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LETTER TO THE EDITOR

Coexistence of superconductivity and antiferromagnetism in the heavy-fermion superconductor $CeCu_2(Si_{1-x}Ge_x)_2$ probed by means of Cu nuclear quadrupole resonance—a test case for the SO(5) theory

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Abstract
We report, on the basis of Cu nuclear quadrupole resonance measurements, that superconductivity (SC) and antiferromagnetism (AF) coexist on a microscopic level in $CeCu_2(Si_{1-x}Ge_x)_2$, once a tiny amount of 1% Ge ($x = 0.01$) is substituted for Si. This coexistence arises because Ge substitution expands the unit-cell volume in nearly homogeneous $CeCu_2Si_2$ where the SC coexists with *slowly fluctuating magnetic waves*. We propose that the underlying exotic phases of SC and AF in nearly homogeneous and in slightly Ge-substituted $CeCu_2Si_2$ can be accounted for on the basis of the SO(5) theory that unifies the SC and AF. We suggest that the SC and AF in $CeCu_2Si_2$ have a common mechanism.

1. Introduction

$CeCu_2Si_2$ was the first heavy-fermion (HF) superconductor ($T_c \sim 0.65$ K) [1]. For antiferromagnetic (AF) HF $CeCu_2Ge_2$ ($T_N = 4.15$ K), which has the same lattice and electronic structure as $CeCu_2Si_2$, it was found that an AF phase meets a superconducting (SC) one at a critical pressure $P_c \sim 7.6$ GPa [2]. Its pressure–temperature (P–T) phase diagram is depicted in figure 1(a); $T_c \sim 0.65$ K is constant for $P_c < P < 12$ GPa, but increases at pressures exceeding $P \sim 12$ GPa. $CeCu_2Si_2$ was observed to behave at $P = 0$ much as $CeCu_2Ge_2$ does at P_c. Thus the superconductivity in $CeCu_2Si_2$ is anticipated to take place close to an AF phase at $P = 0$. The P–T phase diagram of $CeCu_2Si_2$ is also shown in figure 1(a) [3,4]; T_c remains constant for $0 < P < 2$ GPa, and then shows a rapid increase in the range $2 < P < 3$ GPa.

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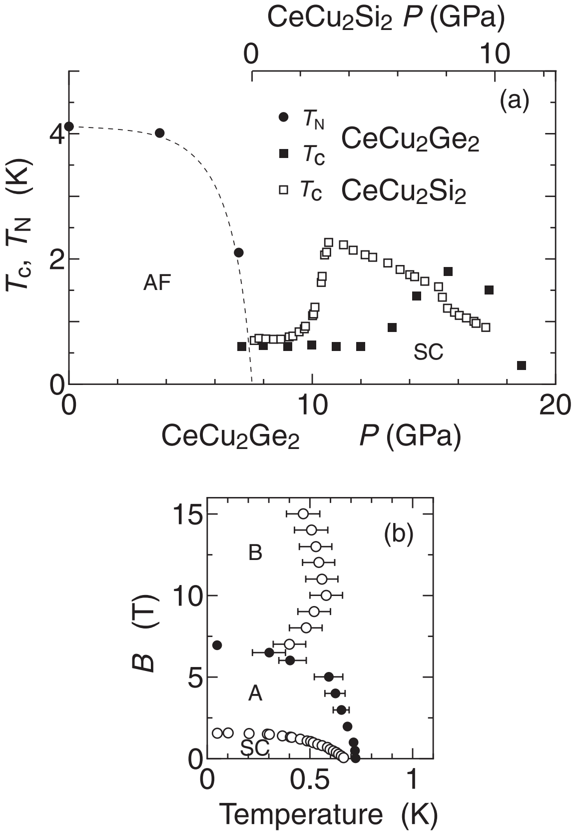

Figure 1. (a) The pressure (P) versus temperature (T) phase diagram for HF AF $CeCu_2Ge_2$ (reference [2]) and HF SC $CeCu_2Si_2$ (reference [3]). (b) The magnetic field (B) versus T phase diagram [10].

In recent years, there has been increasing evidence that the SC phase is observed even on a border at which the AF order is suppressed by applying pressure to the AF HF compounds $CePd_2Si_2$ and $CeIn_3$ [5]. It was suggested that this kind of SC phase is only viable extremely close to the critical lattice density D_c at which the AF order is suppressed, and in crystals of extremely high purity [5]. When a magnetic medium is near D_c, the waves of electron spin density tend to propagate over a long range. Therefore, it was argued that the binding of the Cooper pairs could be described in terms of the emission and absorption of waves of electron spin density. However, since the P–T phase diagrams were constructed from the resistivity measurements, the detailed magnetic nature near D_c or P_c in these systems has not yet been experimentally examined.

For metallic and insulating materials, when approaching an AF phase transition point (T_N; $T \to T_N$ or $T_N \to 0$ near D_c), *slowly fluctuating magnetic waves* become dominant. The nuclear spin–lattice relaxation rate, $1/T_1$, can be used to probe their low-frequency components; it is derived as $1/T_1 \propto \omega_Q/(\omega_Q^2 + \omega_N^2)$. Here ω_Q and ω_N are a characteristic frequency of magnetic waves and either a nuclear magnetic resonance (NMR) or a nuclear quadrupole resonance (NQR) frequency, respectively. When $T \to T_N$ or $T \to 0$ near D_c ($T_N \sim 0$), $\omega_Q \propto \sqrt{T - T_N}$ and hence $1/T_1$ reveals a sharp peak at T_N. Reduction of the NQR/NMR intensity also becomes pronounced because the magnetic waves develop at very low frequencies comparable to ω_N. Thus the NQR/NMR T_1 is one of the key measures for probing the existence of the magnetic waves of interest near D_c.

It is necessary to examine the characteristics of the magnetic medium near D_c to prove the existence of magnetically mediated superconductivity. Extensive experiments including NQR measurements revealed surprisingly disparate SC and normal-state properties that suggest *a break-up* of heavy quasiparticles in nearly homogeneous $CeCu_2Si_2$ samples, denoted as type I and type II, as follows [6–8]:

(i) Type I: polycrystal $Ce_{0.99}Cu_{2.02}Si_2$ (Ce0.99) shows a peak in $1/T_1$ at T_c, followed by a pronounced deviation from the $1/T_1 \propto T^3$ behaviour observed in a type-II sample. The fact that $1/T_1T$ tends toward a constant value well below T_c is indicative of a gapless SC nature with a finite density of states at the Fermi level (see figures 3(a) and 3(b), later) [7,8]. At pressures exceeding $P_A \sim 0.1$ GPa, the development of *slowly fluctuating magnetic waves* is depressed. Simultaneously, a SC phase that is consistent with a line-node gap model emerges at zero magnetic field ($B = 0$) [6, 8].

(ii) Type II: polycrystal $CeCu_{2.05}Si_2$ (Ce1.00) behaves as the best single crystal does, revealing a pronounced reduction in Cu NQR intensity above T_c. This indicates the development of *slowly fluctuating magnetic waves* in the normal state [7,9]. Below T_c at $B = 0$, however, such magnetic fluctuations are expelled [7, 9, 10]. As a result, the typical behaviour of $1/T_1 \propto T^3$ is observed in the HF SC state, consistent with the line-node gap (see figure 3, later) [7].

The NQR experiments do not give evidence of any static magnetic ordering in the type-I and type-II samples, at least at $B = 0$, but suggest that the former is extremely close to the border of a magnetic ordering. When the magnetic field exceeds an upper critical field H_{c2} for the type-I and type-II samples at $P = 0$ and suppresses the superconductivity, on the other hand, measurements of the elastic constant, the thermal expansion [10], $1/T_1T$ [4], and the specific heat [11] revealed an evolution from a SC phase into some magnetic phase that was called 'phase A'. This phase emerges below T_A close to the T_c at $B = 0$. Furthermore, the resistivity measurements on a single crystal suggested that 'phase A' seems to behave as a spin-density-wave-type (SDW-type) ordering [6], and the phase marked 'B' is also taken to be magnetic in origin [6, 10]. These field-induced phases are presented in figure 1(b).

Application of pressure compresses the unit-cell volume, V, and increases the lattice density $D = 1/V$. By contrast, partial Ge substitution for Si expands V linearly and hence decreases D [12]. The magnetic and SC phase diagram for $CeCu_2(Si_{1-x}Ge_x)_2$ for the range $0.02 < x < 1$ was reported using various types of measurement. Coexistence of 'phase A' and superconductivity on a microscopic scale was suggested [12].

In order to gain further insight into these exotic SC and magnetic phases and to clarify the characters of the magnetic fluctuations near D_c, we have investigated $CeCu_2(Si_{1-x}Ge_x)_2$ through Cu T_1-measurements and NQR spectrum measurements. We show that the SC and AF order coexist on a microscopic level, once a tiny amount of 1% Ge ($x = 0.01$) is substituted for Si. We propose that these exotic phases found in $CeCu_2Si_2$, which have been long-standing concerns—for over a decade, can be accounted for on the basis of the SO(5) theory that unifies antiferromagnetism and d-wave superconductivity [13]. We shed new light on SC states in strongly correlated electron systems near D_c.

2. Experimental details and results

We report here a Cu NQR study on $CeCu_2(Si_{1-x}Ge_x)_2$ at $x = 0.01$, 0.02, 0.1, and 0.2, which are the respective pieces of the same samples as were used in the previous work [12]. The high-frequency ac susceptibility (ac χ) was measured by using an *in situ* NQR coil to establish the bulk nature of the superconductivity. The bulk SC nature at $x = 0.01$ and 0.02 is corroborated by comparable sizes of the SC diamagnetism at low temperatures for the ac χ-data at various values of pressure for the type-I Ce0.99 as presented in figure 2, where the data at $x = 0.01$ are represented. The respective SC transition temperatures were determined as 0.5 and 0.4 K for $x = 0.01$ and 0.02. Note that the SC natures of the type-I Ce0.99 for 0.85 GPa $< P$ are also typical for the other HF SC compounds with the line-node gap [6, 8]. $T_c = 0.2$ K and 0.15 K were reported for $x = 0.1$ and 0.15, respectively, in reference [12]. The samples were moderately crushed into grains with diameters larger than 100 μm in order to avoid some crystal distortion. The Cu NQR spectrum was obtained by plotting the spin-echo intensity as a function of frequency at $B = 0$ and in a T-range 0.1–4.2 K. T_1 was measured by the conventional saturation–recovery method at $B = 0$ for $T = 0.05$–50 K.

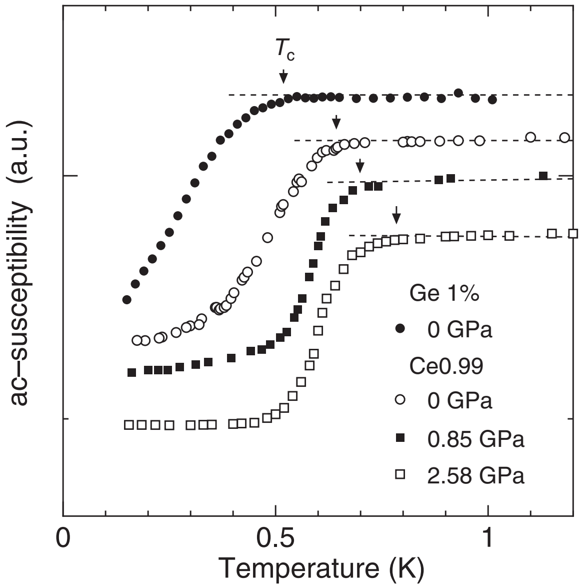

Figure 2. A representative ac susceptibility for 1% Ge-doped $CeCu_2Si_2$ along with ones for the type-I Ce0.99 at ambient pressure ($P = 0$), $P = 0.85$, and $P = 2.58$ GPa [8]. Note that the SC natures of the type-I Ce0.99 for 0.85 GPa $< P$ are also typical for the other HF SC compounds with the line-node gap reported thus far [6, 8].

Figures 3(a) and 3(b) show the respective T-dependences of $1/T_1$ and $1/T_1T$ for $x = 0.02$ along with those for Ce0.99 and Ce1.00 [7]. A single T_1-value was determined from the simple exponential recovery of the nuclear magnetization in the measured T-range except below T_N. Clear peaks in both quantities for $x = 0.02$ ($x = 0.01$, not shown) emerge at $T_N = 0.75$ (0.65) K, serving as a probe for the development of slow magnetic fluctuations towards an AF-type phase transition point. The data for $x = 0.02$ in the normal state ($T_N < T < 2$ K) appear to be consistent with a behaviour $1/T_1T = 6.6/\sqrt{T - 0.75} + 43$ (the solid line in the inset of figure 3(b)). A nearly or weakly AF itinerant-electron model (SCR theory) was applied to provide an understanding of the unusual normal and SC properties of the HF compounds near to an AF instability [14, 15]. In this SCR scheme, $1/T_1T \propto \sqrt{\chi_Q(T)} \propto 1/\sqrt{T - T_N}$ was predicted, since $\chi_Q(T)$ follows $C/(T - T_N)$. In fact, such a behaviour was reported for the AF HF superconductor UNi_2Al_3 [16]. The fact that the $1/T_1T$ data for the Ge-doped sample are fitted by including the term $6.6/\sqrt{T - 0.75}$

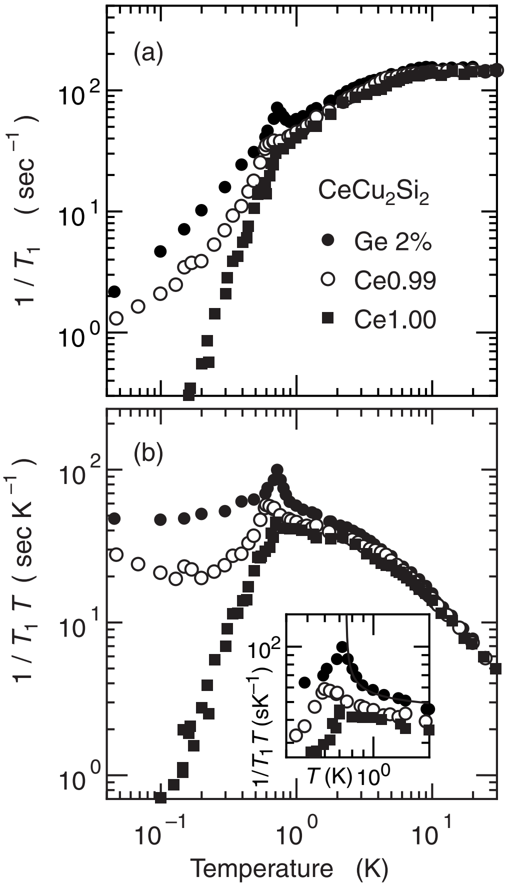

Figure 3. The temperature (T) dependence of (a) $1/T_1$ and (b) $1/T_1T$ for 2% Ge-doped CeCu$_2$Si$_2$ ($T_N = 0.75$ K and $T_c = 0.4$ K), Ce$_{0.99}$Cu$_{2.02}$Si$_2$ ($T_c = 0.65$ K), and CeCu$_{2.05}$Si$_2$ ($T_c = 0.7$ K) [7]. The inset indicates the $1/T_1T$ versus T plot on scales that are both expanded. The solid line indicates a fit with $1/T_1T = 6.6/\sqrt{T - 0.75} + 43$ that is consistent with the data for $T_N < T < 2$ K for $x = 0.02$.

demonstrates the long-range nature of the magnetic ordering. Furthermore, the emergence of an AF-type order below $T_N \sim 0.75$ K is corroborated by the appearance of internal fields (H_{int}) that are manifested by the significant increase of the NQR spectral width. These spectra are indicated in figure 4. It is at present hard to identify the magnetic structure from the hyperfine-broadened shape of the Cu NQR spectrum below T_N, although some SDW-like ordering appears likely from the distribution in H_{int}.

The T_N thus determined is plotted in figure 5(a) where the phase diagram of CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ is indicated together with that under pressure [8]. In the figure are shown an effective Fermi temperature T_F^* below which a T_1T = constant behaviour would be expected and T_m below which *slowly fluctuating magnetic waves are dominant* [8]. We conclude that $T_c < T_N$ and hence that the AF and SC order coexist, once Ge is slightly substituted for Si in the type-I Ce0.99, and eventually D becomes smaller than D_c ($D < D_c$). We should also remark that the T_1T = constant behaviour in the SC state (see figure 3(b)) evidences a

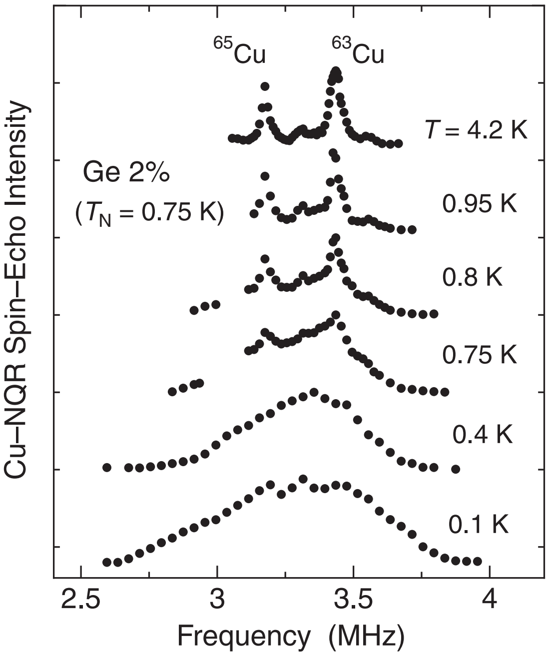

Figure 4. The T-dependence of the Cu NQR spectrum. The spectrum shows the hyperfine broadening due to the emergence of magnetic ordering below $T_N \sim 0.75$ K.

gapless SC state with a finite density of states[4]. This contrasts with the line-node gap state that is observed in the type-I Ce0.99 for 0.85 GPa $< P$ and the type-II Ce1.00 (see figure 3). The present NQR results have, for the first time, given convincing experimental evidence that the SC and AF order coexist for $D < D_c$ on *a microscopic scale*. It is noteworthy that the temperature T_A below which 'phase A' emerges [6, 10, 12] is higher than either T_N or T_c, but comparable to T_m. In this context, 'phase A', which was proposed to exist even at $B = 0$ on the basis of the various bulk measurements [6, 12], is shown to be neither an AF-type nor a SDW-type phase at $B = 0$ [7, 8].

Noting that $D = D_{\mathrm{Si}}[1 - (V_{\mathrm{Ge}} - V_{\mathrm{Si}})x/V_{\mathrm{Ge}}]$ for Ge doping and that D increases with the pressure, we present a combined phase diagram as a function of D in figure 5(b). In the phase diagram of figure 5(b), we denote the AF-type phase for $D < D_{c1}$ as (P-0), the coexistence phase of SC and AF order for $D_{c1} < D < D_c$ as (P-1), and the SC phase at $B = 0$ for $D_c \leqslant D \leqslant D_{c2}$ as (P-2). In (P-2), note that the SC state evolves into 'phase A' and subsequently 'phase B' as the magnetic field increases and exceeds H_{c2} as seen in figure 1(b) [10]. The SC region is wide for $D_{c1} < D$, whereas the AF order disappears at $B = 0$ for $D_c < D$. Thus, with increasing D, D_{c1} and D_c are characterized as the respective critical lattice densities at which the superconductivity sets in and the AF order is suppressed at $B = 0$. We suppose that D_{c2} is the third critical density, and that when this is exceeded there is no longer a field-induced 'phase A' [6].

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[4] We note that the absence of any clear reductions in $1/T_1$ and $1/T_1T$ below $T_c \sim 0.4$ K ($x = 0.02$) is in remarkable contrast to the bulk SC signatures. This is because low-frequency magnetic excitations in the AF state that govern the nuclear relaxation channel persist even in the SC state. This guarantees that the SC phase is not spatially segregated, but coexists with the AF order on a microscopic scale.

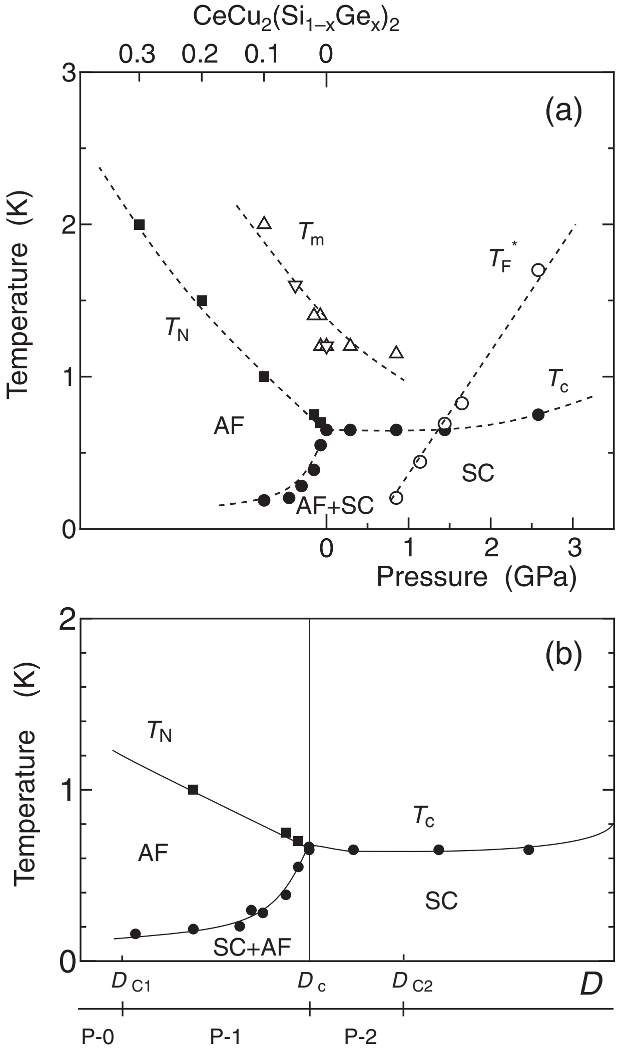

Figure 5. (a) The AF and SC combined phase diagram for CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ and for the type-I sample under pressure. Also shown are an effective Fermi temperature T_F^* below which a T_1T = constant behaviour would be expected and T_m below which slowly fluctuating magnetic waves are dominant [8]. (b) The AF and SC combined phase diagram as a function of the lattice density D in CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ ($D < D_c$) and in CeCu$_2$Si$_2$ ($D_c \leqslant D$) under pressure P. Note that $D \propto 1/V$, where V is the unit-cell volume, and $D = D_{\mathrm{Si}}[1-(V_{\mathrm{Ge}}-V_{\mathrm{Si}})x/V_{\mathrm{Ge}}]$ in the former case.

3. Phenomenology of the SO(5) theory

We now propose that the SO(5) theory that was proposed by Shou-Cheng Zhang [13] is promising as regards achieving an understanding of the underlying phases found in CeCu$_2$Si$_2$. In this unified theory of AF and SC order, a concept of *superspin* is introduced. It is a five-dimensional vector, $\vec{n}_i = (n_1, n_2, n_3, n_4, n_5)$. The magnitude of the vector is preserved with the constraint $\sum_{i=1}^{5} n_i^2 = 1$ or $|\Delta|^2 + |S_Q|^2 = 1$. Here

$$|\Delta|^2 = n_1^2 + n_5^2 \qquad |S_Q|^2 = n_2^2 + n_3^2 + n_4^2$$

are the respective amplitudes of the SC and AF order parameters (OP). *In this new theory, the AF and SC order are complementary and the same magnetic interaction as leads to an AF state also gives rise to pair binding.* In the low-energy asymmetric SO(5) Hamiltonian, a μ-term, $-2\mu Q_c$ (where Q_c is the total charge), is added, appearing as a 'gauge coupling' in the field theory. Then an effective potential energy is given by

$$V_{eff} = -\frac{g}{2}|S_Q|^2 - \frac{(2\mu)^2}{2}|\Delta|^2(\chi_c|\Delta|^2 + \chi_\pi|S_Q|^2).$$

Here χ_c is the charge compressibility and $Q_c = 2\mu(D)\chi_c$ is a function of the lattice density D. χ_π is the newly introduced 'π'-susceptibility which enables one to rotate the AF OP into the SC OP and vice versa. In order for the SO(5) symmetry to be approximately valid, these parameters must be close in value. For $|\Delta|^2 = 0$, since the AF phase is selected, a coupling constant g in the presence of explicit symmetry breaking is fixed at $g > 0$. Zhang predicted that the richness of the phase diagram at $T = 0$ comes entirely from varying $\mu(D)$ in the present case, as follows:

(1) [SO(5)-0]: an AF phase for $[2\mu(D)]^2 \leqslant g/\chi_\pi$;
(2) [SO(5)-1]: a coexistence phase of AF and SC order for $g/\chi_\pi < [2\mu(D)]^2 < g/(2\chi_c-\chi_\pi)$ when $\chi_c < \chi_\pi < 2\chi_c$; and
(3) [SO(5)-2]: a SC state for $(g/\chi_c) \leqslant [2\mu(D)]^2$ when $\chi_\pi \leqslant \chi_c$.

We point out that the underlying phases observed in $CeCu_2Si_2$ (figure 5) can be accounted for on the basis of the SO(5) theory by assuming that $\chi_\pi(D_{c1}) = g/[2\mu(D_{c1})]^2$ at D_{c1}, $\chi_\pi(D_c) = \chi_c(D_c) = g/[2\mu(D_c)]^2 \equiv \chi_0$ at D_c, and $\chi_\pi = 0$ at D_{c2}. A feature is that $\chi_c(D)$ and $\chi_\pi(D)$ are assumed to vary as functions of D as schematically indicated in figure 6(a). Here note that the variation in χ_c might be moderate because of the metallic states on both

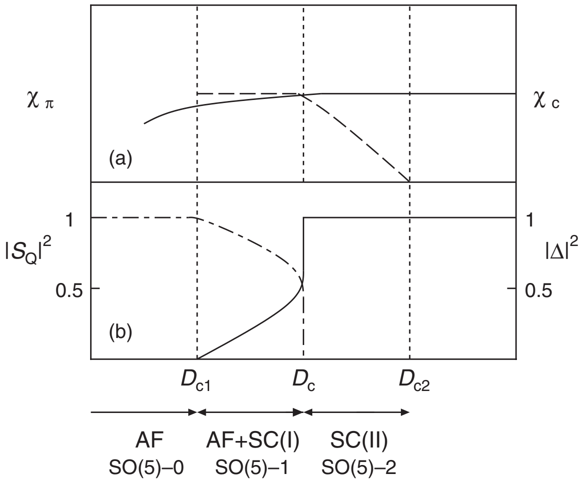

Figure 6. (a) The schematic variation in the charge compressibility χ_c (solid line) and the 'π'-susceptibility χ_π (dashed line) (see the text) shown as functions of D to display the consistency with the complex phases (P-0), (P-1), and (P-2) in figure 5(b). The phases predicted at $T = 0$ on the basis of the SO(5) theory are assigned as follows: [SO(5)-0] to the AF phase (P-0); [SO(5)-1] to the coexistence phase of AF and SC order (P-1); and [SO(5)-2] to the SC phase (P-2). (b) Possible variations in the AF OP, $|S_Q|^2$ (dash–dot line), and the SC OP, $|\Delta|^2$ (solid line), against D in [SO(5)-1].

sides of D_c and that

$$\chi_\pi(D_{c1}) \sim \left[1 + 2\left(\frac{\partial\mu}{\partial D}\right)\frac{D_c - D_{c1}}{\mu(D_c)}\right]\chi_0$$

is experimentally determined. Indeed, $\chi_\pi(D_{c1})$ and $\chi_0(D_c)$ are close in value, because $(D_c - D_{c1})/D_c \sim 0.01$ and $(\partial\mu/\partial D)$ is very small [8, 12]. The SO(5) symmetry is hence supposed to be approximately valid. We assign the respective phases (P-0), (P-1), and (P-2) in figure 5(b) to [SO(5)-0], [SO(5)-1], and [SO(5)-2]. Schrieffer *et al* described the phase [SO(5)-1] as a spin-bag phase in terms of pairing the eigenstates of the AF background [17]. We suggest that the AF OP and SC OP change as functions of D in the phase [SO(5)-1] as indicated in figure 6(b).

On the basis of the SO(5) theory, it is possible to account for the exotic SC phase in the type-I Ce0.99 which exhibits anomalies associated with *slowly fluctuating magnetic waves* [7, 9]. In the case where $\chi_0 \simeq \chi_c(D) \leqslant \chi_\pi(D)$ and $\mu(D) \simeq \mu(D_c)$, we may infer that the AF and SC order coexist with $|\Delta|^2 = |S_Q|^2 = 1/2$ at $T = 0$. For a finite temperature in the range $T_c < T \leqslant T_N$, on the other hand, we suggest that SC fluctuations prevent the onset of AF order when the characteristic frequencies of fluctuations are imposed such that

$$\omega_Q(T) \simeq \omega_{\mathrm{SC}}(T) < [2\mu(D_c)]\sqrt{\frac{(T_N - T)}{T_N}|\Delta(T)|^2[|\Delta(T)|^2 + \frac{\chi_\pi}{\chi_0}|S_Q(T)|^2]}.$$

Here note that $|S_Q(T)|^2 < |\Delta(T)|^2 < 1$. Therefore, *slowly fluctuating magnetic waves* may survive even below T_c, as was actually observed in the type-I Ce0.99 [7–9]. In the case where $\chi_c(D) < \chi_\pi(D)$ and $\mu(D) < \mu(D_c)$, however, the onset of the AF order becomes energetically more stable and this order coexists with the SC phase for $T < T_c < T_N$. The SO(5) theory accounts for the unusual SC and magnetic phenomena exhibited by the type-I Ce0.99 [6, 7, 9] as well as the coexistence of SC and AF order in $\mathrm{CeCu_2(Si_{x-1}Ge_x)_2}$. In this context, the type-I Ce0.99 is anticipated to be quite close to the condition for the SO(5) symmetry to be exact, i.e. $\chi_\pi = \chi_c$.

We next argue, on the basis of the SO(5) theory [18], in favour of a magnetic field-induced 'phase A' in the type-II samples where $D \leqslant D_{c2}$. In the *superspin* picture, starting from a SC state at $B_z = 0$, $\vec{n}$ lies in the (n_1, n_5) SC subplane. As B_z increases, a uniform spin polarization is induced along a SC vortex over a London penetration depth, λ. At the same time, the staggered polarization, $n_2^2 + n_3^2 = |S_Q^\perp|^2$ perpendicular to B_z, is induced over a SC coherence length, ξ, where $|\Delta(r)|^2 = |\Delta|^2[1 - \exp(-r/\xi)]$. This is because the magnitude of the *superspin* is preserved. In the presence of B_z and μ, by taking a general Ginzburg–Landau potential of an approximate SO(5) model [18], it was argued that the magnetic field induces either a first-order phase transition from the SC state to the AF state at a critical value of the magnetic field B_c or two second-order phase transitions with an intervening mixed-phase region where the SC and AF order coexist. It is here proposed that 'phase A' in the type-II sample (see figure 1(b)) corresponds to this magnetic field-induced AF state in [SO(5)-2], although further investigations are required to determine the magnetic structure of 'phase A'. Indeed, we believe that the phenomenological approach based on the SO(5) theory has given a coherent interpretation for

(1) the coexistence phase of SC and AF order in $\mathrm{CeCu_2(Si_{1-x}Ge_x)_2}$;
(2) the exotic SC phase in the type-I Ce0.99 near D_c where we propose $\chi_\pi \sim \chi_c$; and
(3) the magnetic field-induced 'phase A' in the type-II Ce1.00.

4. Implications of the new concept for superconductivity

The unconventional interplay between superconductivity and magnetism found in homogeneous $CeCu_2Si_2$ has been a long-standing problem—unresolved for over a decade. It was shown to originate from the system being just on the border of the AF phase ($D \sim D_c$) and the SC and AF coexisting once $D < D_c$. We have proposed that the SO(5) theory constructed on the basis of quantum-field theory (references [13, 18, 19]) is able to give a coherent interpretation for these exotic phases found in the strongly correlated HF superconductor $CeCu_2Si_2$. In this context, we may suggest that the superconductivity in $CeCu_2Si_2$ could be mediated by the same magnetic interaction as leads to the AF state in $CeCu_2(Si_{1-x}Ge_x)_2$. This is in marked contrast to the BCS case, in which the pair binding is mediated by phonons—vibrations of the lattice density. We believe that this model could shed further light on current ideas regarding the magnetic mechanism of the superconductivity in other strongly correlated electron systems—perhaps even the high-T_c cuprates [13, 19] and the organic [20] superconductors.

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