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

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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 low temperatures ($T \lessapprox 0.1$ K) provided evidence for a fully gapped superconducting order parameter. In this Colloquium, after a historical overview the apparently conflicting results of numerous experimental studies on this compound are surveyed. The different theoretical scenarios that have been applied to understanding the particular gap structure are then addressed, 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 noninteracting electron bands. This property can be contrasted 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 material-specific band structure. 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, Nagaosa, and Wen, 2006; Proust and Taillefer, 2019), iron-based pnictides and chalcogenides (Stewart, 2011; Si, Yu, and Abrahams, 2016), organic charge-transfer salts (Lang and Müller, 2004; Maple *et al.*, 2004; Kanoda, 2008), and the moiré structures of graphene and transition-metal dichalcogenides (Cao *et al.*, 2018; Andrei and MacDonald, 2020).

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 detected by Triplett and Phillips (1971) for the dilute magnetic alloys CuCr and CuFe, the impurity-derived "incremental" low-temperature specific heat is proportional to temperature [$\Delta C(T) = \gamma T$], with a large 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 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; Stewart, 1984; Ott, 1987; Fulde, Keller, and Zwicknagl, 1988; Kuramoto and Kitaoka, 2012). 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, Graebner, and Ott, 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 CuFe. In addition, the low-temperature resistivity of $CeAl_3$ was observed to follow a $\Delta\rho(T) = AT^2$ dependence with a large prefactor A. These early findings were ascribed to a $4f$ virtual bound state at E_F. A large γ coefficient of the low-T specific heat similar 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 strong 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, Steglich, and Schotte, 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" characteristics, and the effective mass of the charge carriers exceeds that of ordinary conduction electrons by a factor up to about 1000 (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^2 mol [Fig. 1(a)], and there is a correspondingly enhanced temperature-independent Pauli spin susceptibility (Sales and Viswanathan, 1976; Grewe and Steglich, 1991) (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 involving 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 large A coefficient, whereas $CeCu_2Si_2$ exhibits non-Fermi-liquid behavior, as discussed in Sec. III.C.

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-Yoshida (RKKY) magnetic exchange interaction (Ruderman and Kittel, 1954; Kasuya, 1956; Yosida, 1957), which tends to stabilize the f-electron moments. While predominant Kondo screening results in a paramagnetic heavy-fermion ground

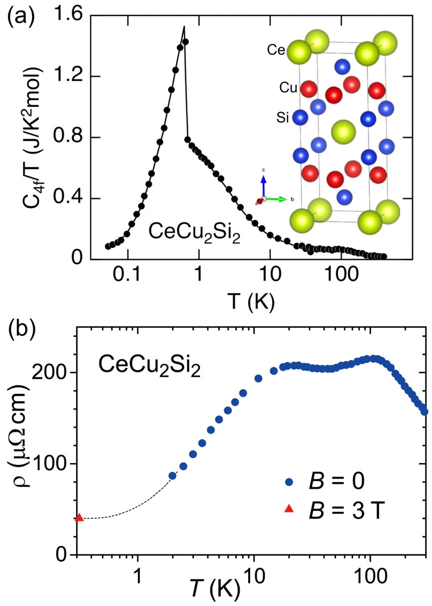

FIG. 1. (a) Contribution of the $4f$ electrons to the specific heat of $CeCu_2Si_2$ plotted as C_{4f}/T vs T on a logarithmic scale. The solid line is a guide for the eye. Inset: crystal structure of $CeCu_2Si_2$ ($ThCr_2Si_2$-type structure, space group $I4/mmm$), where the green, red, and blue spheres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

state, a dominant RKKY interaction causes magnetic, most frequently antiferromagnetic order. For 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 nonthermal control parameter, for instance, pressure, doping, or magnetic fields (Stewart, 2001; Gegenwart, Si, and Steglich, 2008; Si and Steglich, 2010; Sachdev, 2011). To eliminate the large residual entropy accumulated at the QCP, symmetry-broken novel phases are often observed, notably "unconventional" superconductivity that cannot be accounted for by the electron-phonon-mediated pairing mechanism of Bardeen-Cooper-Schrieffer (BCS) theory (Norman, 2011, 2014; 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 significant research interest again. While it was considered a single-band d-wave superconductor for many years (Ishida et al., 1999; Fujiwara et al., 2008), the observation of a fully developed energy gap at low temperatures (Kittaka et al., 2014; Takenaka et al., 2017; Yamashita et al., 2017; Pang et al., 2018) has led to proposals of $CeCu_2Si_2$ being a two-band s-wave superconductor both with (Ikeda, Suzuki, and Arita, 2015; Li et al., 2018) and without (Takenaka et al., 2017; Yamashita et al., 2017; Tazai and Kontani, 2018; Tazai and Kontani, 2019) a sign change of the order parameter.

In this Colloquium, after a 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 understanding 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, Yu, and Si, 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 $s_\pm$ scenario (Ikeda, Suzuki, and Arita, 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 Colloquium. 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 multiorbital systems and discuss future efforts that may shed further light on this canonical problem in the field of strongly correlated electron systems.

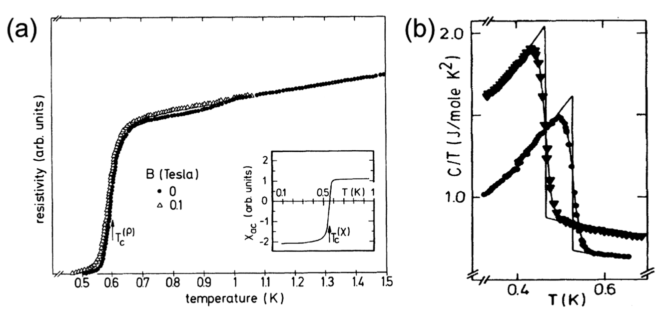

FIG. 2. (a) Resistivity $\rho(T)$, ac susceptibility $\chi_{ac}(T)$, and (b) specific heat as C/T vs T for polycrystalline $CeCu_2Si_2$ indicating bulk superconductivity at $T_c \approx 0.5$ K. The Pauli susceptibility ($T > T_c$) shown in the inset amounts to $\chi_P = 82 \times 10^{-9}$ m^3/mol (Aarts,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

II. HISTORY OF HEAVY-FERMION SUPERCONDUCTIVITY

Given the strong pair-breaking effect of diluted localized spins in conventional superconductors (Matthias, Suhl, and Corenzwit, 1958; Abrikosov and Gor'kov, 1960), the discovery of bulk superconductivity in $CeCu_2Si_2$ (Steglich et al., 1979) was surprising. In a BCS superconductor, a small amount of randomly distributed magnetic impurities fully suppresses the superconducting state (Maple, 1968; Riblet and

TABLE I. Chronology of discoveries and early studies on heavy fermions, heavy-fermion superconductivity, and related topics (1969–1989). PM, paramagnetic; AFM(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 or achievement	Material	Reference(s)
1969	First synthesis	$CeCu_2Si_2$	Rieger and Parthé (1969)
1969	Superconductivity, $T_c = 1.47$ K	U_2PtC_2	Matthias *et al.* (1969)
1971	Fe- or Cr-derived specific heat $\Delta C(T) = \gamma T$ at $T \ll T_K$, $\gamma \approx 1(16)$ J/mol K^2	Cu(Fe, Cr) 80 (20–50) ppm	Triplett and Phillips (1971)
1972	Superfluidity	Liquid ^{3}He	Osheroff, Richardson, and Lee (1972) and Osheroff *et al.* (1972)
1974	Theory of local FL of an $S = 1/2$ 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	UBe_{13}	Bucher *et al.* (1975)
1975	Heavy FL; $\gamma = 1.62$ J/mol K^2 *I*: due to $4f$-virtual bound state	$CeAl_3$	Andres, Graebner, and Ott (1975)
1975	Treatment of KE by renormalization group		Wilson (1975)
1975	Theory of superfluid phases	Liquid ^{3}He	Leggett (1975)
1976	Magnetic properties *I*: intermediate-valence compound	$CeCu_2Si_2$	Sales and Viswanathan (1976)
1978	$T_K = 5$ K, $T_N = 3.9$ K, $\gamma_{AFM} = 0.135$ J/mol K^2 KE/AFO treated by RLM/MF: $\gamma_{PM} = 1.7$ J/mol K^2	$CeAl_2$	Bredl, Steglich, and Schotte (1978)
1978	Superconducting transition at $T_c \approx 0.5$ K in resistivity and susceptibility *I*: due to spurious phase(s)	$CeCu_2Si_2$	Franz *et al.* (1978)
1979	Bulk superconductivity, $T_c \approx 0.6$ K (first HF superconductor) $\gamma \approx 1$ J/mol K^2, heavy fermions (introduction of the term HF)	$CeCu_2Si_2$	Steglich *et al.* (1979)
1982	Lower and upper critical fields Meissner effect, strong Pauli limiting, *I*: even-parity pairing	$CeCu_2Si_2$	Rauchschwalbe *et al.* (1982)
1983	HF superconductivity ($T_c \approx 0.85$ K, $\gamma \approx 1.1$ J/mol K^2)	UBe_{13}	Ott *et al.* (1983)
1983	Suppression of superconductivity by ≈1% impurity substitution *I*: unconventional superconductivity	$CeCu_2Si_2$	Spille, Rauchschwalbe, and Steglich (1983)
1984	HF superconductivity in single crystals	$CeCu_2Si_2$	Assmus *et al.* (1984) and Ōnuki, Furukawa, and Komatsubara (1984)
1984	HF superconductivity ($T_c = 0.5$ K, $\gamma = 0.4$ J/mol K^2)	UPt_3	Stewart *et al.* (1984)
1984	Hump in $C(T)/T$, *I*: due to Kondo-lattice coherence	$CeCu_2Si_2/CeAl_3$	Bredl *et al.* (1984)
1984	$C(T) \sim T^3 (T \ll T_c)$ *I*: gap point nodes, *p*-wave superconductivity	UBe_{13}	Ott *et al.* (1984)
1984	NMR: $1/T_1 \sim T^3$, *I*: gap line nodes	$(U_{1-x}Th_x)Be_{13}$	MacLaughlin *et al.* (1984)
1984	Theory of superconductivity in Kondo lattice by Grüneisen-parameter coupling		Razafimandimby, Fulde, and Keller (1984)
1984	Theory of triplet pairing in HF superconductors		Anderson (1984)
1984	HF superconductivity ($T_c \approx 1.5$ K, $\gamma \approx 0.075$ J/mol K^2)	U_2PtC_2	Meisner *et al.* (1984)
1984	HF superconductivity ($T_c \approx 0.8$–1.5 K, $\gamma \approx 0.07$ J/mol K^2) MF-type transition at $T_0 = 17.5$ K, *I*: into SDW or CDW	URu_2Si_2	Schlabitz *et al.* (1984, 1986), Palstra *et al.* (1985), and Maple *et al.* (1986)
1985	dc Josephson effect across $CeCu_2Si_2$/Al weak link: ordinary critical pair current size	$CeCu_2Si_2$	Steglich, Rauchschwalbe *et al.* (1985)
1985	Second transition below T_c, *I*: unconventional superconductivity	$(U_{1-x}Th_x)Be_{13}$	Ott *et al.* (1985)
1985	Second transition below T_c, *I*: SDW transition	$(U_{1-x}Th_x)Be_{13}$	Batlogg *et al.* (1985)
1986	Evidence for two superconducting states	$(U_{1-x}Th_x)Be_{13}$	Lambert *et al.* (1986)
1986	Penetration depth: $\lambda(T) \sim T^2 (T \ll T_c)$, *I*: gap point nodes	UBe_{13}	Gross *et al.* (1986)
1986	Theory of even-parity pairing caused by spin fluctuations		Miyake, Schmitt-Rink, and Varma (1986)
1986	Theory of *d*-wave pairing near a SDW instability		Scalapino, Loh, and Hirsch (1986)
1987	Evidence for two coexisting superconducting order parameters	$(U_{1-x}Th_x)Be_{13}$	Rauchschwalbe, Steglich *et al.* (1987)
1988	de Haas–van Alphen oscillations: direct observation of HFs	UPt_3	Taillefer and Lonzarich (1988)
1988	Penetration depth: $\lambda(T) \sim T^2 (T \ll T_c)$, *I*: gap nodes	UPt_3, $CeCu_2Si_2$	Gross *et al.* (1988)
1989	Second transition below T_c, *I*: unconventional superconductivity	UPt_3	Fisher *et al.* (1989)
1989	Weak AFMO, decrease of magnetic Bragg intensity below T_c	UPt_3	Aeppli *et al.* (1989)
1989	Theory on broken symmetry in an unconventional superconductor model for double transition in UPt_3		Hess, Tokuyasu, and Sauls (1989)
1989	Phenomenological theory of multiple pairing states	UPt_3	Machida, Ozaki, and Ohmi (1989)

Winzer, 1971; Maple et al., 1972; Steglich and Armbrüster, 1974), but the superconductivity is robust against doping with nonmagnetic impurities (Anderson, 1959; Balatsky, Vekhter, and Zhu, 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 nonsaturated, 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)].

Two early observations have led to the conclusion that $CeCu_2Si_2$ must be an unconventional bulk superconductor: (i) the nonmagnetic reference compound $LaCu_2Si_2$ is not a superconductor, at least down to 20 mK (Steglich et al., 1979), and (ii) a small amount of nonmagnetic (as well as magnetic) substitution at the level of 1 at. % may lead to a complete suppression of superconductivity in $CeCu_2Si_2$ (Spille, Rauchschwalbe, and Steglich, 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 $\mathrm{mJ/mol\,K^2}$; they substantially increase upon lowering the temperature and extrapolate to about 1 $\mathrm{J/mol\,K^2}$ 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, as with $CeAl_3$, the measured specific heat in this low-temperature range is practically identical to the electronic contribution ($\approx C_{4f}$). The corresponding renormalized kinetic energy $k_B T_F^*$ corresponds to the Kondo screening energy $k_B T_K$ ($T_K \approx 15$ K) (Stockert et al., 2011). Therefore, the ratio T_c/T_F^* is of the order of 0.04, compared to $T_c/T_F \approx 10^{-3}$–10^{-4} for an ordinary BCS superconductor, highlighting $CeCu_2Si_2$ as a "high-T_c superconductor" in a normalized sense (Steglich et al., 1979). On the other hand, the ratio T_F^*/θ_D, where θ_D is the Debye temperature, also amounts to about 0.05, while in a main group metal or transition metal T_F/θ_D is of the order of 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 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 $CeCu_2Si_2$ by a coupling of the heavy charge carriers to the breathing mode (Razafimandimby, Fulde, and Keller, 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, $CeCu_2Si_2$ was soon regarded generally as an unconventional, i.e., non-phonon-driven, superconductor. Because of the phenomenological similarity of heavy-fermion superconductivity in $CeCu_2Si_2$ with the superfluidity in ^3He (Osheroff, Richardson, and Lee, 1972; Osheroff et al., 1972), a magnetic coupling mechanism appeared to be most natural (Anderson, 1984).

The jump height at the superconducting transition $\Delta C/T_c$ is comparable to the Sommerfeld coefficient extrapolated to $T = 0$ [$\gamma_0 = C(T \to 0)/T \approx 1\ \mathrm{J/mol\,K^2}$] [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 small that within the scatter of the data it would not be resolvable in Fig. 2(b). Note that recent theoretical considerations have shown that in order to form Cooper pairs in $CeCu_2Si_2$, a large kinetic-energy cost, exceeding the binding energy by a factor as high as 20, is necessary to overcompensate for 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^*, which 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) that existed throughout the first few years after the first report of bulk superconductivity in $CeCu_2Si_2$ (Steglich et al., 1979), which was subsequently confirmed (Aliev et al., 1983b, 1984; Ishikawa, Braun, and Jorda, 1983; Ōnuki, Furukawa, and Komatsubara, 1984). The cause for these "sample dependences" [see also Aliev et al. (1983b) and Stewart, Fisk, and Willis (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 QCP that is located inside the narrow homogeneity range (Steglich et al., 2001; Lengyel et al., 2011); see Sec. III.C. The aforementioned skepticism would be overcome a few years later, when high-quality single crystals of $CeCu_2Si_2$ were prepared (Assmus et al., 1984; Ōnuki, Furukawa, and Komatsubara, 1984) and found to show even more pronounced superconducting phase transition anomalies than polycrystals do. The upper critical field curve $B_{c2}(T)$ of such a single crystal is displayed in Fig. 3. It reveals the following:

(i) Only a small anisotropy between the field being applied parallel and perpendicular to the basal tetragonal plane (inset of Fig. 1), contrasting with a pronounced anisotropy in the electrical resistivity (Schneider et al., 1983).

(ii) A shallow maximum was found at around $T = 0.15$ K (inset of Fig. 3) that seems to correspond

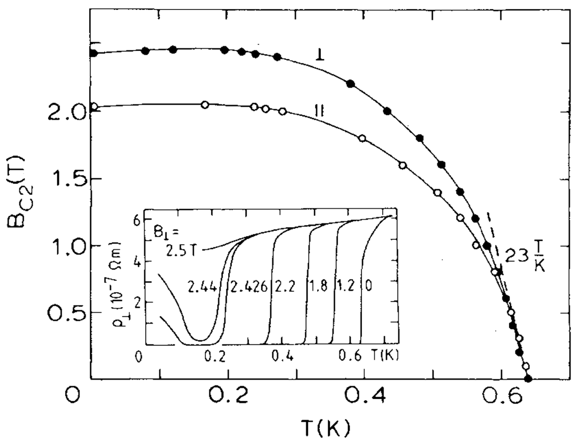

FIG. 3. Upper critical magnetic field B_{c2} vs T of a $CeCu_2Si_2$ single crystal for fields applied within (∥) and perpendicular to (⊥) the Ce planes obtained from $\rho(T)$ measured parallel to the respective field. There is only a moderate anisotropy, but a large initial slope at T_c is found for $B_{c2}(T)$. 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. From Assmus *et al.*, 1984.

to a low-temperature hump in $C(T)/T$ (Bredl *et al.*, 1984). It was also observed for $CeAl_3$ (Flouquet *et al.*, 1982; Bredl *et al.*, 1984; Steglich, Rauchschwalbe *et al.*, 1985), 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 to its relation to antiferromagnetic correlations (Steglich *et al.*, 1996; Stockert *et al.*, 2004). For UBe_{13}, a broad peak in $C(T)/T$ at $T_L \approx 0.6$ K had been detected (Rauchschwalbe, Ahlheim *et al.*, 1987; Rauchschwalbe, Steglich *et al.*, 1987) 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) suggest a spin-density-wave (SDW) transition, pressure studies (Lambert *et al.*, 1986) and results of the lower critical field (Rauchschwalbe, Ahlheim *et al.*, 1987) highlight a superconducting nature of the transition at T_{c2}.

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

(iv) A strong Pauli limiting effect is displayed in the low-temperature regime for both field configurations. This discards the odd-parity (spin-triplet) pairing 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 low temperatures close to the upper critical field based on Cu-NMR results was recently proposed (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, Rauchschwalbe *et al.*, 1985). 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, Schmitt-Rink, and Varma, 1986; Scalapino, Loh, and Hirsch, 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. More recently the Kondo effect has been incorporated into the study of heavy-fermion quantum criticality (Gegenwart, Si, and Steglich, 2008), with an emphasis on the notion of Kondo destruction (Coleman *et al.*, 2001; Si *et al.*, 2001). A corresponding theory was advanced for quantum-criticality-driven superconductivity in Kondo-lattice models (Hu, Cai, Chen, Deng *et al.*, 2021).

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 (Schlabitz *et al.*, 1984, 1986; Palstra *et al.*, 1985; Maple *et al.*, 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, Behnia, and Sierro, 1992), $CeRh_2Si_2$ (Movshovich *et al.*, 1996), $CeIn_3$, and $CePd_2Si_2$ (Mathur *et al.*, 1998). In the ensuing years, 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 TIn_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 $T_c = 18.5$ K for this class of superconductors (Sarrao *et al.*, 2002). Its Rh homolog $PuRhGa_5$ (Wastin *et al.*, 2003) as well as $NpPd_5Al_2$ (Aoki *et al.*, 2009) also show enhanced T_c values of 8.7 and 4.9 K, respectively. The discovery of heavy-fermion superconductivity in the noncentrosymmetric compound $CePt_3Si$ (Bauer *et al.*, 2004) stimulated the search for noncentrosymmetric heavy-fermion as well as weakly correlated superconductors (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 $CeRh_2As_2$, which has a locally noncentrosymmmetric crystal structure, two-phase superconductivity has recently been reported, 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 had previously been established for UPt_3 (Joynt and Taillefer, 2002), and multifaceted behavior has been reported for thoriated UBe_{13}

(Ott et al., 1985; Heffner et al., 1990; Oeschler et al., 2003) as well as URu_2Si_2, exhibiting a hidden-order phase (Mydosh, Oppeneer, and Riseborough, 2020). The three last materials all show a superconducting state with broken time-reversal symmetry (Heffner et al., 1990; Luke et al., 1993; Schemm et al., 2014, 2015).

There are currently only two known Yb-based heavy-fermion superconductors. β-$YbAlB_4$ with $T_c = 80$ mK (Nakatsuji et al., 2008) is an intermediate-valence compound showing quantum criticality without tuning (Matsumoto et al., 2011). $YbRh_2Si_2$ (Schuberth et al., 2016; Nguyen et al., 2021; Schuberth, Wirth, and Steglich, 2022; Shan et al., 2022) exhibits an antiferromagnetic QCP at $B \approx 0$ that is induced by nuclear spin order (below $T_A = 2.3$ mK). The latter strongly competes with the primary $4f$-electronic order ($T_N = 70$ mK) and causes the emergence of heavy-fermion superconductivity at ultralow temperatures ($T_c = 2$ mK). As shown by Schuberth, Wirth, and Steglich (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) (Custers et al., 2003). 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, Cai, Chen, and Si, 2021). Correlated Pr-based superconductors were also found. $PrOs_4Sb_{12}$ 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 $PrTi_2Al_{20}$, PrV_2Al_{20}, and $PrIr_2Zn_{20}$ are quadrupolar Kondo-lattice systems exhibiting superconductivity and quadrupolar order (Onimaru et al., 2011; Sakai, Kuga, and Nakatsuji, 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), URhGe (Lévy et al., 2005), and UCoGe (Huy et al., 2007; Hattori et al., 2012), as well as UPt_3 (Tou et al., 1998) and UNi_2Al_3 (Ishida et al., 2002). Also under discussion is UTe_2 (Aoki et al., 2019; Ran et al., 2019). It 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 (Duan et al., 2020, 2021; Thomas et al., 2020; Chen et al., 2021; Knafo et al., 2021).

In concluding this survey, we can state that currently about 50 heavy-fermion superconductors are known. Most of these materials were discussed by Pfleiderer (2009). They are complemented by the previously mentioned compounds β-$YbAlB_4$, $Pr(Ti,V)_2Al_{20}$, $PrIr_2Zn_{20}$, $YbRh_2Si_2$, UTe_2, and $CeRh_2As_2$. The majority of heavy-fermion superconductors are believed to have anisotropic even-parity Cooper pairing. In the following section, we present early evidence for single-band d-wave superconductivity in $CeCu_2Si_2$ down to about $T = 0.1$ K; see also Stockert et al. (2012).

III. EVIDENCE FOR d-WAVE SUPERCONDUCTIVITY IN $CeCu_2Si_2$ ABOVE 0.1 K

A. Phase diagram

One of the major distinguishing features that sets $CeCu_2Si_2$ apart from previously known BCS superconductors is the 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 that are entirely antiferromagnetic (A type), superconducting (S type), or exhibit both superconductivity and magnetism (A/S type) (Steglich et al., 1996; Seiro et al., 2010). While magnetism and superconductivity are generally considered antagonistic within the context of BCS theory, superconductivity on the border of magnetism is a common feature of broad classes of unconventional superconductors (Norman, 2011, 2014; Stewart, 2017), including heavy-fermion superconductors (Pfleiderer, 2009; Steglich and Wirth, 2016), cuprates (Lee, Nagaosa, and Wen, 2006; Proust and Taillefer, 2019), iron-based pnictides and chalcogenides (Stewart, 2011; Si, Yu, and Abrahams, 2016), organic superconductors (Lang and Müller, 2004; Maple et al., 2004; Kanoda, 2008), and twisted graphene superlattices (Cao et al., 2018; Andrei and MacDonald, 2020), 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 $T_N = 0.69$ K and a subsequent superconducting transition at $T_c = 0.46$ K. The application of moderate pressure rapidly suppresses T_N, while T_c shows a slight increase, and once T_N is suppressed below T_c no antiferromagnetic transition is observed. When a magnetic field is applied, both T_N and T_c are suppressed, but the more rapid decrease of T_c with field allows for T_N (extrapolated to $B = 0$) to be tracked as a function of pressure to lower temperatures. From extrapolating the positions of B_c^0 (where T_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 superconductivity, 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

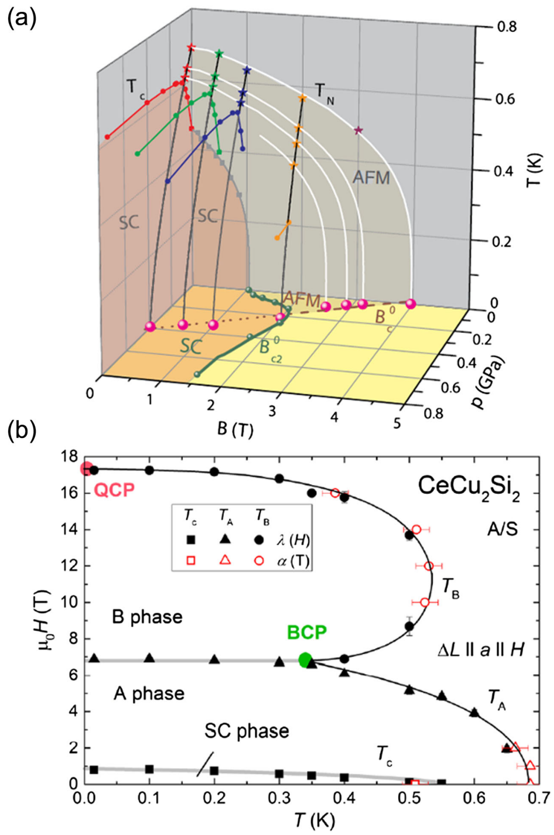

FIG. 4. (a) Temperature-pressure-magnetic-field phase diagram of a single crystal of A/S-type $CeCu_2Si_2$. From Lengyel et al., 2011. (b) Magnetic-field–temperature diagram of single crystal $CeCu_2Si_2$, where positions of the field-induced bicritical point (BCP) and QCP are also displayed. From Weickert et al., 2018.

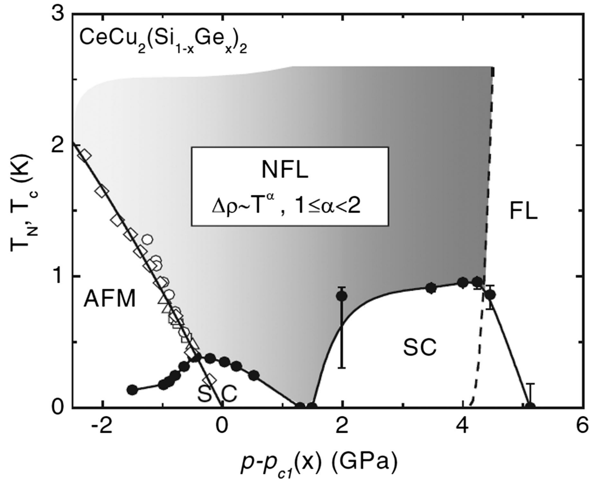

FIG. 5. Temperature-pressure phase diagram of $CeCu_2(Si_{1-x}Ge_2)_2$ that exhibits two superconducting domes (for $x = 0.1$), one centered around a lower pressure p_{c1} associated with an antiferromagnetic QCP, while the dome at higher pressures is near a possible valence transition. The diamonds, 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. The solid lines are a guide for the eye. From Yuan et al., 2006.

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 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 the A to the B phase is still to be determined, where the small change in magnetization between the two phases suggests that B (like A; see forthcoming discussion) also corresponds to a 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 unusual compared to other heavy-fermion superconductors (Mathur et al., 1998; Knebel et al., 2006), 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). 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 the other has 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 (Yuan et al., 2003; Holmes, Jaccard, and Miyake, 2004). 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 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 muon spin relaxation (μSR) measurements (Uemura et al., 1988, 1989) in the late 1980s, both of which detected 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. 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 cusplike 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). To unravel the nature of the A phase in pure $CeCu_2Si_2$, an approach to studying the magnetic order in the Ge-substituted system $CeCu_2(Si_{1-x}Ge_x)_2$ was chosen. Starting with 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 small single crystals were available, enabling only thermodynamic and transport measurements. With substantially improved crystal growth techniques (Seiro *et al.*, 2010; Cao *et al.*, 2011), large single crystals of $CeCu_2Si_2$ and $CeCu_2(Si_{1-x}Ge_x)_2$ (up to $\sim$cm^3 size) could be synthesized. When single crystal neutron diffraction is performed on $CeCu_2(Si_{1-x}Ge_x)_2$, the antiferromagnetic order could be followed to much lower Ge concentrations (Stockert *et al.*, 2003, 2005). 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 $\mathbf{k} = \mathbf{Q}_{AFM} = (0.215, 0.215, 0.53)$ at $T = 50$ mK agrees well with theoretical calculations of the Fermi surface using a renormalized band method (Zwicknagl, 1992; Zwicknagl and Pulst, 1993; Stockert *et al.*, 2004). The vectors 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. 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. Note that the propagation vectors in $CeCu_2(Si_{1-x}Ge_x)_2$ are 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).

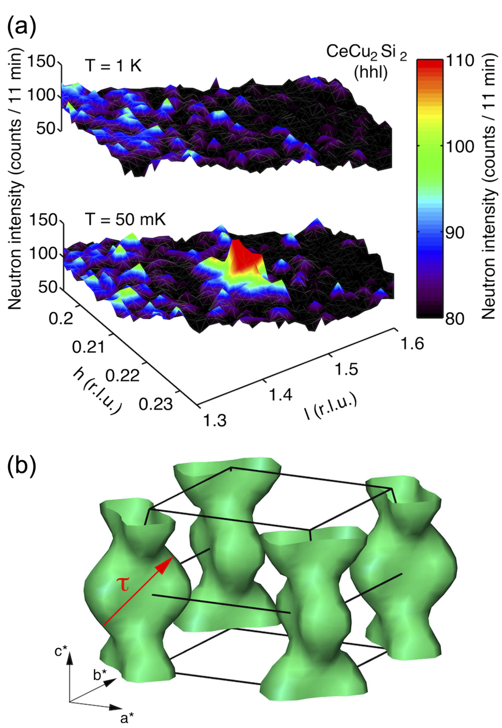

FIG. 6. (a) Neutron-diffraction intensity map of the reciprocal (hhl) plane around $\mathbf{Q} = (0.21, 0.21, 1.45)$ in A-phase $CeCu_2Si_2$ at $T = 50$ mK and 1 K. (b) Main heavy Fermi surface sheet in $CeCu_2Si_2$ indicating the columnar nesting with wave vector $\boldsymbol{\tau}$. From Stockert *et al.*, 2004.

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 (Luke *et al.*, 1994; Feyerherm *et al.*, 1997; Stockert *et al.*, 2006), in contrast to earlier reports on polycrystalline samples (Uemura *et al.*, 1988). Neutron diffraction on large A/S-type $CeCu_2Si_2$ single crystals also revealed that magnetic order and superconductivity do not coexist in $CeCu_2Si_2$ on a microscopic scale (Thalmeier *et al.*, 2005; Arndt *et al.*, 2010).

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 (the 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 (Stewart, 2001, 2006; Löhneysen *et al.*, 2007; Sachdev, 2011). 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; Si *et al.*, 2001; Senthil, Vojta, and Sachdev, 2004) 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), $YbRh_2Si_2$ (Paschen *et al.*, 2004; Gegenwart *et al.*, 2007; Friedemann *et al.*, 2010), and $CeRhIn_5$ (Shishido *et al.*, 2005; Park *et al.*, 2006). For paramagnetic $CeRhIn_5$ ($p > p_c$), the quantum-critical behavior changes at a certain crossover energy scale $E^* = k_B T^*$ (T. Park *et al.*, 2011), suggesting that the critical fluctuations of the Kondo effect, i.e., partial Mott physics, may be dominating above the crossover scale. $CeCu_2Si_2$ 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 with a $T^{3/2}$ dependence of the resistivity, as well as a $-T^{1/2}$ dependence of the specific-heat coefficient (Gegenwart *et al.*, 1998). In addition, the spin-excitation spectrum at the nesting wave vector $\boldsymbol{\tau} \approx \mathbf{Q}_{\mathrm{AFM}}$ in the normal state of superconducting (S-type) $CeCu_2Si_2$ displays an almost critical slowing down when superconductivity is suppressed by a magnetic field (Arndt *et al.*, 2011), as expected for a compound located 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 $CeCu_2Si_2$ a 3D SDW-type QCP seems to be realized, which is in line with the aforementioned thermodynamic and transport measurements (Arndt *et al.*, 2011; Stockert *et al.*, 2011). Measurements of the damping rate from inelastic neutron scattering (INS) have 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.

Note that $\mathbf{Q}_{\mathrm{AFM}}$ is not a singular point in $(\mathbf{Q}, \omega)$ space, but paramagnons are emerging out of $\mathbf{Q}_{\mathrm{AFM}}$ with an initial linear dispersion (Arndt *et al.*, 2011; Stockert *et al.*, 2011). When the magnetic response in the normal state of superconducting (S-type) $CeCu_2Si_2$ was compared to the antiferromagnetic state in A-type $CeCu_2Si_2$, the dispersion of the (para)magnons in both states was found to be similar, with merely a 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 occurring only at low-energy transfers below 0.5 meV due to the formation of a spin gap (Stockert *et al.*, 2011); see Sec. III.D. Recently INS experiments on S-type $CeCu_2Si_2$ have been extended to higher energy transfers up to several meV (Song *et al.*, 2021). These measurements 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; Stockert *et al.*, 2011; Huesges *et al.*, 2018). However, in addition, the dispersive spin excitations are now found to change to a dispersionless column in energy above ≈1.5 meV (Song *et al.*, 2021). The transition from dispersive to dispersionless magnetic excitations occurs around $k_B T_K$, i.e., the characteristic local energy scale in $CeCu_2Si_2$. Currently if and how these high-energy spin excitations are related to the unconventional heavy-fermion superconductivity in $CeCu_2Si_2$ are open questions.

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 $CeCu_2Si_2$, $\Delta\rho(T) = A'T^{\alpha}$. As mentioned, this was found to be $\alpha = 3/2$ by Gegenwart *et al.* (1998), whereas $\alpha = 1$ was reported by 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 (Luke *et al.*, 1994; Feyerherm *et al.*, 1997), these samples contain a minority phase of the 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 $CeCu_2Si_2$

Owing to the small magnetic moment and the low transition temperatures, INS experiments were performed on superconducting $CeCu_2Si_2$ using cold-neutron triple-axis spectroscopy. While the INS spectra in the normal state yield a quasielastic magnetic response at $\mathbf{Q}_{\mathrm{AFM}}$ with slowing down and scaling behavior, as mentioned, the spin dynamics in the superconducting state well below $T_c = 0.6$ K show a clear spin-excitation gap at $\mathbf{Q}_{\mathrm{AFM}}$ (Stockert *et al.*, 2008, 2011) followed by a well-defined maximum that is often called a spin resonance [Fig. 7(a)]. Note that this maximum exceeds the magnetic response in the normal state, in contrast to a simple s-wave superconductor, where no 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.

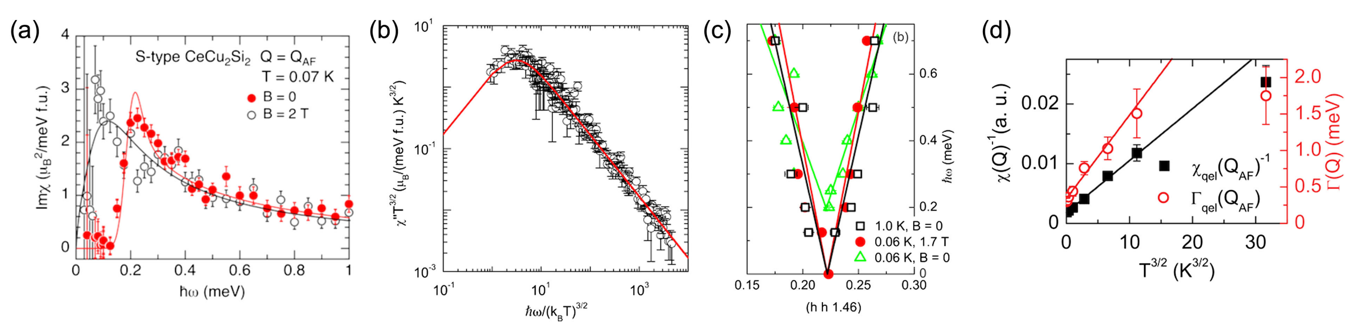

FIG. 7. Spin dynamics in $CeCu_2Si_2$. (a) Low-energy spin excitations in S-type $CeCu_2Si_2$ at $\mathbf{Q}_{\mathrm{AFM}}$ and $T = 0.07$ K in the superconducting state ($B = 0$) and the normal state ($B = 2$ T). From Stockert *et al.*, 2011. (b) Scaling of the normal-state quasielastic response in S-type $CeCu_2Si_2$ at $\mathbf{Q}_{\mathrm{AFM}}$ 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 $CeCu_2Si_2$. (d) Relaxation rate Γ and inverse susceptibility $\chi(\mathbf{Q})^{-1}$ of the normal-state magnetic response at $\mathbf{Q} = \mathbf{Q}_{\mathrm{AFM}}$ in S-type $CeCu_2Si_2$ vs $T^{3/2}$. (b)–(d) From Arndt *et al.*, 2011.

With a spin-gap size of about 0.2 meV, the maximum is located at $4k_BT_c$ and its position is therefore smaller than $2\Delta = 5k_BT_c$ 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 low-energy peak in the INS spectra of UPd_2Al_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). As in the latter case as well as in $CeCoIn_5$ (Song *et al.*, 2016, 2020), the peak in $CeCu_2Si_2$ develops in the one-particle channel, i.e., out of the aforementioned quasielastic line that persists to far above T_c for the two Ce-based compounds, and to well above T_N ($> T_c$) for UPd_2Al_3. This is different from the cuprates, where it manifests a singlet-triplet excitation of the d-wave condensate (Fong *et al.*, 1995; Sidis *et al.*, 2004).

Although this distinct maximum in the INS data at the edge of the spin-excitation gap should not be called a spin resonance, for the previously given reasons, it nevertheless highlights a sign-changing superconducting order parameter. Namely, if one considers coupling between a magnetic mode (such as a magnon or magnetic exciton) and the itinerant quasiparticles or Cooper pairs (Bernhoeft *et al.*, 1998, 2006), the observation in $CeCu_2Si_2$ of a significant low-energy enhancement of the INS intensity along the propagation vector $\mathbf{Q}_{AFM}$ 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 $CeCoIn_5$ 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, Kontani, and Sato, 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 (Yu *et al.*, 2009; Duan *et al.*, 2021). Second (and related to the first reason), for this scenario to occur the universality of the INS peak in the superconducting state that 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 can 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 $CeCu_2Si_2$ even in the normal state the paramagnons do not appear to be overdamped, as suggested by their highly visible dispersion at low energies [see Fig. 7(c)], even up to $k_BT_K \approx 1.5$ meV (Song *et al.*, 2021).

The experimentally determined propagation vector $\mathbf{Q}_{AFM}$ agrees well with the theoretically obtained nesting wave vector $\boldsymbol{\tau}$ shown in Fig. 6(b) (Zwicknagl, 1992; Zwicknagl and Pulst, 1993; Stockert *et al.*, 2004), which connects nested parts of the heavy quasiparticle band, highlighting intraband nesting. $\mathbf{Q}_{AFM}$ does not connect extended regions of different bands (interband nesting) of, for instance, electron and hole bands (Sec. IV.B), as required by the $s_\pm$-pairing model that has been considered for certain 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 sensitive to magnetic impurities, the nonmagnetic 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, Vekhter, and Zhu, 2006). Indeed, the high sensitivity of $CeCu_2Si_2$ to a 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, Rauchschwalbe, and Steglich, 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, Rauchschwalbe, and Steglich, 1983), and culminating in 20% for Th^{4+} (Ahlheim *et al.*, 1990). This trend with chemical pressure is analogous to that found when hydrostatic pressure is applied to $CeCu_2Si_2$ 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.

Needing such small critical substitutions on the transition-metal side to suppress T_c proves that this cannot be due simply 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 much 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

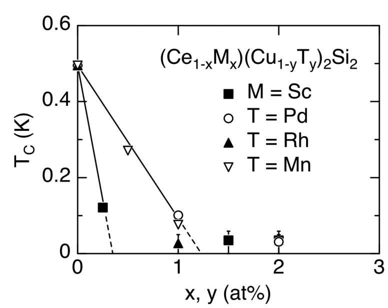

FIG. 8. Dependence of the superconducting transition temperature of $CeCu_2Si_2$ polycrystals on substitutions for Ce and Cu. Adapted from Spille, Rauchschwalbe, and Steglich, 1983.

domes in the temperature-pressure phase diagram, one dome sitting near a magnetic QCP and the higher pressure dome potentially lying near a valence transition (Fig. 5) (Yuan *et al.*, 2003; Holmes, Jaccard, and Miyake, 2004). 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 was reported that the suppression of T_c upon the introduction of disorder by electron irradiation was not as rapid as expected for sign-changing pairing states such as those in the cuprates or iron pnictides, but instead was 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 is 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 4) 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. previous results on yttrium barium copper oxide 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, Yu, and Abrahams, 2016). However, from the cuprates it is also known that atomic substitution can be 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). This is similar to the results of the aforementioned substitution experiments on $CeCu_2Si_2$ (Spille, Rauchschwalbe, and Steglich, 1983; Ahlheim *et al.*, 1988; 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 pair breaking, while certain kinds of lattice rearrangements, induced, for instance, 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.

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, which is in line with other Ce-based heavy-fermion superconductors (Thompson and Fisk, 2012), cuprate materials (Scalapino, 1995; Lee, Nagaosa, and Wen, 2006), and organic superconductors (Lang and Müller, 2004; Kanoda, 2008). A decrease of the Knight shift below T_c, the ordinary size of the dc Josephson effect between polycrystalline $CeCu_2Si_2$ and Al, and evidence for Pauli limiting of the upper critical field (Fig. 3) confirmed early on 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–nuclear quadrupole resonance (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 (Ishida *et al.*, 1999; Fujiwara *et al.*, 2008), although we note that the $1/T_1(T)$ data of Ishida *et al.* (1999) also showed some deviation from T^3 behavior at the lowest temperatures, as demonstrated in recent NQR experiments extended to somewhat lower temperature (Kitagawa *et al.*, 2017). 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 $\boldsymbol{\tau} \approx \mathbf{Q}_{AFM}$ 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 angular dependence of the upper critical field in the *a*-*b* 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

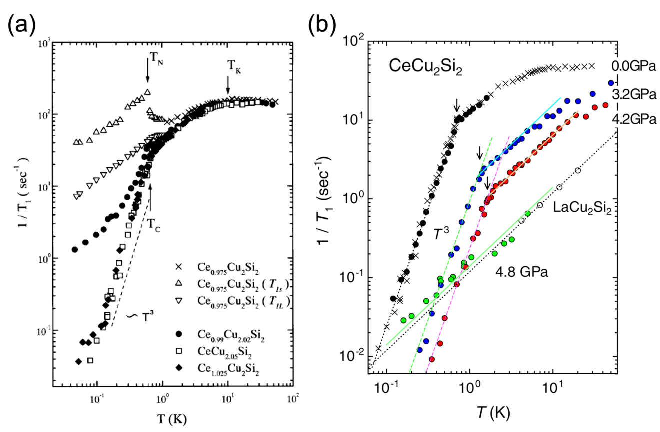

FIG. 9. Temperature dependence of the spin lattice relaxation rate $1/T_1(T)$ of $CeCu_2Si_2$, obtained from Cu-NQR measurements for (a) various superconducting and nonsuperconducting polycrystals, and (b) single crystals of superconducting $CeCu_2Si_2$ under hydrostatic pressure, as well as $LaCu_2Si_2$.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

nodes was long considered to be the most likely candidate for a pairing state.

IV. FULLY GAPPED UNCONVENTIONAL SUPERCONDUCTIVITY IN $CeCu_2Si_2$

A. Evidence for a nodeless gap structure

The understanding of the superconducting state of $CeCu_2Si_2$ underwent a radical overhaul following the results from low-temperature specific-heat measurements of Kittaka *et al.* (2014, 2016), which revealed that the superconducting gap is fully open over the entire 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 $\Delta_0 = 0.39 k_B T_c$. Since this is considerably less than the value of $1.76 k_B T_c$ 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 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, as 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 still needs to be clarified by future work. Upon rotating the field within the *a*-*b* plane, no modulation of the specific heat is observed, whereas in the single-band *d*-wave scenario a fourfold oscillation is predicted theoretically (Vorontsov and Vekhter, 2007; Boyd *et al.*, 2009) and observed experimentally in the Ce*T*In_5 series of heavy-fermion superconductors (Aoki *et al.*, 2004; An *et al.*, 2010; Lu *et al.*, 2012). Furthermore, measurements as a function of the polar angle θ reveal simply the twofold oscillations arising naturally from the tetragonal symmetry (Kittaka *et al.*, 2016).

Note that early evidence for a potentially exponential temperature dependence of the low-temperature specific heat was provided by measurements of $CeCu_2Si_2$ polycrystals, which revealed power-law behavior with an exponent of 2 near T_c but close to 3 at $T = 0.05$ K (Steglich, Ahlheim *et al.*, 1985). Further early evidence for fully gapped superconductivity was reported from a point contact study of $CeCu_2Si_2$ measured at 0.03 K (De Wilde *et al.*, 1994). De Wilde *et al.* 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.

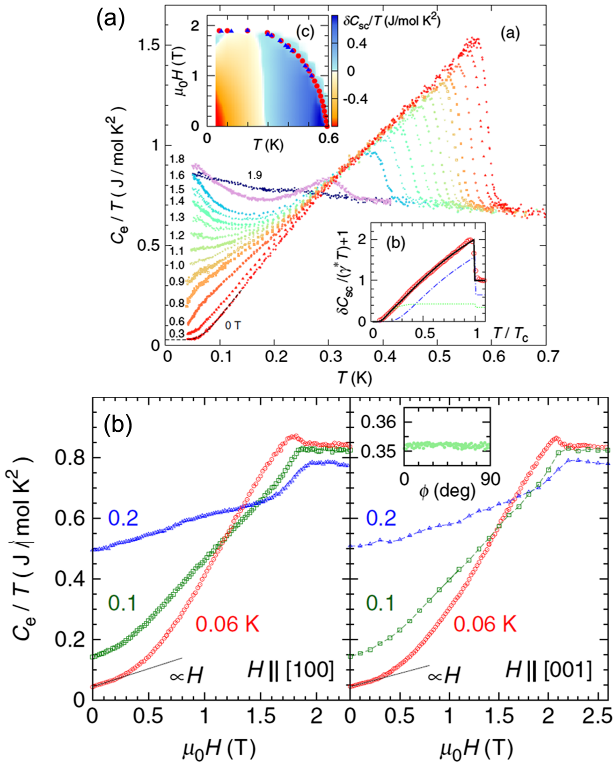

FIG. 10. (a) Temperature dependence of the electronic contribution to the specific heat as C_e/T of $CeCu_2Si_2$ down to temperatures of 0.04 K. Lower inset: analysis of the data using a nodeless two-gap model. Upper panel: the data as a contour plot. (b) The field dependence of the electronic specific-heat coefficient at various temperatures for fields along the (left panel) [100] and (right panel) [001] directions. At 0.06 K, linear behavior is observed at low fields for both field orientations. Inset in the right panel: C_e/T as a function of the in-plane azimuthal field angle ϕ, which remains constant. From Kittaka *et al.*, 2014.

Penetration depth measurements performed down to $T \approx 0.05$ K also demonstrate a fully open gap (Takenaka *et al.*, 2017; Yamashita *et al.*, 2017; Pang *et al.*, 2018). 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 $0.5 k_B T_c - 1 k_B T_c$). 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 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).

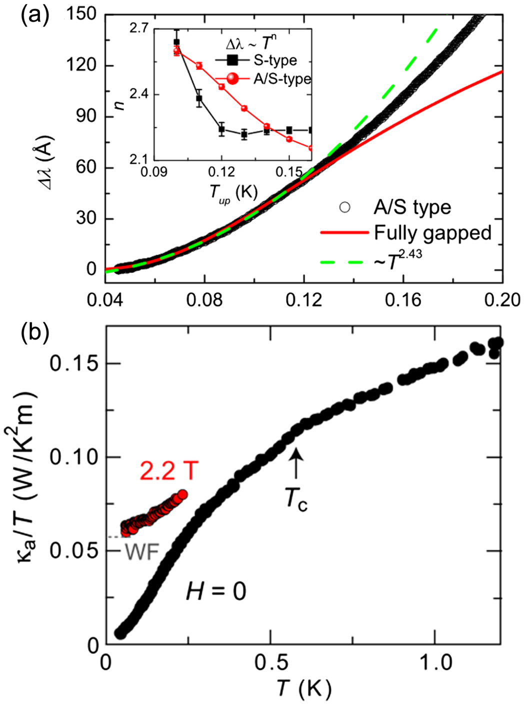

FIG. 11. (a) Temperature dependence of the magnetic penetration depth shift of an A/S-type $CeCu_2Si_2$ single crystal, measured using the tunnel-diode-oscillator-based method. The solid line shows the fit to the low-temperature data with a model for a fully gapped superconductor, where the lack of nodes in the gap is corroborated by an exponent n that is consistently larger than 2 (inset). From Pang *et al.*, 2018. (b) Temperature dependence of the in-plane thermal conductivity κ_a/T of $CeCu_2Si_2$, which in the superconducting state ($H = 0$) extrapolates to zero at $T = 0$, demonstrating fully gapped superconductivity. Also shown are the data in the normal state, which demonstrates the validity of the Wiedemann-Franz law. From 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 the applied field in the low-field region. Note 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 nonsuperconducting fractions, as well as better contacts between the heater and the sample. Earlier NQR measurements, however, were found to exhibit a T^3 dependence of $1/T_1(T)$ down to around 0.1 K (Ishida *et al.*, 1999; Fujiwara *et al.*, 2008); more recent results show a deviation from this behavior at low temperatures that can be accounted for by a small but nodeless gap (Kitagawa *et al.*, 2017).

While most recent low-temperature measurements, including small-angle neutron-scattering measurements, have indicated that the superconducting gap is fully open (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 recently; see Sec. IV.B. The tunneling spectra measured at low temperatures show two clear features at different voltage biases, providing clear evidence for multiple gaps or an anisotropic gap structure (Enayat *et al.*, 2016). However, the data were best accounted for using a model where the large gap is fully open but the small gap is nodal. The reason for this discrepancy is not clear, but note 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

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 (Zwicknagl and Pulst, 1993; Pourovskii *et al.*, 2014; Ikeda, Suzuki, and Arita, 2015; Zwicknagl, 2016; Li *et al.*, 2018; Luo *et al.*, 2020), 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 (Z. Wu *et al.*, 2021). Figure 12 summarizes the ARPES results from a typical S-type single crystal. The experimental Fermi surface of $CeCu_2Si_2$ 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 (the $\bar{M}$ point at the surface Brillouin zone corner); see Figs. 12(a) and 12(b). Measurements of the energy-momentum dispersion show that

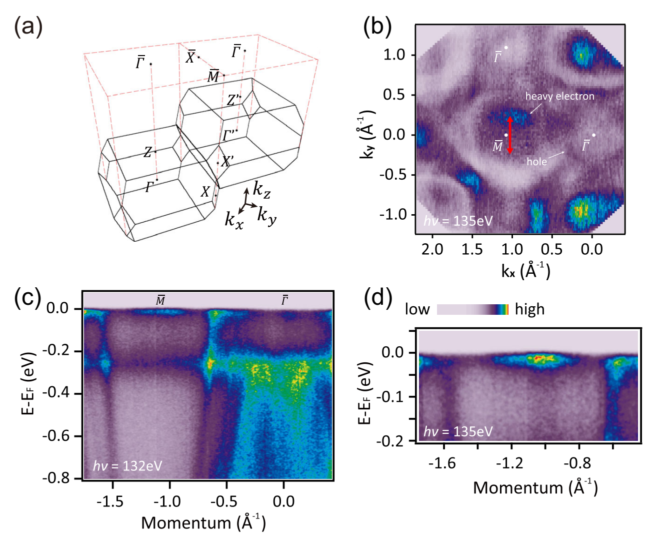

FIG. 12. Fermi surface and quasiparticle dispersion of S-type $CeCu_2Si_2$ from ARPES measurements. (a) Three-dimensional bulk Brillouin zone (black solid lines) and the projected surface Brillouin zone (red dashed lines) of $CeCu_2Si_2$. (b) Experimental k_x-k_y map at 10 K taken with 135 eV photons. The red arrow indicates the in-plane component of the SDW ordering wave vector $\mathbf{Q}_{\mathrm{AFM}}$ observed using neutron diffraction (Sec. III.B). (c) Band dispersion along $\bar{\Gamma}$-$\bar{M}$ at 10 K. (d) Enlargement of the heavy-electron band near E_F. From Z. Wu *et al.*, 2021.

the quasi-2D electron band is of predominant $4f$ character and possesses a large effective mass [Figs. 12(c) and 12(d)], while the hole bands near the $\bar{\Gamma}$ point are derived mainly 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 to heavy-fermion superconductivity (Zwicknagl and Pulst, 1993). Photon-energy-dependent scans and a 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 using the resolution-convoluted Fermi-Dirac distribution function and then fitting the extracted quasiparticle dispersion with a parabola. Owing 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 and Pulst, 1993; Zwicknagl, 2016). 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 7 times smaller than in the low-temperature limit (Steglich, 1990), the estimated effective masses indicate a good correspondence between ARPES and the specific heat. As illustrated in Fig. 6(b), renormalized band calculations (Zwicknagl and Pulst, 1993; Stockert *et al.*, 2004) reveal that this heavy-electron band has a warped part with flat parallel sides connected with a nesting vector $\boldsymbol{\tau}$, which is in excellent agreement with the SDW ordering wave vector $\mathbf{Q}_{\mathrm{AFM}}$ observed in neutron diffraction [Figs. 6(a) and 6(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 (Im *et al.*, 2008; Chen *et al.*, 2017; Jang *et al.*, 2020; Y. Wu *et al.*, 2021). 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. The $d+d$ matrix-pairing state

As discussed in Sec. III.F, the majority of the experiments in superconducting $CeCu_2Si_2$ 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 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 that changes sign within the heavy cylindrical bands near the edge of the Brillouin zone, as illustrated in Fig. 13.

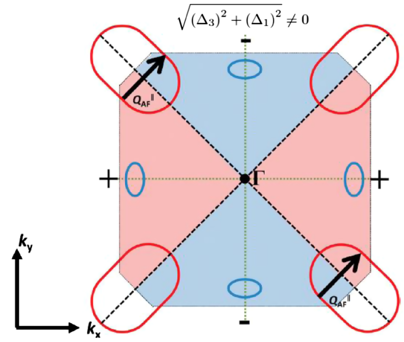

FIG. 13. Projection of the renormalized heavy Fermi surface and ordering wave vector $\mathbf{Q}_{\mathrm{AFM}} = (0.215, 0.215, 0.53)$ onto the $k_z = 0$ plane of the 3D Brillouin zone at zero temperature. The dashed lines indicate the nodes of the individual components of the $d+d$ pairing. The diagonal black and vertical and 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, the $d+d$ 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 $\mathbf{Q}^{||}_{\mathrm{AFM}}$ connects parts of the cylindrical Fermi surface near the edges (the red pill shapes), where $\Delta_3 \propto d_{x^2-y^2}$ has the opposite sign, leading to the emergence of a pronounced peak inside the superconducting gap in INS experiments. From Pang *et al.*, 2018.

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 $CeCu_2Si_2$ that 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 time-reversal symmetries. We instead consider a pairing state that can reconcile the features of superconducting $CeCu_2Si_2$ while preserving the previously mentioned symmetries. This is a multiband $d+d$ pairing of concurrent intraband $d_{x^2-y^2}$ and interband d_{xy} waves (Nica, Yu, and Si, 2017; Nica and Si, 2021). In its most general form, the $d+d$ 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 intraband and interband components Δ_3 and Δ_1 transform as $d_{x^2-y^2}$ and d_{xy}, respectively. This matrix-pairing state, which is intrinsically multiband, has additional structure due to the band space on which it is defined. The intraband $d_{x^2-y^2}$ component naturally satisfies the required sign change, much like a single-band d-wave pairing. In contrast to the latter, the matrix structure of the $d+d$ pairing, due to the anticommuting 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 by Nica and Si (2021), the $d+d$ pairing is a natural d-wave analog 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 the 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 Sec. IV.D.

While the $d+d$ 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 or spin space of the paired electrons. Matrix-pairing states that 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 multiband superconductors. [The properties of other Fe-selenide superconductors with a similar or higher T_c to 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 (Q.-Y. Wang *et al.*, 2012), are similar (Si, Yu, and Abrahams, 2016).] In spite of the difference in the nature of their basic constituents, these Fe-based superconductors share some of the experimental signatures that are similar to those in $CeCu_2Si_2$, namely, fully gapped superconductivity, as indicated by ARPES experiments (Mou *et al.*, 2011; Wang *et al.*, 2011; X.-P. Wang *et al.*, 2012; Xu *et al.*, 2012). This is supported by a spin resonance in the INS spectrum at the wave vector $\mathbf{Q}_{\text{alkaline FeSe}} = (0.5, 0.25, 0.5)$ (J. T. Park *et al.*, 2011; Friemel *et al.*, 2012), which is distinct from what one could expect from the sign-changing s-wave pairing. In any case, the latter scenario is unlikely given the absence of hole pockets at the center of the Brillouin zone. Nica, Yu, and Si (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 multiorbital $s\tau_3$ pairing is equivalent to the $d+d$ pairing in the band basis (Nica, Yu, and Si, 2017; Nica and Si, 2021). On the other hand, $s\tau_3$ transforms as a single B_{1g} irreducible representation of the D_{4h} point group. This implies that the $d+d$ 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, Yu, and Si, 2017; Nica and Si, 2021). Finally, we note that the $s\tau_3$ pairing and the equivalent $d+d$ pairing are energetically favored: they are stabilized in a multiorbital $t - J_1 - J_2$ model (Nica, Yu, and Si, 2017) in the regime where the A_{1g} and B_{1g} pairing channels are quasidegenerate.

Following the important precedent of the alkaline Fe selenides, Nica and Si (2021) constructed a microscopic candidate for an even-parity, spin-singlet $d+d$ pairing that incorporates the nature of the electronic states in $CeCu_2Si_2$. Matrix-pairing candidates can be constructed within the quasilocalized f-electron sector corresponding to the $f-f$ pairing, but also in the $f-c$ and $c-c$ sectors, where c stands for a conduction electron. As indicated in several experiments (Goremychkin and Osborn, 1993; Rueff *et al.*, 2015; Amorese *et al.*, 2020) and in studies utilizing local-density approximation with dynamical mean-field theory (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 symmetries of $f-f$ pairing states are determined entirely by the form factor. This implies that the $f-f$ pairing is not likely to support the $d+d$ pairing. Nica and Si (2021) considered an alternative in the $f-c$ pairing sector.

Conduction electron states that belong to the Γ_6 irreducible representation can be constructed by first taking linear combinations of the Cu $d_{x^2-y^2}$ orbitals that 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

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

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

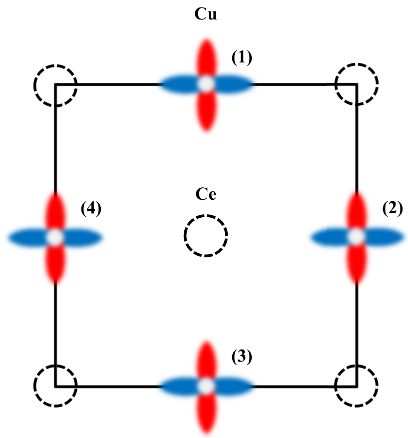

FIG. 14. Single Cu plane in the unit cell of $CeCu_2Si_2$. The four sites labeled (1)–(4) correspond to Cu $d_{x^2-y^2}$ orbitals in the plane. The dashed-line circles represent the Ce sites projected onto the Cu plane. From Nica and Si, 2021.

where ϕ denotes a spin-1/2 state. 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 the Γ_3 $f-c$ pairing is the analog of the $s\tau_3$ pairing introduced in the context of the alkaline Fe selenides. Thus, $s\Gamma_3$ provides a microscopic candidate for the $d+d$ pairing in $CeCu_2Si_2$. Evidence supporting this type of pairing was provided by x-ray absorption spectroscopy experiments (Amorese et al., 2020) that indicated a finite admixture of the f electron Γ_6 in the ground state of $CeCu_2Si_2$. Recall that the microscopic candidate for the $d+d$ pairing introduced by 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 the $d+d$ 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 band structure of the normal state.

Sign-changing $s_\pm$-pairing states were also advanced to explain the gapped, sign-changing superconductivity in $CeCu_2Si_2$ (Ikeda, Suzuki, and Arita, 2015; Li et al., 2018). We now summarize two of the most important differences between the $d+d$ and $s_\pm$ pairing states. First, although both candidates are sign changing and therefore conducive to a large peak in the INS intensity inside the superconducting gap, they also imply significantly different ways to involve the states on the Fermi surface. Li et al. (2018) [see also Ikeda, Suzuki, and Arita (2015)] carried out calculations using density-functional theory including the Hubbard parameter U, which capture neither the Kondo effect nor the associated renormalization toward heavy single-electron excitations. Physically, the proposed $s_\pm$ picture invokes a wave vector that spans the distance between the heavy cylindrical Fermi surface (the red pockets in Fig. 13) and the hole pocket near the Z point (the bulk Brillouin zone) projected from light bands (not shown in Fig. 13), 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 the 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 significantly different shapes and effective masses. In contrast, the $d+d$ pairing implies a wave vector spanning within the same cylindrical heavy Fermi surface (the 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. Second, the $d+d$ and $s_\pm$ pairing states have distinct nodal structures. As discussed, the $d+d$ pairing state has no nodes on the Fermi surface; see Fig. 13. By contrast, the $s_\pm$-pairing state has gap zeros that would generally be expected to intersect the extended hole Fermi surface of $CeCu_2Si_2$, leading to nodal excitations. This is different than 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 $s_\pm$ picture is not viable.

We conclude this section by revisiting the effects of disorder on the paired states in $CeCu_2Si_2$. As mentioned in Sec. III.E, the weak suppression of T_c in electron-irradiated samples was argued to point toward a more conventional order parameter that does not change sign (Takenaka et al., 2017; Yamashita et al., 2017), an interpretation that 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 nonmagnetic potential scattering (Anderson, 1997). Studies in models with strong short-range exchange interactions are consistent with this expectation (Garg, Randeria, and Trivedi, 2008; Chakraborty, Kaushal, and Ghosal, 2017). 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 the alkaline Fe selenides, where the $d+d$ state in the form of a $s\tau_3$ pairing was stabilized in a multiorbital $t-J_1-J_2$ model (Nica and Si, 2021). We expect strong correlations to also protect the $d+d$ pairing state in $CeCu_2Si_2$. In contrast, as previously mentioned, T_c can be sharply suppressed in $CeCu_2Si_2$ via atomic substitution, as is the case, for instance, in high-T_c superconductors with Zn substituted for Cu on the CuO_2 planes (Loram, Mirza, and Freeman, 1990).

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. (2018) and Smidman et al. (2018) found that the temperature dependence could be described by both an isotropic two-gap model and one for the $d+d$ band-mixing pairing state, which is displayed in Fig. 15. In the latter case, a simple model of the gap function is given by $\Delta(T,\phi) = \{[\Delta_1(T)\cos 2\phi]^2 + [\Delta_2(T)\sin 2\phi]^2\}^{1/2}$, which has a fourfold oscillatory component where one of the gap parameters corresponds to the gap minimum and the other to

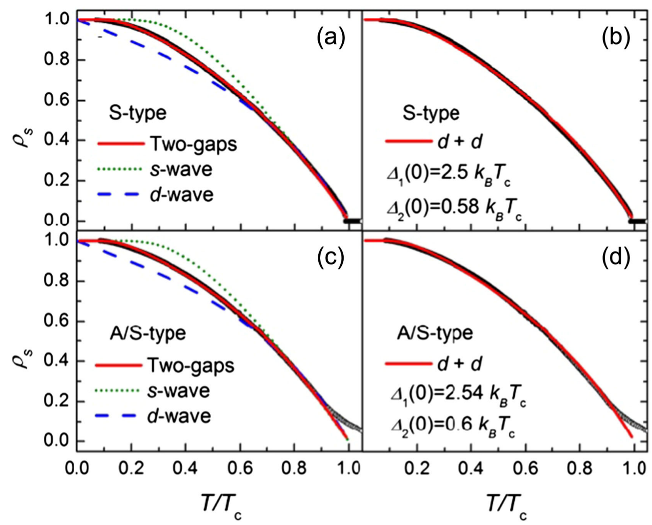

FIG. 15. Temperature dependence of the superfluid density derived from penetration depth measurements using the tunnel-diode-oscillator-based method. (a),(b) Results fitted to the superfluid density of an S-type sample with an isotropic two-gap model and $d + d$ band-mixing pairing model, respectively.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

the maximum. The basis for applying this model is explained in Sec. IV.C, and it is found that this model describes the data well across the entire temperature range. 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. (2014).

In the case of recent NQR measurements, this $d + d$ band-mixing pairing model can account well for $1/T_1(T)$ across the entire 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 $s_\pm$ scenario this peak is somewhat suppressed, but still present in the model. In analogy with Fe-based superconductors, effects such as quasiparticle damping and impurity-induced bound states (in the case of $s_\pm$ 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. Unambiguously discriminating between different fully gapped models will likely require high-resolution momentum-resolved experimental probes of the superconducting gap at low temperatures, which is 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 pair breakers. The origin of this nonuniversal impact of substitutional disorder on the superconductivity of $CeCu_2Si_2$ and the dichotomy between “harmful” and “harmless” (for instance, electron-irradiation-induced) disorder are interesting open questions to be unraveled in future work.

We also note that $CeCu_2Si_2$ has often been regarded as a prototypical example of both heavy-fermion superconductivity and 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 $Ce(Co,Ir)In_5$ (Izawa et al., 2001; Kasahara et al., 2008; Park et al., 2008; An et al., 2010; Lu et al., 2012; Allan et al., 2013; Zhou et al., 2013).

The spin-excitation spectrum of $CeCu_2Si_2$ consists of both long-wavelength SDW-type fluctuations (paramagnons) and 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 that is only about half the value of the critical pressure p_c at $B = 0$. When increasing the pressure at $p \ll p_\mathrm{c}$ far 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 one further approaches the QCP (at $p_\mathrm{c}/2 < p < p_\mathrm{c}$) 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 $T_\mathrm{c}(p)$ for the low-pressure dome displayed in Fig. 5 and may also apply to other correlated metals showing a superconducting dome centered at a 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 nonmonotonic evolution suggests that the Mott-type critical excitations are pair promoting, while the ultralow-temperature (below $T^* = 1$ K) SDW-type critical excitations in $CeCu_2Si_2$ are pair breaking. The theoretical work of Hu, Cai, Chen, Deng et al. (2021) for a 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 (Shishido et al., 2005; Park et al., 2006; Schuberth et al., 2016; Nguyen et al., 2021; Schuberth, Wirth, and Steglich, 2022; Shan et al., 2022).

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, Steglich, and Schotte, 1978; Bredl et al., 1984) 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 Colloquium. In the following, we 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 doublets and a Γ_6 Kramers doublet (Amorese et al., 2020), was recently verified by ARPES experiments performed at 10 K (Z. Wu et al., 2021), which is 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 $m^* \approx 120 m_e$ is in good agreement with that obtained from specific-heat results at the same temperature [Fig. 1(a)] (Steglich, 1990). In addition, ARPES revealed a hole band with a small but significant $4f$ contribution near the bulk Z point that corresponds to the distinct Fermi surface pocket with a moderately enhanced $m^* (\approx 5 m_e)$ 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 that probe the large Fermi surface of the Kondo-lattice state of $CeCu_2Si_2$ at finite temperatures, transport measurements (Sun and Steglich, 2013; Shan et al., 2022) appear to be dominated down to 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, Anderson, and Ramakrishnan (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 (Yuan et al., 2003, 2006; Holmes, Jaccard, and Miyake, 2004).

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 surprise to most researchers in the field of superconductivity, this might have been expected by researchers working on superfluid ^3He (Vollhardt and Wölfle, 1990). With the discovery of a heavy-fermion low-temperature phase in $CeAl_3$ (Andres, Graebner, and Ott, 1975), which resembles the renormalized normal phase of charge-neutral liquid ^3He at sufficiently low temperatures, the following question might arise: Is there a superconducting analog in a heavy-fermion metal like $CeAl_3$ to the superfluid phases in ^3He? Not surprisingly, magnetically driven superconductivity in heavy-fermion metals was proposed early on by theorists (Anderson, 1984; Miyake, Schmitt-Rink, and Varma, 1986; Scalapino, Loh, and Hirsch, 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 nonmagnetic analog compound $LaCu_2Si_2$ is not a superconductor (Steglich et al., 1979) as well as the drastic pair-breaking effect of certain nonmagnetic impurities, notably when substituted for Cu in $CeCu_2Si_2$ (Spille, Rauchschwalbe, and Steglich, 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, Si, and Steglich, 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 was 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 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, Si, and Steglich, 2008; Smidman et al., 2018). Theoretical studies that incorporate the Kondo-destruction physics in quantum-criticality-driven superconductivity have recently been developed (Hu, Cai, Chen, Deng et al., 2021).

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 (Gegenwart, Si, and Steglich, 2008; Arndt *et al.*, 2011; Smidman *et al.*, 2018), $T^* \sim 1$–2 K can be inferred, which is of 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 Mott-type fluctuations of local Ce moments with frequencies in the range $k_{\rm B}T^*/\hbar$ to $k_{\rm B}T_{\rm K}/\hbar$. The existence of a nonzero Kondo-destruction energy scale $k_{\rm B}T^*$ 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 for 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 20, which is 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 $B_{\rm c2}$ (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 notable 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 $T_{\rm c}$ is around 3 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 $T_{\rm c}$ vs p plateau, the existence of a distinct second high-pressure dome was apparent only 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 (Yuan *et al.*, 2003; Holmes, Jaccard, and Miyake, 2004).

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 $T_{\rm c}$ and a T^3 dependence of $1/T_1(T)$ (Ishida *et al.*, 1999; Fujiwara *et al.*, 2008). 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 (Takenaka *et al.*, 2017; Yamashita *et al.*, 2017; Pang *et al.*, 2018), thermal conductivity (Yamashita *et al.*, 2017), and more recent NQR measurements on $CeCu_2Si_2$ single crystals (Kitagawa *et al.*, 2017) that revealed 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, Suzuki, and Arita, 2015; Li *et al.*, 2018), as well as a $d + d$ matrix-pairing state (Nica, Yu, and Si, 2017; Pang *et al.*, 2018; Nica and Si, 2021); 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 $\mathbf{Q}_{\rm AFM}$. 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; Z. Wu *et al.*, 2021). This maximum demonstrates a sign change of the superconducting order parameter along $\boldsymbol{\tau}$, which means intraband pairing, as previously discussed. No such sign change is possible for isotropic BCS-type pairing. In addition, such on-site pairing is unfavorable in a heavy-fermion superconductor, as the heavy charge carriers forming the Cooper pairs have only a small kinetic energy of the order of $k_{\rm B}T_{\rm K}$, which is of the same order as their renormalized Coulomb repulsion. For an on-site pairing to operate in a BCS superconductor, the kinetic energy must be 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 reconcile with the exclusion of the on-site pairing associated with the strong Coulomb repulsion of the $4f$ electrons.

(ii) Anisotropic s-wave pairing also cannot explain the superconductivity of $CeCu_2Si_2$. To account for the pronounced peak observed in INS at $\mathbf{Q}_{\rm AFM}$ inside the superconducting gap, there would need to be interband nesting connected by the SDW ordering wave vector, whereas ARPES measurements (Z. Wu *et al.*, 2021) 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 $s_\pm$ scenario, where the sign changes between the hole and electron pockets (Li *et al.*, 2018); see also Ikeda, Suzuki, and Arita (2015).

TABLE II. 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(s)
Resistivity under field	Paramagnetic limiting of B_{c2}	Singlet pairing	Assmus *et al.* (1984)
Specific heat	$C \sim T^3$	$\cdots$	Steglich, Ahlheim *et al.* (1985)
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) and 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	Fourfold $B_{c2}(\phi)$	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	Takenaka *et al.* (2017), Yamashita *et al.* (2017), and 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(s)
Loop nodal $s_\pm$ state	Nodal	✓ (interband)	Ikeda, Suzuki, and Arita (2015)
$d + d$ pairing	Nodeless	✓ (intraband)	Nica, Yu, and Si (2017) and Nica and Si (2021)
Multipole mediated s wave	Nodeless	✗	Tazai and Kontani (2018, 2019)
$s_\pm$ state	Nodal, nodeless	✓ (interband)	Li *et al.* (2018)

A $d + d$ pairing state with intraband and interband components provides a natural resolution to all currently available experimental results and is in line with the importance of the nonperturbative 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 $CeCu_2Si_2$. On the microscopic level, a $d + d$ pairing is equivalent to a matrix-pairing state between f electrons in Γ_7 doublets and conduction electrons belonging to Γ_6 doublets. The nontrivial 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, Yu, and Si, 2017; Nica and Si, 2021), 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 $CeCu_2Si_2$ still requires a fully developed microscopic theory together with additional experimental results able to discriminate among different scenarios.

Taking all these together, $CeCu_2Si_2$, the first unconventional superconductor discovered, continues to grow in its role as a model system for strong correlation physics. The historical intuition about $CeCu_2Si_2$ 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 $CeCu_2Si_2$ are formed by the extremely heavy charge carriers existing in the low-temperature phase of the Kondo lattice proved that the superconducting pairing mechanism is incompatible with the conventional one of BCS theory. In modern times, $CeCu_2Si_2$, like $CeCu_{6-x}Au_x$ (Löhneysen *et al.*, 1994; Schröder *et al.*, 2000), $CePd_2Si_2$ (Mathur *et al.*, 1998), $CeCoIn_5$ (Paglione *et al.*, 2003), and $CeRhIn_5$ (Shishido *et al.*, 2005; Park *et al.*, 2006), has served as a model system for heavy-fermion antiferromagnetic quantum criticality. Here the Landau-type, SDW-type quantum criticality interacts with the beyond-Landau Mott-type quantum criticality in different energy ranges below the Kondo temperature. Progress over the past few years has shown $CeCu_2Si_2$ emerging as a model system for multiband superconductivity with strongly correlated carriers. We certainly will not be surprised if the future holds still more surprises about the superconductivity in $CeCu_2Si_2$ and related heavy-fermion systems.

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Correction: The sixth sentence of Sec. III D contained an error in wording and has been fixed.