified experimentally (Aeppli *et al.*, 1989; Sato *et al.*, 2001). In the case of $CeCu_2Si_2$, it became clear from the outset that a BCS-type phonon-mediated Cooper-pairing mechanism is incapable of explaining why the non-magnetic analogue compound $LaCu_2Si_2$ is not a superconductor (Steglich *et al.*, 1979) as well as the drastic pair-breaking effect of certain non-magnetic impurities, notably when substituted for Cu in $CeCu_2Si_2$ (Spille *et al.*, 1983).

In more recent years, $CeCu_2Si_2$, along with $CeCu_{6-x}Au_x$, $YbRh_2Si_2$ and $CeRhIn_5$, have played a prominent role in the understanding of heavy-fermion quantum criticality (Gegenwart *et al.*, 2008). Theoretical studies of Kondo-lattice models have led to the notion of Kondo destruction (Coleman *et al.*, 2001; Si *et al.*, 2001), which characterizes Mott-type quantum criticality for an electron localization-delocalization transition. More recently, it has been argued that partial Mott quantum criticality also forms the basis for the ferromagnetic instabilities in the heavy-fermion metals $YbNi_4(P_{1-x}As_x)_2$ (Steppke *et al.*, 2013) and $CeRh_6Ge_4$ (Shen *et al.*, 2020). In $CeCu_2Si_2$, it has been suggested that SDW-type critical excitations operate below an energy scale of T^* that is nonzero but much smaller than the Kondo temperature, while the Mott-type critical excitations describe the quantum criticality above this energy scale (Gegenwart *et al.*, 2008; Smidman *et al.*, 2018). Theoretical studies that incorporate the Kondo destruction physics in quantum-criticality-driven superconductivity have recently been developed (Hu *et al.*, 2021a).

In the low-temperature normal state of S-type $CeCu_2Si_2$, the critical exponent of the power-law T-dependence of the resistivity turned out to be ambiguous, i.e. 1.5 (Gegenwart *et al.*, 1998) or 1 (Yuan *et al.*, 2003, 2006), presumably due to the spatial distribution of a magnetically ordered minority phase (Stockert *et al.*, 2011) that may modifiy the volume-integrated response in resistivity experiments. From the temperature dependences of both $C(T)/T$ and the damping rate measured in the INS spectrum (Arndt *et al.*, 2011; Gegenwart *et al.*, 2008; Smidman *et al.*, 2018), $T^* \sim 1 - 2$ K can be inferred, which is on the same order of magnitude as the spin excitation gap in the magnetic response in the superconducting state. Nevertheless, the linear paramagnon dispersion relation observed above the spin-gap energy $\hbar\omega_{\rm gap}$ extends to about 1.5 meV (Song *et al.*, 2021). Except for these paramagnon excitations, the magnetic INS response comprises of Mott-type fluctuations of local Ce-moments with frequencies in the range $k_B T^*/\hbar$ to $k_B T_K/\hbar$. The existence of a nonzero Kondo-destruction energy scale $k_B T^*$ that is small compared to the Kondo temperature has also been inferred from the large kinetic energy loss as $CeCu_2Si_2$ goes from the normal to the superconducting state; this kinetic energy loss overcompensates the majority of the exchange energy saving in the same process (Stockert *et al.*, 2011). This overcompensation results in a pair-formation energy that is smaller than the exchange energy by a factor of about twenty - characteristic of magnetically-driven Cooper pairing of slowly propagating Kondo singlets (Stockert *et al.*, 2011). As far as the magnetism in $CeCu_2Si_2$ is concerned, the nature of the high-field B-phase (Bruls *et al.*, 1994), and of its QCP at about 17 T (Weickert *et al.*, 2018) as well as the first-order phase transition between this B-phase and the adjacent low-field SDW A-phase (Tayama *et al.*, 2003) need further detailed exploration. Meanwhile the field dependence of the specific heat in the superconducting state exhibits an unusual upturn at intermediate fields culminating in a strongly enhanced value just below B_{c2} (Kittaka *et al.*, 2014, 2016). The origin of this behavior still needs to be determined, especially whether it is related to a spatially modulated superconductivity (Kitagawa *et al.*, 2018) or other inferred effects of strong Pauli-paramagnetic limiting (Campillo *et al.*, 2021).

Another striking phenomenon is the occurrence of a second superconducting dome in $CeCu_2Si_2$ at pressures well above the critical pressure at which SDW order disappears. There, the T_c is around three times larger than in low pressure conditions (Yuan *et al.*, 2003, 2006). Although such a scenario was hinted at by the unusual shape of the T_c vs p plateau, the existence of a distinct second high pressure dome was only apparent upon doping with Ge to weaken the superconductivity (Fig. 5), and suggests a different unconventional pairing mechanism at higher pressures, namely one related to valence fluctuations (Holmes *et al.*, 2004; Yuan *et al.*, 2003).

For a long time, $CeCu_2Si_2$ was believed to be a single-band d-wave superconductor with line nodes in the energy gap. The strongest evidence for this conclusion came from NQR measurements down to 0.1 K, which revealed the absence of a Hebel-Slichter peak at T_c and a T^3 dependence of $1/T_1(T)$ (Fujiwara *et al.*, 2008; Ishida *et al.*, 1999). A $d_{x^2-y^2}$ state was concluded from INS (Eremin *et al.*, 2008; Stockert *et al.*, 2011), while d_{xy} was deduced from the anisotropy of the upper critical field determined from the resistivity (Vieyra *et al.*, 2011). This understanding was overturned by the results of low-temperature specific-heat (Kittaka *et al.*, 2014, 2016), penetration depth (Pang *et al.*, 2018; Takenaka *et al.*, 2017; Yamashita *et al.*, 2017), thermal conductivity (Yamashita *et al.*, 2017), and more recent NQR measurements on $CeCu_2Si_2$ single crystals (Kitagawa *et al.*, 2017) which reveal a small, but finite fully open superconducting gap. Theoretical proposals to account for these findings include both isotropic (non-sign-changing) (Takenaka *et al.*, 2017; Yamashita *et al.*, 2017) and anisotropic (sign-changing) s-wave pairings (Ikeda *et al.*, 2015; Li *et al.*, 2018), as well as a $d+d$ matrix pairing state (Nica and Si, 2021; Nica *et al.*, 2017; Pang *et al.*, 2018)(see Table II).

The aforementioned s-wave pairings are disfavored for the following reasons:

(i) As discussed in Sec. III.D, a pronounced maximum is observed in the INS intensity inside the superconducting gap, exactly at the SDW ordering wave vector $\mathbf{Q}_{\mathrm{AF}}$. The latter equals the nesting vector $\boldsymbol{\tau}$ inside the warped part of the cylindrical heavy-electron band at the X-point of the bulk Brillouin zone (Smidman *et al.*, 2018; Wu *et al.*, 2021b). This maximum demonstrates a sign-change of the superconducting order parameter along $\boldsymbol{\tau}$, which means intraband pairing as already discussed. No such sign change is possible for isotropic BCS-type pairing. In addition, such onsite pairing is unfavorable in a heavy-fermion superconductor as the heavy charge carriers forming the Cooper pairs only have a tiny kinetic energy of order $k_{\mathrm{B}}T_{\mathrm{K}}$, which is of the same order as their renormalized Coulomb repulsion. For an onsite pairing to operate in a BCS superconductor, the kinetic energy is much larger than the effective Coulomb repulsion. In an innovative approach, (Tazai and Kontani, 2018, 2019) succeeded in showing that both phonon-mediated and electronically-driven s-wave heavy fermion superconductivity can arise from higher multipole charge fluctuations. However, the *magnetically-driven* nature of the superconductivity in $CeCu_2Si_2$ (Stockert *et al.*, 2011) necessitates sign-changing superconductivity (Scalapino, 2012). More generally, such an s-wave pairing without a sign change is difficult to be reconciled with the exclusion of onsite pairing associated with the strong Coulomb repulsion of the $4f$ electrons.

(ii) Anisotropic s-wave pairing also cannot explain the superconductivity of $CeCu_2Si_2$. In order to account for the pronounced peak observed in INS at $\mathbf{Q}_{\mathrm{AF}}$ inside the superconducting gap, there would need to be *interband* nesting connected by the SDW ordering wave vector, whereas ARPES measurements (Wu *et al.*, 2021b) and calculations of the renormalized electronic structure (Zwicknagl, 1992; Zwicknagl and Pulst, 1993) demonstrate that this ordering wave vector must connect regions within the heavy electron pocket, as indeed revealed by neutron diffraction (Stockert *et al.*, 2004), see Fig.6(b). This confirms that there is a sign change of the order parameter within this band, in contrast to the s_{+-} scenario where the sign changes between the hole and electron pockets (Li *et al.*, 2018) (see also Ikeda *et al.*, 2015).

A $d+d$ pairing state with intra and interband components provides a natural resolution to all currently available experimental results, and is in line with the importance of the non-perturbative effect of the strong Coulomb repulsion of the $4f$-electrons in the form of a Kondo effect. The intraband d-wave component accounts for the sign change on the heavy warped cylindrical bands. The two distinct components added in quadrature also ensure a fully gapped Fermi surface. This pairing state belongs to a single irreducible representation of the point group, which coincides with that of a single-band d-wave, and therefore implies a single transition to the superconducting phase, as observed in $\mathrm{CeCu_2Si_2}$. On the microscopic level, $d+d$ pairing is equivalent to a matrix-pairing state between f-electrons in Γ_7 doublets and conduction electrons belonging to Γ_6 doublets. The non-trivial matrix structure ensures the presence of the two d-wave components in the band basis. Similar $d+d$ candidates were proposed in the context of the alkaline Fe-selenides (Nica and Si, 2021; Nica *et al.*, 2017), suggesting a common theme in unconventional superconductivity. Nevertheless, in line with other classes of unconventional superconductors, the unambiguous determination of the pairing state and mechanisms of $\mathrm{CeCu_2Si_2}$ still requires a fully developed microscopic theory together with additional experimental results able to discriminate between different scenarios.

Taking all these together, $\mathrm{CeCu_2Si_2}$, the very first unconventional superconductor ever discovered, continues to grow in its role as a model system for strong correlation physics. The historical intuition about $\mathrm{CeCu_2Si_2}$ as a solid-state generalization of the superfluidity observed in liquid ^{3}He inspired the early considerations regarding the interplay between antiferromagnetic correlations and d-wave superconductivity. The observation that the Cooper pairs in $\mathrm{CeCu_2Si_2}$ are formed by the extremely heavy charge carriers existing in the low-temperature phase of the Kondo lattice proved the superconducting pairing mechanism to be incompatible with the conventional one of BCS theory. In modern times, $\mathrm{CeCu_2Si_2}$, like $\mathrm{CeCu_{6-x}Au_x}$ (Löhneysen *et al.*, 1994; Schröder *et al.*, 2000), $\mathrm{CePd_2Si_2}$ (Mathur *et al.*, 1998), $\mathrm{CeCoIn_5}$ (Paglione *et al.*, 2003), and $\mathrm{CeRhIn_5}$ (Park *et al.*, 2006; Shishido *et al.*, 2005), has served as a model system for heavy fermion antiferromagnetic quantum criticality. Intriguingly, here, the Landau-type SDW quantum criticality interplays with the beyond-Landau Mott-type quantum criticality in different energy ranges below the Kondo temperature. Moving further to just over the past few years, $\mathrm{CeCu_2Si_2}$ has emerged as a model system for multiband superconductivity with strongly correlated carriers. We certainly won't be surprised if the future will bring out yet more surprises about the superconductivity in $\mathrm{CeCu_2Si_2}$ and related heavy-fermion systems.

Acknowledgements

We would like to thank Wolf Aßmus, Ang Cai, Lei Chen, Piers Coleman, Pengcheng Dai, Onur Erten, Zach Fisk, Philipp Gegenwart, Christoph Geibel, Norbert Grewe, Malte Grosche, Haoyu Hu, Kenji Ishida, Kevin Ingersent, Lin Jiao, Hirale S. Jeevan, Stefan Kirchner, Michael Lang, Michael Loewenhaupt, Alois Loidl, Bruno Lüthi, Brian Maple, Kazumasa Miyake, Guiming Pang, Silke Paschen, Jed H. Pixley, Noriaki Sato, Doug Scalapino, Erwin Schuberth, Andrea Severing, Yu Song, Günter Sparn, Greg R. Stewart, Joe D. Thompson, Hao Tjeng, Roxanne Tutchton, Hilbert von Löhneysen, Franziska Weickert, Steffen Wirth, Rong Yu, Jinglei Zhang, Jian-Xin Zhu, and Gertrud Zwicknagl for useful discussions. Part of these discussions took place at the 2019 Zhejiang Workshop on Correlated Matter, which provided the initial motivation for this work. This work was supported by the National Key R&D Program of China (2022YFA1402203), the Key R&D Program of Zhejiang Province, China (2021C01002), the National Natural Science Foundation of China (12222410, 12034017, 11974306 and 12174331), the Zhejiang Provincial Natural Science Foundation of China (LR22A040002), at Arizona State University by the NSF Grant No. DMR-2220603 and an ASU startup grant; at Rice University by the NSF Grant No. DMR-2220603 and the Robert A. Welch Foundation Grant No. C-1411. Q.S. acknowledges the hospitality of the Aspen Center for Physics, which is supported by NSF Grant No. PHY-1607611.

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TABLE I Chronology of discoveries and early studies on heavy fermions, heavy-fermion superconductivity, and related topics (1969-1989).The following abbreviations are used: PM: paramagnetic, AF(O): antiferromagnetic (order), SDW: spin-density wave, CDW: charge-density wave, MF: mean field, FL: Fermi liquid, HF: heavy fermion, KE: Kondo effect, RLM: resonance level model (Schotte and Schotte, 1975), I: interpretation.

Year	**Discovery/Achievement**	**Material**	**Reference**
1969	First synthesis	$\mathrm{CeCu_2Si_2}$	Rieger and Parthé, 1969
1969	Superconductivity, $T_{\rm c} = 1.47$ K	$\mathrm{U_2PtC_2}$	Matthias *et al.*, 1969
1971	Fe/Cr-derived specific heat $\Delta C(T) = \gamma T$ at $T \ll T_{\rm K}$, $\gamma \approx 1(16)$ J/mol K^2	**Cu**(Fe,Cr) 80 (20-50) ppm	Triplett and Phillips, 1971
1972	Superfluidity	Liquid ^{3}He	Osheroff *et al.*, 1972a,b
1974	Theory of local FL of $S = \frac{1}{2}$ Kondo ion		Noziéres, 1974
1975	Superconducting transition at $T_c = 0.97$ K T_c decreases by 30% in $B = 6$ T. **I:** due to U - filaments	$\mathrm{UBe_{13}}$	Bucher *et al.*, 1975
1975	Heavy FL; $\gamma = 1.62$ J/mol K^2 **I:** due to $4f$-virtual bound state	$\mathrm{CeAl_3}$	Andres *et al.*, 1975
1975	Theory of superfluid phases	Liquid ^{3}He	Leggett, 1975
1976	Magnetic properties **I:** Intermediate-valence compound	$\mathrm{CeCu_2Si_2}$	Sales and Viswanathan, 1976
1978	$T_{\rm K} = 5$ K, $T_{\rm N} = 3.9$ K, $\gamma_{\rm AF} = 0.135$ J/mol K^2 KE/AFO treated by RLM/MF: $\gamma_{\rm PM} = 1.7$ J/mol K^2	$\mathrm{CeAl_2}$	Bredl *et al.*, 1978
1978	Superconducting transition at $T_c \approx 0.5$ K in resistivity and susceptibility **I:** due to spurious phase(s)	$\mathrm{CeCu_2Si_2}$	Franz *et al.*, 1978
1979	Bulk superconductivity, $T_{\rm c} \approx 0.6$ K (first HF superconductor) $\gamma \approx 1$ J/mol K^2, heavy fermions (introduction of the term “HF”)	$\mathrm{CeCu_2Si_2}$	Steglich *et al.*, 1979
1982	Lower and upper critical fields Meissner effect, strong Pauli limiting, **I:** even-parity pairing	$\mathrm{CeCu_2Si_2}$	Rauchschwalbe *et al.*, 1982
1983	HF superconductivity ($T_{\rm c} \approx 0.85$ K, $\gamma \approx 1.1$ J/mol K^2)	$\mathrm{UBe_{13}}$	Ott *et al.*, 1983
1983	Suppression of superconductivity by $\approx 1\%$ impurity substitution **I:** Unconventional superconductivity	$\mathrm{CeCu_2Si_2}$	Spille *et al.*, 1983
1984	HF superconductivity in single crystals	$\mathrm{CeCu_2Si_2}$	Assmus *et al.*, 1984 Ōnuki *et al.*, 1984
1984	Hump in $C(T)/T$, **I:** due to Kondo lattice coherence	$\mathrm{CeCu_2Si_2/CeAl_3}$	Bredl *et al.*, 1984
1984	$C(T) \sim T^3 (T \ll T_{\rm c})$ **I:** gap point nodes, p-wave superconductivity	$\mathrm{UBe_{13}}$	Ott *et al.*, 1984
1984	NMR: $1/T_1 \sim T^3$, **I:** gap line nodes	$\mathrm{(U_{1-x}Th_x)Be_{13}}$	MacLaughlin *et al.*, 1984
1984	Theory of superconductivity in Kondo lattice by Grüneisen-parameter coupling		Razafimandimby *et al.*, 1984
1984	Theory of triplet pairing in HF superconductors		Anderson, 1984
1984	HF superconductivity ($T_{\rm c} \approx 1.5$ K, $\gamma \approx 0.075$ J/mol K^2)	$\mathrm{U_2PtC_2}$	Meisner *et al.*, 1984
1984	HF superconductivity ($T_{\rm c} \approx 0.8 - 1.5$ K, $\gamma \approx 0.07$ J/mol K^2) MF-type transition at $T_0 = 17.5$ K, **I:** into SDW/CDW	$\mathrm{URu_2Si_2}$	Schlabitz *et al.*, 1984, 1986 Palstra *et al.*, 1985 Maple *et al.*, 1986
1985	dc-Josephson effect across $\mathrm{CeCu_2Si_2}$/Al weak link: ordinary critical pair current size	$\mathrm{CeCu_2Si_2}$	Steglich *et al.*, 1985b
1985	Second transition below $T_{\rm c}$, **I:** Unconventional superconductivity	$\mathrm{(U_{1-x}Th_x)Be_{13}}$	Ott *et al.*, 1985
1985	Second transition below $T_{\rm c}$, **I:** SDW transition	$\mathrm{(U_{1-x}Th_x)Be_{13}}$	Batlogg *et al.*, 1985
1986	Evidence for two superconducting states	$\mathrm{(U_{1-x}Th_x)Be_{13}}$	Lambert *et al.*, 1986
1986	Penetration depth: $\lambda(T) \sim T^2 (T \ll T_{\rm c})$, **I:** gap point nodes	$\mathrm{UBe_{13}}$	Gross *et al.*, 1986
1986	Theory of even-parity pairing caused by spin fluctuations		Miyake *et al.*, 1986
1986	Theory of d-wave pairing near an SDW instability		Scalapino *et al.*, 1986
1987	Evidence for two coexisting superconducting order parameters	$\mathrm{(U_{1-x}Th_x)Be_{13}}$	Rauchschwalbe *et al.*, 1987b
1988	dHvA oscillations: direct observation of HFs	$\mathrm{UPt_3}$	Taillefer and Lonzarich, 1988
1988	Penetration depth: $\lambda(T) \sim T^2 (T \ll T_{\rm c})$, **I:** gap nodes	$\mathrm{UPt_3}$, $\mathrm{CeCu_2Si_2}$	Gross *et al.*, 1988
1989	Second transition below $T_{\rm c}$, **I:** Unconventional superconductivity	$\mathrm{UPt_3}$	Fisher *et al.*, 1989
1989	Weak AFO, decrease of magnetic Bragg intensity below $T_{\rm c}$	$\mathrm{UPt_3}$	Aeppli *et al.*, 1989
1989	Theory on broken symmetry in an unconventional superconductor model for double transition in $\mathrm{UPt_3}$		Hess *et al.*, 1989

TABLE II A summary of experimental probes of the superconducting gap structure of $CeCu_2Si_2$, together with proposed theories for the superconducting pairing state.

Experiments

Probe	**Results**	**Interpretation**	**Reference**
Resistivity under field	Paramagnetic limiting of B_{c2}	Singlet pairing	Assmus *et al.*, 1984
Specific heat	$C \sim T^3$	–	Steglich *et al.*, 1985a
NMR Knight shift	Knight shift decrease below T_c	Singlet pairing	Ueda *et al.*, 1987
Penetration depth	$\lambda \sim T^2$	Gap nodes	Gross *et al.*, 1988
Point contact spectroscopy	Flat dV/dI	Nodeless gap	De Wilde *et al.*, 1994
Cu-NQR	$1/T_1 \sim T^3$	Gap line nodes	Ishida *et al.*, 1999 Fujiwara *et al.*, 2008
Inelastic neutron scattering	Peak in magnetic response below T_c	Sign-changing order parameter	Stockert *et al.*, 2011
Field angle dependent resistivity	4-fold $B_{c2}(\phi)$	d_{xy} state	Vieyra *et al.*, 2011
Specific heat ($T < 0.1$ K)	Exponential $C(T)$ as $T \to 0$	Two nodeless gaps	Kittaka *et al.*, 2014, 2016
Scanning tunneling microscopy	Spectra analysis	Nodal + nodeless gaps	Enayat *et al.*, 2016
Penetration depth ($T < 0.1$ K)	Exponential $\lambda(T)$ as $T \to 0$	Nodeless gap	Yamashita *et al.*, 2017 Takenaka *et al.*, 2017 Pang *et al.*, 2018
Thermal conductivity ($T < 0.1$ K)	Vanishing κ/T as $T \to 0$	Nodeless gap	Yamashita *et al.*, 2017
Cu-NQR ($T < 0.1$ K)	Exponential $1/T_1$ as $T \to 0$	Nodeless gap	Kitagawa *et al.*, 2017
Small angle neutron scattering	Form factor analysis	Two nodeless gaps	Campillo *et al.*, 2021

Theory

Theory	**Gap structure**	**Sign-change?**	**Reference**
Loop nodal s_{+-} state	Nodal	✓(interband)	Ikeda *et al.*, 2015
$d + d$ pairing	Nodeless	✓(intraband)	Nica *et al.*, 2017 Nica and Si, 2021
Multipole mediated s-wave	Nodeless	×	Tazai and Kontani, 2018, 2019 Tazai and Kontani, 2019
s_{+-} state	Nodal, nodeless	✓(interband)	Li *et al.*, 2018