

Chapter 2

Non-centrosymmetric Heavy-Fermion Superconductors

N. Kimura and I. Bonalde

Abstract In this chapter we discuss the physical properties of a particular family of non-centrosymmetric superconductors belonging to the class heavy-fermion compounds. This group includes the ferromagnet UIr and the antiferromagnets CeRhSi₃, CeIrSi₃, CeCoGe₃, CeIrGe₃ and CePt₃Si, of which all but CePt₃Si become superconducting only under pressure. Each of these superconductors has intriguing and interesting properties. We first analyze CePt₃Si, then review CeRhSi₃, CeIrSi₃, CeCoGe₃ and CeIrGe₃, which are very similar to each other in their magnetic and electrical properties, and finally discuss UIr. For each material we discuss the crystal structure, magnetic order, occurrence of superconductivity, phase diagram, characteristic parameters, superconducting properties and pairing states. We present an overview of the similarities and differences between all these six compounds at the end.

2.1 CePt₃Si

The enormous interest in superconductors without inversion symmetry started with the discovery of superconductivity in the heavy-fermion compound CePt₃Si [1], which exhibits long-range antiferromagnetic (AFM) order below the Neel temperature $T_N = 2.2$ K and becomes superconducting at the critical temperature $T_c = 0.75$ K. CePt₃Si is the only known heavy-fermion (HF) compound without

N. Kimura (✉)
Center for Low Temperature Science,
Tohoku University,
Sendai Miyagi 980-8578, Japan
email: kimura@mail.clts.tohoku.ac.jp

I. Bonalde
Centro de Física, Instituto Venezolano de Investigaciones Científicas,
1020-A, Caracas, 20632 Apartado, Venezuela
email: ijbonalde@gmail.com

Table 2.1 Normal and superconducting parameters of CePt₃Si

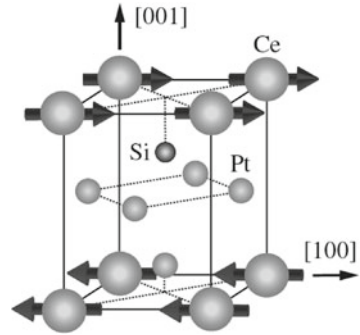
Crystal structure	Tetragonal
Space group	$P4mm$
Lattice parameters	$a = 4.072 \text{ \AA}$ $c = 5.442 \text{ \AA}$
Sommerfeld value of specific heat at T_c	$\gamma_n = 300 - 400 \text{ mJ/mol K}^2$
Effective electron mass (Fermi sheet α)	$m^* \sim 11 - 23 m_0$
Mean free path	$l = 1200 - 2700 \text{ \AA}$
Antiferromagnetic transition temperature	$T_N = 2.2 \text{ K}$
Antiferromagnetic propagation vector	$\mathbf{q} = (0, 0, 1/2)$
Staggered magnetic moment $\mathbf{m}_Q$ along	$[100]$
Ordered moment per Ce atom	$\mu_s = 0.16 \mu_B$
Superconducting transition temperature	$T_c = 0.75 \text{ K (or } 0.5 \text{ K ?)}$
Specific heat jump at T_c	$\Delta C / \gamma_n T_c \approx 0.25$
Upper critical field (small anisotropy)	$H_{c2}(0) \sim 3 \text{ T}$
Thermodynamic critical field	$H_c(0) = 26 \text{ mT}$
Ginzburg-Landau coherence length	$\xi(0) \sim 104 \text{ \AA}$
Ginzburg-Landau parameter	$\kappa = 82$
London penetration depth	$\lambda(0) \approx 0.86 \mu\text{m}$
Nodal structure	Line nodes

inversion symmetry that superconducts at ambient pressure, as opposed to the cases of CeIrSi₃, CeRhSi₃, CeCoGe₃, CeIrGe₃ and UIr. The heavy-fermion character has been established from the large Sommerfeld coefficient $\gamma_n \approx 0.39 \text{ J/K}^2 \text{ mol}$ [1]. The coexistence of antiferromagnetism and superconductivity has been proved by zero-field muon-spin relaxation [2] and neutron scattering [3]. Although CePt₃Si has been intensively studied both theoretically and experimentally many questions regarding its superconducting state remain unresolved, in part because the observed properties have some sample dependence. We will focus here on the superconducting properties of CePt₃Si, giving only a brief review on normal state and magnetic behaviors (a detailed review of these can be found in Refs. [4, 5]).

2.1.1 Crystal Structure, Sample Growth and Characteristic Parameters

CePt₃Si crystallizes in a tetragonal crystal structure with space group $P4mm$ (No. 99) without inversion symmetry [1]. The lattice parameters are listed in Table 2.1. The unit cell has one formula unit with one Ce, one Si and two Pt inequivalent sites. The absence of inversion symmetry comes from the missing mirror plane $(0, 0, \frac{1}{2})$ (see Fig. 2.1). The antiferromagnetic lattice has an ordered wave vector

Fig. 2.1 Crystal and magnetic structures of CePt_3Si



(0,0,1/2), with a magnetic moment oriented ferromagnetically along the axis [100] and antiferromagnetically along the axis [001], as indicated in Fig. 2.1.

The melting temperature of CePt_3Si is about 1390 °C [6]. An isothermal section of the Ce-Pt-Si phase diagram at 600 °C was presented by Gribanov et al. [6], who indicated that the interaction of Ce, Pt and Si leads to the formation of at least nine stable ternary phases. Seven of these ternary phases have a fixed composition. Polycrystalline samples of CePt_3Si have been grown by argon arc melting and high-frequency melting and single crystals by the Bridgman and high-frequency techniques. These samples are usually annealed under high vacuum around 900 °C for 2–3 weeks. Interestingly, the annealing process has been linked to a Si excess [7]. Growing very high quality single crystals of CePt_3Si has taken a long path that has led to the resolution of most of the problems in identifying the true superconducting properties of this compound.

2.1.2 Normal State

2.1.2.1 Phase Diagram and Magnetic Properties

Figure 2.2 shows the temperature-pressure phase diagram of CePt_3Si determined by specific heat, resistivity and ac magnetic susceptibility measurements [8]. The antiferromagnetic T_N and superconducting T_c transition temperatures decrease with increasing pressure and become zero around $P_{AF} = 0.7 \text{ GPa}$ and $P_c = 1.6 \text{ GPa}$, respectively.

The phase diagram indicates that there are two distinct superconducting phases: one below P_{AF} coexisting with the AFM phase and another above P_{AF} being presumably the only ordered phase. The coexistence of the superconducting and AFM phases was confirmed by neutron-scattering measurements that clearly show two superlattice peaks below and above the superconducting critical temperature (see Fig. 2.3) [3]. The observed peaks (0 0 1/2) and (1 0 1/2) correspond to an AFM vector $Q_0 = (0 \ 0 \ 1/2)$. The magnetic structure consists of ferromagnetic sheets of rather small Ce moments of $0.16 \mu_B/\text{Ce}$ stacked antiferromagnetically along the

Fig. 2.2 Temperature–pressure phase diagram of CePt_3Si showing the coexistence of the antiferromagnetic and superconducting phases for pressures below 0.7 GPa [8]

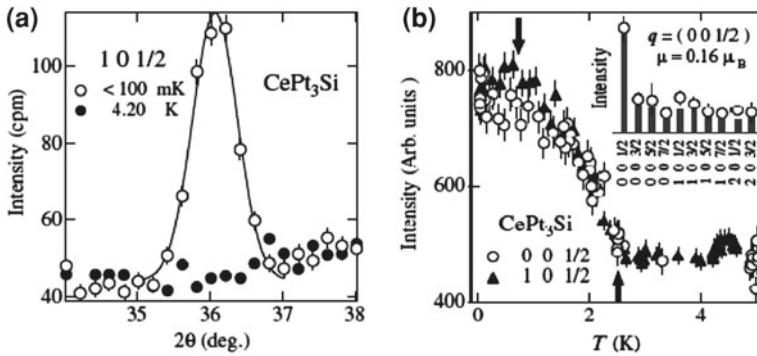
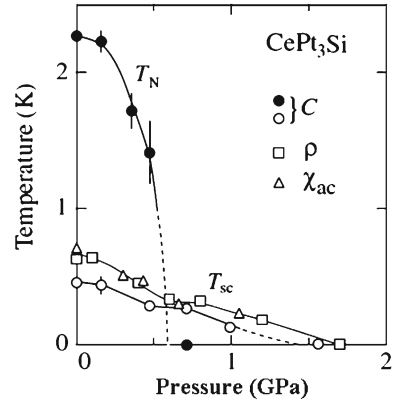


Fig. 2.3 **a** (101/2) AFM Bragg reflection observed below 0.1 K (*open circles*) and the background measured at 4.2 K (*solid circles*). **b** The intensity of (001/2) and (101/2) magnetic reflections as a function of temperature, shown by *open circles* and *solid triangles*, respectively. Up and down pointing arrows indicate T_N and T_c , respectively [3]

c axis (see Fig. 2.1). Here, μ_B is the Bohr magneton. The small value of the moment relative to $2.54 \mu_B/\text{Ce}$ of Ce^{3+} may partially be explained through the itinerant character of Ce $4f$ -electrons involved in the formation of the heavy quasiparticles. In parts the reduction of the moment is also due to the Kondo screening effect viewing these electrons as almost localized moments [1]. In general, the magnetic response of CePt_3Si in the normal state involves also the interplay of crystal electric field splitting of the $4f$ -orbitals and Kondo interaction (see, for example, Ref. [5]).

2.1.3 Superconducting State

At ambient pressure superconductivity in CePt_3Si appears within a Fermi-liquid state, as evidenced by quantum oscillations [9] and resistivity measurements [1, 10].

However, CePt₃Si becomes a non-Fermi-liquid under pressure, as indicated by the linear temperature behavior of the resistivity above 0.4 GPa [10].

Most of the unusual superconducting properties found initially in CePt₃Si have been clarified by now, but others have appeared. First results, like second anomalies or a small peak just below the superconducting transition in the NMR $1/T_1 T$, are not observed in the latest measurements carried out on new single crystals. These early results were associated with sample dependence: sample preparation, off-stoichiometry, impurity phases and/or annealing conditions. The new puzzling feature is a transition temperature that falls sometimes below 0.5 K [11–14].

Several theoretical approaches have been developed to try to understand this superconductor [15–19]. The models take into account the splitting of the spin-degenerate bands caused by the absence of inversion symmetry. Antiferromagnetic order effects in the heavy-fermion superconductors are also considered [18, 19]. Even though much experimental and theoretical efforts have been dedicated to clarify its physics, CePt₃Si continues to be the most interesting and challenging of all superconductors without spatial inversion symmetry.

2.1.3.1 Probing the Pairing Symmetry

Several techniques have been employed to test the Cooper pairing state in non-centrosymmetric CePt₃Si. We will review what has been done thus far.

Spin State

One of the most direct probes of the spin state of the pairing is the ratio of the superconducting to the normal electron-spin paramagnetic susceptibility, χ_s and χ_n , respectively, which for a spin-singlet pairing state may be written as

$$\frac{\chi_s}{\chi_n} = -2 \int_0^\infty d\xi \left\langle \frac{\partial f(E)}{\partial E} \right\rangle_{\hat{k}}. \quad (2.1)$$

The integration is over ξ , the energy of the free electrons relative to the Fermi level, f is the Fermi distribution function, and $E = \sqrt{\xi^2 + \Delta(\hat{k})^2}$ is the energy of the quasiparticles. The bracket $\langle \cdots \rangle_{\hat{k}}$ denotes the angular average. Here, $\Delta(\hat{k})$ is the energy gap that in general depends on the momentum direction $\hat{k}$ and temperature. For $T \ll T_c$ the ratio χ_s/χ_n goes as $(1/\sqrt{T}) \exp(-\Delta_0/k_B T)$ for an s -wave pairing state with an isotropic gap and as T^2 for a d -wave pairing state where the gap has line nodes (Fig. 2.4(a)). Δ_0 is the zero-temperature value of Δ and k_B is the Boltzmann constant.

For spin-triplet pairing the ratio χ_s/χ_n is more complicated and depends on the field orientation. In the most simple cases, the susceptibility does not change across the transition if the field is perpendicular to the $\mathbf{d}$ vector denoting the spin-triplet gap function, but decreases continuously down to zero at $T = 0$ if the applied field is parallel to the $\mathbf{d}$ vector (Fig. 2.4(b)).

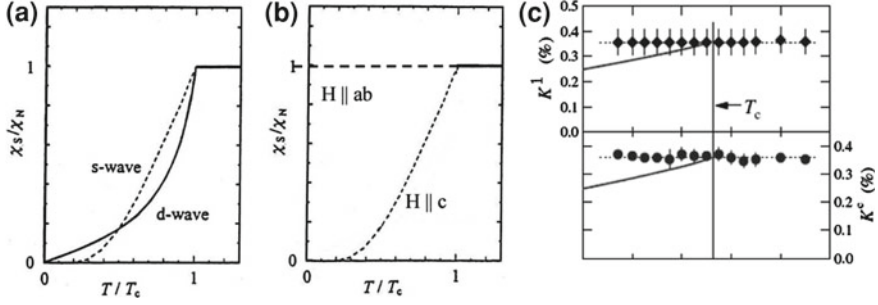


Fig. 2.4 Electronic spin susceptibility expected in **a** spin-singlet states *s*-wave and *d*-wave and **b** spin-triplet states. **c** Experimental electronic spin susceptibility of CePt₃Si showing no change across the superconducting transition for all orientations of the applied magnetic field (*upper panel*: $\mathbf{H} \perp \hat{z}$ and *lower panel* $\mathbf{H} \parallel \hat{z}$) [20]

In CePt₃Si the experimental result of χ_s/χ_n is puzzling: χ_s/χ_n does not change at all in the superconducting phase for any orientation of the field as shown in Fig. 2.4(c) [20]. Model calculations including a sizable ASOC characteristic for the non-centrosymmetric CePt₃Si predict that for both spin-singlet and spin-triplet states $\chi_s(0)/\chi_n \rightarrow 1$ for fields parallel to the *z* axis and $\chi_s(0)/\chi_n \rightarrow 1/2$ for fields perpendicular to *z* [21, 22]. On the other hand, if electron correlations are included, χ_s/χ_n can be calculated to be constant across the transition independently of the field orientation [18, 19]. Thus, in CePt₃Si spin-susceptibility measurements are not quite useful to distinguish between spin-singlet and spin-triplet pairings.

The upper critical field H_{c2} can also be used to get information about the spin configuration of a pairing state. A magnetic field induces pair breaking via paramagnetic and orbital mechanisms. The Pauli paramagnetic limiting field H_P can be estimated by comparing the (zero-field) superconducting condensation energy with the Zeeman energy

$$\frac{1}{2}(\chi_n - \chi_s)H_P^2 = \frac{1}{2}N_0\Delta_0^2, \quad (2.2)$$

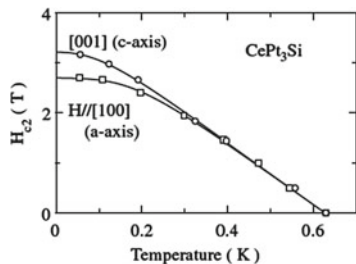
where N_0 is the density of states at the Fermi energy. The Pauli susceptibility χ_n is given by $\chi_n = (g\mu_B)^2 N_0/2$, where g is the gyromagnetic ratio. H_P is then derived as

$$H_P = \frac{\sqrt{2}\Delta_0}{g\mu_B\sqrt{1 - \chi_s/\chi_n}}. \quad (2.3)$$

As discussed above, for spin-singlet superconductors χ_s goes to zero as $T \rightarrow 0$. Then, using the BCS value $\Delta_0 = 1.76k_B T_c$ for a weak-coupling superconductor and $g = 2$ for free electrons, we obtain the well-known estimate

$$H_P^{BCS} = H_P(0) = 1.86T_c[\text{T/K}]. \quad (2.4)$$

Fig.2.5 Temperature dependence of the upper critical field in CePt₃Si showing a weakly anisotropic behavior [10]



This expression is also valid for spin-triplet superconductors if the field is applied parallel to the $\mathbf{d}$ vector. On the other hand, if the field is applied perpendicular to the $\mathbf{d}$ vector $\chi_s = \chi_n$ as $T \rightarrow 0$ and Eq. (2.3) yields $H_P \rightarrow \infty$. This would imply the absence of the Pauli paramagnetic limiting effect for fields along the ab plane.

Measurements on high-quality single crystals of CePt₃Si show a weak anisotropy for the upper critical field $H_{c2}(0)$ with a value around 3 T (Fig. 2.5) [10] that exceeds the standard BCS weak-coupling paramagnetic limit $H_P(0) \approx 1$ T.

From Eq. (2.3) and the predictions for χ_s/χ_n in the non-centrosymmetric superconductors, no limiting behavior is expected for fields parallel to the c axis, whereas $H_P(0) = \Delta_0/\mu_B \approx 1.4$ T for fields in the ab plane. As mentioned above, modifications of χ_s due to correlation effects as well as magnetic ordering could eliminate the paramagnetic limiting for all field directions [18, 19]. It was also suggested that the realization of a so-called helical phase for fields perpendicular to the c axis would strongly reduce paramagnetic limiting effects [23, 24].

The orbital limiting field H_{orb} is expressed by

$$H_{orb}(T) = \frac{\Phi_0}{2\pi\xi^2(T)}, \quad (2.5)$$

where Φ_0 is the flux quantum. $H_{orb}(T=0)$ can be in principle obtained by using the BCS expression $\xi(0) = 0.18\hbar v_F/(k_B T_c)$, where v_F is the Fermi velocity. However, $H_{orb}(T=0)$ is usually estimated from the formula [25]

$$H_{orb}(T) = h(T)H'_{c2}T_c. \quad (2.6)$$

Here, $H'_{c2} \equiv -dH_{c2}/dT|_{T=T_c}$. $h(0) = 0.727$ for weak-coupling BCS superconductors in the clean limit. Using the data of Fig. 2.5 we estimate $H'_{c2} \approx -6.8$ T/K and from Eq. (2.6) $H_{orb}^{BCS} \approx 3.7$ T. This is about the value of $H_{c2}(0)$ for both field orientations, which suggests that CePt₃Si may be restricted by the orbital depairing limit. In such a case the spin-triplet state should be favorable. Both paramagnetic and orbital mechanisms will be discussed in more detail in Sect. 2.2.

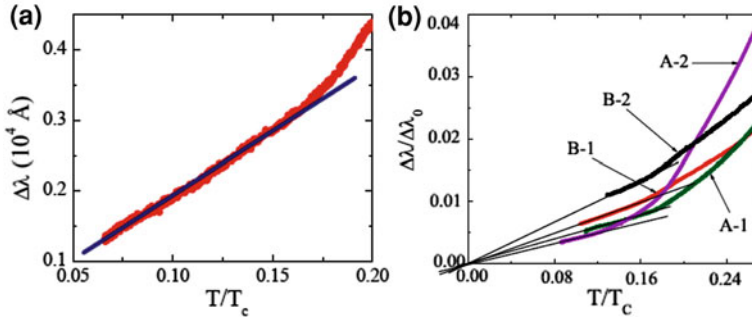


Fig. 2.6 Penetration depth in **a** a polycrystalline sample and **b** single crystals of CePt_3Si . The linear temperature behavior indicates line nodes in the energy gap. The single crystals used in the penetration depth measurements shown in **b** have different defect concentrations, which suggests that the linear behavior is unaffected by disorder [13]

Nodal Structure

The structure of the energy gap is directly related to the symmetry of the Cooper pairing. The energy gap is isotropic for s -wave spin-singlet superconductors and usually has zeroes (nodes) for other symmetries. Thus, the determination of the presence of nodes in the energy gap is crucial to establish pairing with symmetries lower than the s -wave. The existence of nodes in the energy gap leads to low-temperature power laws (T^n) in several superconducting properties, instead of the BCS exponential temperature response observed for an isotropically gapped excitation spectrum. In CePt_3Si magnetic penetration-depth, thermal-conductivity and specific-heat measurements show power-law behaviors indicative of line nodes in the energy gap.

A linear temperature dependence of the magnetic penetration depth $\lambda(T)$ below $0.16 T_c$ was first found in a polycrystalline sample of CePt_3Si (Fig. 2.6(a)) [26], and later also in single crystals [13]. In the local limit of the electrodynamics of superconductors, the magnetic penetration depth is given by

$$\left[\frac{\lambda^2(0)}{\lambda^2(T)} \right]_{ij} = \frac{n_{ij}^s(T)}{n} = 3 \left\langle \hat{k}_i \hat{k}_j \left[1 - \int d\xi \left(\frac{-\partial f}{\partial E_{\mathbf{k}}} \right) \right] \right\rangle_{\hat{k}}. \quad (2.7)$$

In superconductors with inversion symmetry $\Delta\lambda(T) \propto T$ is expected in the low-temperature limit for an energy gap with line nodes [27]. Thus, the linear response in CePt_3Si was taken as evidence for line nodes. Surprisingly, the temperature response in the penetration depth is not affected by the sample quality, as opposed to what occurs in unconventional superconductors with line nodes in the gap [13]. Figure 2.6(b) displays the low-temperature region of the penetration depth of several single crystals of different quality. A clear linear behavior is observed in all of them. We note that the superfluid density $\rho_s(T)$ shows a small anisotropy [28], in agreement with the upper critical field result.

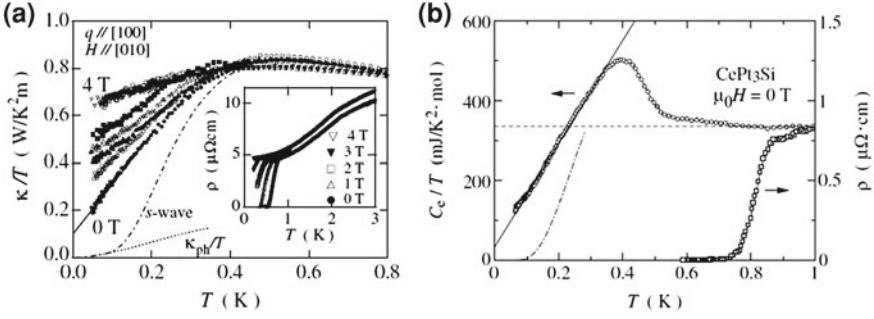


Fig. 2.7 Temperature dependence of **a** thermal conductivity [29] and **b** specific heat [30] of CePt₃Si. The linear temperature response suggests line nodes in the energy gap

Thermal transport measurements also suggest the presence of line nodes by showing a residual linear term in $\kappa(T)/T$ as $T \rightarrow 0$ (Fig. 2.7(a)). In superconductors with inversion symmetry such a linear term is expected when the energy gap has nodes, and is due to impurity scattering. The quasiparticle thermal conductivity has universal components in the low-temperature limit ($k_B T < \gamma$)

$$\kappa_{ii} = \frac{\pi^2}{3} N_0 v_F^2 T \left\langle \hat{k}_i \hat{k}_i \frac{\gamma^2}{(\gamma^2 + \Delta_{\mathbf{k}}^2)^{3/2}} \right\rangle_{\hat{k}} \quad (2.8)$$

that are linear functions of temperature at low T and whose proportionality constants depend on the specific form of the order parameter [27]. γ is the quasiparticle decay rate. In CePt₃Si the experimental $\kappa(T)/T = 0.1 \text{ W}/(\text{K}^2 \cdot \text{m})$ is in good agreement with the calculated universal conductivity limit $0.09 \text{ W}/(\text{K}^2 \cdot \text{m})$ [29]. Moreover, the H dependence of κ follows the prediction by a theory of Doppler-shifted quasiparticles in a superconductor with line nodes [29, 31].

In the low-temperature limit the electronic specific heat of CePt₃Si has been found to follow the expression $C_{el}/T = A + BT$, with $A = 34.1 \text{ mJ}/(\text{K}^2 \cdot \text{mol})$ and $B = 1290 \text{ mJ}/(\text{K}^3 \cdot \text{mol})$ (Fig. 2.7(b)) [30]. In general, the electronic specific heat is given by [27]

$$C_{el} = 2N_0 \int_{-\infty}^{\infty} d\xi \left\langle E_{\mathbf{k}} \frac{\partial f}{\partial E_{\mathbf{k}}} \right\rangle_{\hat{k}}. \quad (2.9)$$

For superconductors with inversion symmetry, in the low-temperature limit $C_{el} \propto T^2$ for a gap with line nodes. The residual linear term in C_{el}/T of CePt₃Si is considered to be caused by impurities or by electrons on part of the Fermi surface that do not participate in superconducting. Thus, the observed behavior of the electronic specific heat is taken as evidence for line nodes in the energy gap [30].

For a theoretical discussion on line nodes in non-centrosymmetric superconductors, see other chapters in this book.

2.2 CeTX₃ Compounds

2.2.1 Crystal Structure and Related Compounds

Most CeTX₃ compounds crystallize in the BaNiSn₃-type tetragonal structure with space group *I4mm* (No.107) [32]. The BaNiSn₃-type structure derives from the BaAl₄-type structure whose basic frame is the body-centered tetragonal lattice shown at the top in Fig. 2.8. There are two other derivatives of the BaAl₄-type structure, the ThCr₂Si₂ and CaBe₂Ge₂ types. Some heavy-fermion superconductors crystallize into the former structure: e.g. CeCu₂Si₂ [33], CeCu₂Ge₂ [34], CePd₂Si₂ [35], CeRh₂Si₂ [36] and URu₂Si₂ [37]. The latter structure is often found in RPt₂X₂ compounds [38], where *R* denotes a rare-earth element. The ThCr₂Si₂- and CaBe₂Ge₂-type structures have an inversion center, while the BaNiSn₃-type structure does not. Fig. 2.8 displays the BaAl₄-type crystal lattice and its three derivatives.

The atomic framework of the BaNiSn₃-type structure can be alternatively displayed as a sequence of planes of the same atoms $R - T - X(1) - X(2) - R - T - X(1) - X(2) - R$ along the *c* axis of the tetragonal structure, where *T* and *X* denote a transition metal and Si/Ge, respectively. The point group of the BaNiSn₃-type structure is C_{4v}, which lacks the mirror plane and a two-fold axis normal to the *c* axis (*z* axis). Therefore, a Rashba-like spin-orbit coupling exists in this system, as it does in CePt₃Si that also belongs to C_{4v} [1].

Of the CeTX₃ compounds, the series CeT₃Si₃ (*T* = Co, Ru, Rh, Pd, Os, Ir and Pt) and CeTGe₃ (*T* = Fe, Co, Rh and Ir) are known to crystallize in the BaNiSn₃-type structure. Among these, CeRhSi₃ [39], CeIrSi₃ [40], CeCoGe₃ [41] and CeIrGe₃ [42] have been found to be pressure-induced superconductors. Single crystals of CeRhSi₃ and CeIrSi₃, as well as of CeRuSi₃ [43], can be obtained by the Czochralski pulling method in a tetra-arc furnace using a pulling speed of 10 mm/h or 15 mm/h. In these compounds annealing treatments at 900 °C, in vacuum, for a week are usually very effective. Notably, for CeRhSi₃ the use of stoichiometric amounts of the components sometimes yields a different crystal; e.g., CeRhSi₂. To obtain a crystal of CeRhSi₃ an off-stoichiometric composition, typically Ce:Rh:Si = 1:1:3.3, works better. Single crystals of CeCoGe₃ are obtained by the Bi-flux method (the Czochralski method is unsuitable) [44]. In this procedure, arc-melt-prepared ingots of CeCoGe₃ and Bi are placed in an alumina crucible and heated up to 1050 °C in an argon atmosphere. After keeping this temperature for a day, the crucible is cooled down to 650 °C over a period of two weeks and then down to room temperature rapidly. Single crystals of CeRhGe₃ can also be obtained by the Bi-flux technique [43]. Single crystals of CeIrGe₃ are grown by the Bi- and Sn-flux methods [42, 43].

Other CeTX₃ compounds with BaNiSn₃-type structure are CeTAl₃ (*T* = Cu, Au) and CeAuGa₃. They order magnetically at low temperatures [45, 46, 47, 48, 49]. In particular, CeCuAl₃ and CeAuAl₃ are suggested to be HF compounds with AFM ground states [46] and have the potential to become non-centrosymmetric HF superconductors. There are at least two CeTX₃ compounds that do not have the BaNiSn₃-type crystal lattice: CeNiGe₃, which has an orthorhombic structure and

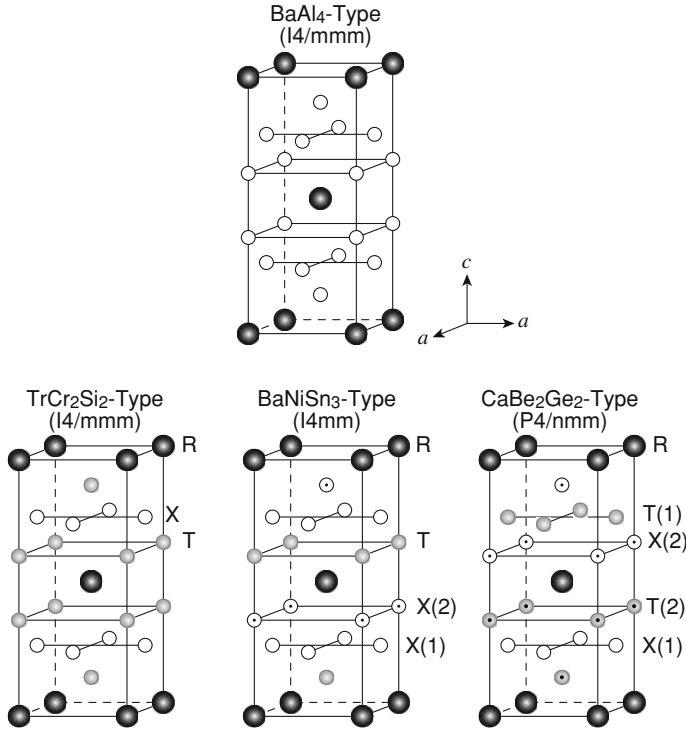


Fig. 2.8 BaAl₄-type crystal structure and its three derivatives TrCr₂Si₂, BaNiSn₃ and CaBe₂Ge₂. Only the BaNiSn₃-type structure does not have an inversion center

exhibits superconductivity under pressure [50], and CeRuGe₃, which is reported to have either orthorhombic [51] or cubic structure [52].

On the other hand, no uranium-based compound (UTX₃) has been fully confirmed to have the BaNiSn₃-type structure. Only UNiGa₃, that exhibits AFM order at 39 K, seems to have this structure [53].

Last, we briefly comment on non-heavy-fermion LaTX₃ compounds. Some of them, like LaRhSi₃, LaIrSi₃ and LaPdSi₃, also have BaNiSn₃-type structures and show superconductivity at the critical temperature 1.9, 0.9 and 2.6 K, respectively [32, 54, 55]. In these materials, however, no evidence for unconventional behavior has been observed [54].

2.2.2 Normal State

2.2.2.1 Magnetic Properties

The CeTX₃ compounds exhibit various magnetic ground states as summarized in Table 2.2. The magnetic ground states vary from AFM to intermediate valence

Table 2.2 Unit-cell volume (V), magnetic ground state, electronic specific-heat coefficient γ_n , ordering temperature (T_N), Weiss temperature (Θ_p) and effective moment (μ_{eff}) for the BaNiSn₃-type CeTX₃ compounds

Compound	a [Å]	c [Å]	V [Å ³]	Magnetism	γ_n [mJ/mol·K ²]	T_N [K]	Θ_p [K]	μ_{eff} [μ _B]	Ref.
<i>*CeT</i> Si ₃									
CeCoSi ₃	4.135	9.567	163.6	PM(IV)	37	—	−840	2.80	[59]
CeRuSi ₃	4.21577	9.9271	176.43	PM(IV)		—			[43]
CeRhSi ₃	4.269	9.738	177.5	AFM(HF)	110	1.6	−128	2.65	[60, 55]
	4.237	9.785	175.7						
CePdSi ₃	4.33	9.631	180.6	AFM	57	5.2/3	−26	2.56	[62, 55]
CeOsSi ₃				PM(IV)					[63]
CeIrSi ₃	4.252	9.715	175.6	AFM(HF)	120	5.0	−142	2.48	[60, 55]
CePtSi ₃	4.3215	9.6075	179.42	AFM	29	4.8/2.4			[64]
<i>*CeT</i> Ge ₃									
CeFeGe ₃	4.332	9.955	186.8	PM(HF)	150	—	−90	2.6	[65]
	4.3371	9.9542	187.24						
CeCoGe ₃	4.320	9.835	183.5	AFM	32	21/19	−51	2.54	[59]
	4.319	9.829	183.3			21/12/8			
CeRhGe ₃	4.402	9.993	193.6	AFM	40	14.6/10/0.55	−28	2.53	[60]
	4.3976	10.0322	194.01			14.9/8.2			
CeIrGe ₃	4.409	10.032	195.0	AFM	80	8.7/4.7/0.7	−21	2.39	[60]
	4.401	10.024	194.2			8.7/4.8			

The abbreviations PM and AFM denote paramagnetic and antiferromagnetic ground states, respectively. The abbreviations IV and HF denote intermediate-valence and heavy-fermion states, respectively. We consider compounds with $\gamma_n > 100$ mJ/mol·K² to be heavy-fermion systems

(IV) through HF states with decreasing unit-cell volume V . For example, CeCoGe₃ ($V = 183.3 \text{ Å}^3$) displays an AFM ground state [44, 56, 57], while CeCoSi₃ ($V = 163.6 \text{ Å}^3$) is thought to be an IV compound [58].

Figure 2.9 shows the Néel temperature T_N and the electronic specific-heat coefficient γ_n as a function of unit-cell volume for CeTX₃ in which T belongs to the Group 9 (Co, Rh and Ir) in the Periodic Table [43]. T_N approximately follows a simple curve which peaks at 186 Å^3 . This behavior supports the Doniach model in which the on-site Kondo effect dominates over the inter-site RKKY interaction with the coupling constant J being effectively enhanced relative to the kinetic energy with decreasing unit-cell volume [43]. γ_n is also described by a simple curve which peaks at the unit-cell volume 176 Å^3 at which T_N goes to zero, suggesting that the γ_n value is enhanced by the magnetic fluctuation arising at the corresponding volume.

Besides the Group 9 (Co, Rh, Ir) compounds, one can consider CeFeGe₃ to be a potential superconductor because its γ_n is comparable to those of CeRhSi₃ and CeIrSi₃. However, in CeFeGe₃ superconductivity has not been observed down to 0.05 K [65].

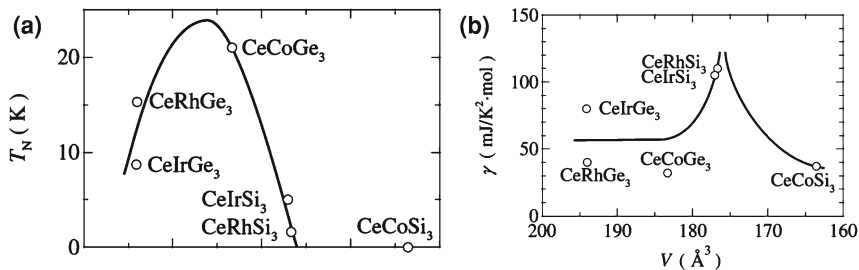


Fig. 2.9 Unit-cell volume dependence of **a** the Néel temperature and **b** the γ_n value in CeTGe_3 and CeTGe_3 (T: Co, Rh, Ir) [43]

CeRhSi₃

As shown in Figs. 2.10(a) and 2.11(d), the magnetic properties of CeRhSi_3 are anisotropic especially at low temperatures [55, 67]. The induced magnetization along the easy axis [100] has a quite small value of $0.1 \mu_B$ at 7 T. The magnetic susceptibility curves for H parallel to the a and c axes show a strong anisotropy at low temperatures, while they obey the Curie-Weiss law above about 150 K. The effective moments μ_{eff} for both field directions are $2.65 \mu_B$, which is close to the expected value for the Ce^{3+} ion. The Weiss temperatures are negative and very large (-112 and -160 K for H parallel to the a and c axes, respectively), as often found for IV compounds. The susceptibility for $H \parallel c$ has a broad peak around 50 K that is characteristic of HF compounds.

From the jump of the specific heat at T_N , the magnetic entropy gain is estimated to be only 12% of $R \ln 2$ [60]. The AFM state is robust against a magnetic field (Fig. 2.12(a)) and survives up to 8 T, although such a strong field should be sufficient to suppress an AFM state with a low T_N of the order of 1 K. The temperature and height of the specific-heat peak decrease with increasing field along the easy axis ($H \parallel a$). The magnetic contribution to the specific heat when the field is aligned with the hard axis ($H \parallel c$) does not change even at 8 T [67]. The tiny entropy gain and the insensitivity to a magnetic field are attributed to the strong Kondo screening of the $4f$ electron. The Kondo temperature is estimated to be about 50 K [60].

The magnetic structure at ambient pressure is revealed by neutron experiments to be a longitudinal spin-density-wave (LSDW) type characterized by the propagation vectors $Q = (\pm 0.215, 0, 0.5)$ with polarization along the a^* axis [69]. The magnetic structure is shown in Fig. 2.13. The staggered moment $0.13 \mu_B$ is quite small [70]. The incommensurate LSDW structure with such a strongly suppressed moment suggests that itinerant-electron magnetism is realized in CeRhSi_3 . The $4f$ electrons are expected to be strongly hybridized with the conduction electrons through the Kondo effect, leading to the formation of the SDW state caused by nesting of the Fermi surface.

It is not obvious whether this magnetic structure persists under pressure. In the pressure-dependent specific-heat curve, a shoulder-like transition is seen below T_N at a pressure of 0.55 GPa [71]. The origin of the transition is unclear at present.

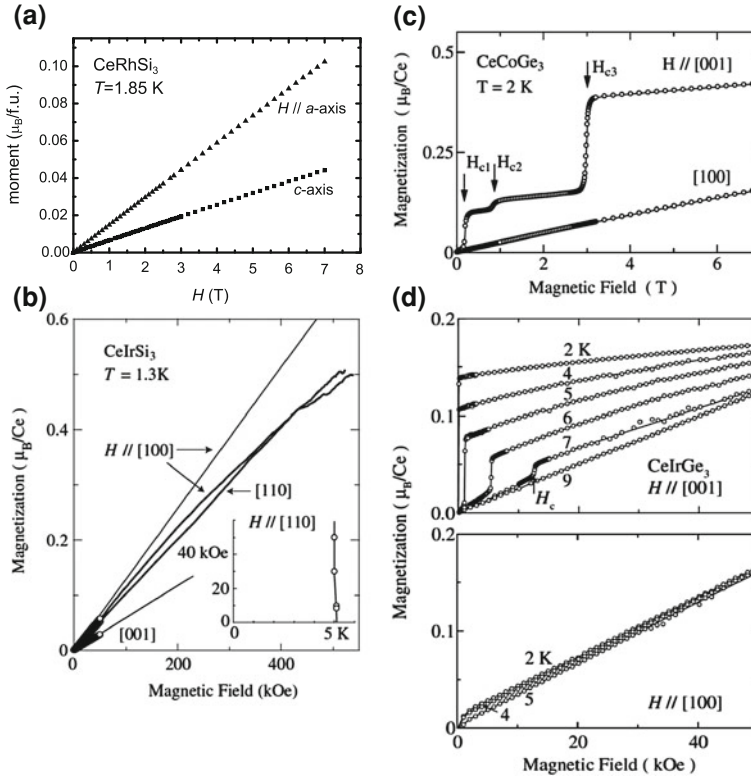


Fig. 2.10 Magnetization curves of CeRhSi_3 , CeIrSi_3 , CeCoGe_3 and CeIrGe_3 [44, 55, 68]

Considering that multiple magnetic transitions are observed in other magnetic CeTX_3 compounds, the magnetic structure realized in CeRhSi_3 at ambient pressure may change under pressure where superconductivity appears.

The electrical resistivity below T_N can be fitted by an antiferro-magnon model [72]. At sufficiently low temperatures ($T < 0.6$ K), the resistivity follows the Fermi-liquid description of $\rho(T) = \rho_0 + AT^2$, where ρ_0 is the residual resistivity. $A = 0.19 \mu\Omega \cdot \text{cm}/\text{K}^2$ for the current J along the a axis and $A = 0.24 \mu\Omega \cdot \text{cm}/\text{K}^2$ for $J \parallel c$. The ratios $A/\gamma_n^2 = 1.6 \times 10^{-5}$ ($J \parallel a$), 2.0×10^{-5} ($J \parallel c$) $\mu\Omega \cdot \text{cm} \cdot \text{K}^2 \cdot \text{mol} \cdot \text{mJ}^{-2}$ are close to $1 \times 10^{-5} \mu\Omega \cdot \text{cm} \cdot \text{K}^2 \cdot \text{mol} \cdot \text{mJ}^{-2}$ as given by the Kadowaki-Woods relation [73].

CeIrSi_3

Although the magnetic structure of CeIrSi_3 is unknown at present, the specific heat, magnetic susceptibility and electrical resistivity are similar to those of CeRhSi_3 . The magnetization curve is anisotropic and a is the easy axis (Fig. 2.10(b)) [68]. The mag-

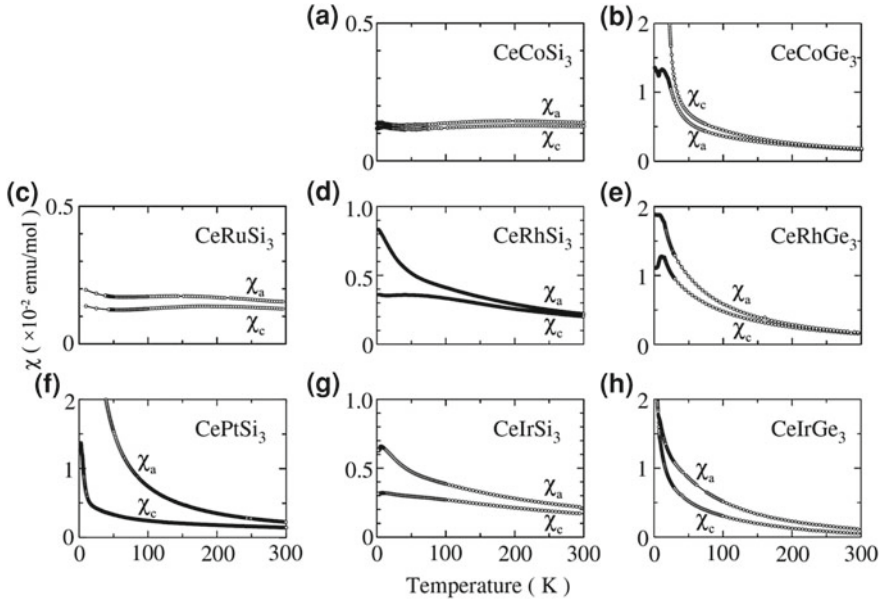


Fig. 2.11 Magnetic susceptibility of CeTX_3 compounds as a function of temperature [43]

netization increases almost linearly with magnetic field. Neither a saturation behavior nor metamagnetic transitions are observed for fields in the basal plane up to 50 T. The induced magnetization is very small and comparable to that of CeRhSi_3 ; $0.1\mu_B$ at 10 T for the easy axis. The temperature dependence of the magnetic susceptibility is anisotropic at low temperatures (Fig. 2.11(g)) [40, 68].

The entropy gain associated with the AFM transition is small, $0.2R \ln 2$, as in CeRhSi_3 (Fig. 2.12(b)) [68]. The AFM state is robust against a magnetic field, as shown in the inset of Fig. 2.10(b). The electrical resistivity below T_N can be fitted by an antiferro-magnon model as done in CeRhSi_3 . The coefficient of the T^2 term $A = 0.04 \mu\Omega \cdot \text{cm}/\text{K}^2$ for $J \parallel c$ and $J \perp c$ [68] is about one fifth of the coefficients of CeRhSi_3 . The ratio $A/\gamma_n^2 = 3.6 \times 10^{-6} \mu\Omega \cdot \text{cm} \cdot \text{K}^2 \cdot \text{mol} \cdot \text{mJ}^{-2}$ is much smaller than the ratios in CeRhSi_3 but still in the range of the Kadowaki-Woods relation.

CeCoGe₃

Unlike CeRhSi_3 and CeIrSi_3 , CeCoGe_3 exhibits three successive AFM transitions [44]. Correspondingly, the magnetization for $H \parallel c$ shows three-step metamagnetic transitions. It reaches $M_s/4$, $M_s/3$ and M_s , where $M_s = 0.43\mu_B/\text{Ce}$, at each transition. The anisotropic magnetization curve indicates an Ising-like magnetism

Fig. 2.12 **a** Magnetic contribution to the specific heat C_{mag} of CeRhSi_3 as a function of temperature for fields along the a and c axes [67]. **b** C_{mag}/T and the entropy S of CeIrSi_3 as a function of temperature [68]

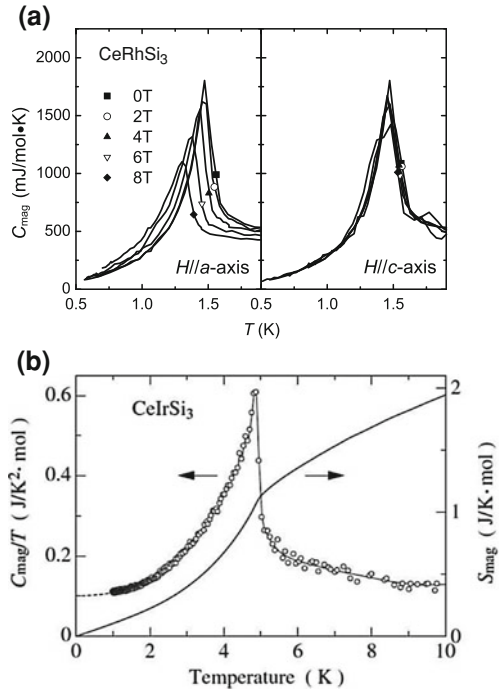
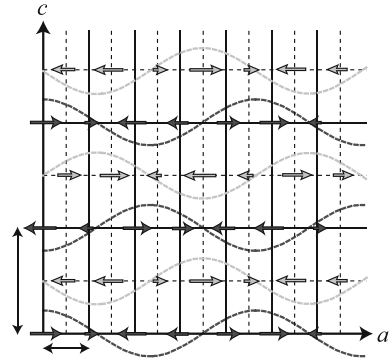


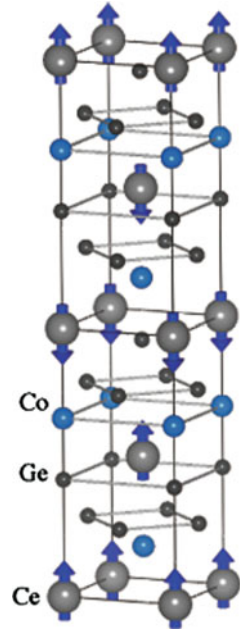
Fig. 2.13 Magnetic structure of CeRhSi_3 . Only the Ce sites at $(0, 0, 0)$ and $(0.5, 0.5, 0.5)$ positions are projected on the ac plane. The arrows are depicted to show the size and the direction of the magnetic moment at the Ce sites [70]



with the easy axis along the c axis, which is different from what is found in CeRhSi_3 and CeIrSi_3 as seen in Fig. 2.11.

The neutron-diffraction experiments revealed that the magnetic structure of the ground state at ambient pressure consists of two components with dominant $q_1 = (0, 0, 1/2)$ and subordinate $q_2 = (0, 0, 3/4)$ [74]. Figure 2.14 shows a possible magnetic structure of the q_1 sublattice. The magnetic moments are parallel to the c axis and alternate in the up-up-down-down sequence. The magnitude of the mag-

Fig. 2.14 Possible magnetic structure for $q_1 = (0, 0, 0.5)$ sublattice in the ground state of CeCoGe_3 [74]



netic moment in the sublattice μ_1 is estimated to be $0.5(1)\mu_B$. A more complete magnetic structure that includes the q_2 sublattice is not clear at present.

The magnetic susceptibility of CeCoGe_3 does not show a peak or a saturated behavior at low temperatures as observed in CeRhSi_3 and CeIrSi_3 . Specific heat measurements revealed that the entropy gain reaches 68% of $R \ln 2$ at $T_{N1} = 21$ K and that 32% of entropy loss is recovered at 38 K. The magnetism of CeCoGe_3 is basically understood in terms of localized $4f$ electron.

CeIrGe₃

The magnetic structure of CeIrGe_3 seems to be more complicated. There are two successive magnetic transitions at $T_{N1} = 8.7$ K and $T_{N2} = 4.8$ K. The former is antiferromagnetic and the latter is unknown although magnetization measurements indicate weak ferromagnetism with a small moment below T_{N2} (see Fig. 2.10(d)). A parasitic ferromagnetism due to the Dzyaloshinsky-Moriya interaction caused by the broken space inversion symmetry is discussed in Ref. [43]. T_{N2} merges into T_{N1} with the application of pressure (see Fig. 2.15(d)).

2.2.2.2 Temperature-Pressure Phase Diagram

All known CeTX_3 superconductors need pressure to become superconducting. The temperature-pressure (T - P) phase diagrams of CeRhSi_3 [72], CeIrSi_3 [75], CeCoGe_3

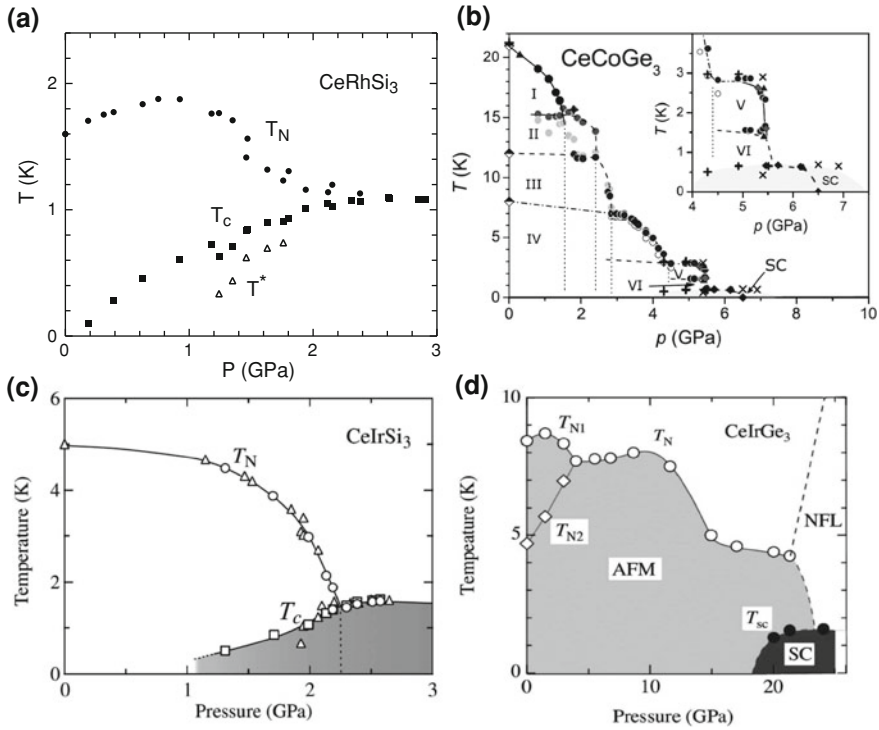


Fig. 2.15 Temperature-pressure phase diagrams of CeRhSi_3 [72], CeIrSi_3 [75], CeCoGe_3 [77] and CeIrGe_3 [42]. T^* in panel **a** denotes an anomaly observed in the resistivity and magnetic-susceptibility measurements [39]

[76] and CeIrGe_3 [42] are shown in Fig. 2.15. In CeRhSi_3 , the Néel temperature T_N initially increases and subsequently decreases with applying pressure, while those of CeIrSi_3 decreases monotonically with pressure. The $T_N(P)$ of CeCoGe_3 and CeIrGe_3 exhibits step-like decreases probably relevant to successive magnetic phase transitions observed at ambient pressure. The superconducting transitions of these four compounds are observed at pressures at which the AFM order still exists. We define here three characteristic pressures: P_1^* where $T_N = T_c$, P_2^* where $T_N \rightarrow 0$ and P_3^* where the superconducting transition temperature T_c reaches a maximum. The values of these pressures are summarized in Table 2.3. P_2^* of CeRhSi_3 is unclear because T_N does not decrease steeply near P_1^* .

The resistivity drop at the superconducting transition of CeRhSi_3 , CeIrSi_3 and probably CeCoSi_3 has its sharpest form at P_3^* . The resistivity drop in the AFM phase, especially far below P_3^* , is very broad. In this pressure region, the drop width depends on the applied current [39, 75]. These observations imply that superconductivity is inhomogeneous or fluctuating in the antiferromagnetic state and is optimum at P_3^* . Such a phenomenon is found in centrosymmetric HF superconductors as well

Table 2.3 Normal and superconducting parameters of CeTX_3 compounds

	CeRhSi ₃	CeIrSi ₃	CeCoGe ₃	CeIrGe ₃
Crystal structure			Tetragonal	
Space group			$I4mm$	
a [Å]	4.237	4.252	4.320	4.401
c [Å]	9.785	9.715	9.835	10.024
γ_n [mJ/mol·K ²]	110	120	32	80
m^* [m_0]	4 – 19	N/A	N/A	N/A
l [Å]	2400 – 3400	N/A	N/A	N/A
T_N [K]	1.6	5.0	21/12/8	8.7/4.8
$\mathbf{q}$	($\pm 0.215, 0, 0.5$)	N/A	(0,0,1/2) (0,0,3/4)	N/A
$\mathbf{m_Q}$ orientation	[001]	[001]	[100]	[100]?
μ_s [μ_B/Ce]	0.13	N/A	0.5	N/A
P_1^* (GPa)	2.4–2.5	2.25	5.5 – 5.6 ^d	> 21
P_2^* (GPa)	?	2.50	5.5 – 5.7 ^d	~ 24
P_3^* (GPa)	2.65	2.63	5.7 ^d or 6.5 ^e	≥ 24
$T_c @ P_3^*$ [K]	1.09	1.56 – 1.59	0.66 ^d or 0.69 ^e	≥ 1.6
$\Delta C/\gamma_n T_c$	N/A	5.7	N/A	N/A
$H_{c2}(0)$ [T] ($H \parallel c$)	30 ± 2^a	45 ± 10^c	$22 \pm 8^{f,g}$	27 ± 10
$H_{c2}(0)$ [T] ($H \perp c$)	7.5 ^b	9.5 ^c	N/A	N/A
$\xi(0)$ [Å] ($H \parallel c$)	~ 33	~ 27	~ 39	~ 35
H'_{c2} [T/K] ($H \parallel c$)	23 ^b	17 ^c	20 ^d	16 ^h
H'_{c2} [T/K] ($H \perp c$)	27 ^b	14.5 ^c	N/A	N/A

$\xi(0)$ is estimated from Eq. (2.5). Here, we regard $H_{c2}(0)$ for $H \parallel c$ as H_{orb} ; that is, the paramagnetic pair-breaking effect is absent. P_1^* , P_2^* and P_3^* are the pressures at which $T_N = T_c$, $T_N \rightarrow 0$ and T_c reaches a maximum, respectively. $\mathbf{q}$, $\mathbf{m_Q}$ and μ_s denote magnetic propagation vector, magnetic moment and value of ordered moment, respectively, and $H'_{c2} \equiv -dH_{c2}/dT|_{T=T_c}$.

^a at 2.85 GPa

^b at 2.6 GPa

^c at 2.65 GPa

^d determined from heat-capacity measurements[77].

^e determined from electrical-resistivity measurements [43].

^f at 6.5 GPa

^g estimated from Fig. 2.25.

^h at 24 GPa

and, thus, seems to be realized irrespective of the presence or absence of inversion symmetry.

A similar evolution of superconductivity in the AFM state is seen by the heat capacity C (Fig. 2.16). The heat capacity jump (ΔC) at the superconducting transition below P_1^* is small and broad, while it becomes sharper with increasing pressure and is strongly enhanced above P_1^* [75]. In CeIrSi₃, as shown in Fig. 2.16, $\Delta C/C_n$, where C_n is the normal state value just above T_c , reaches 5.7 ± 0.1 at 2.58 GPa, which is much larger than the 1.43 value expected from the weak-coupling

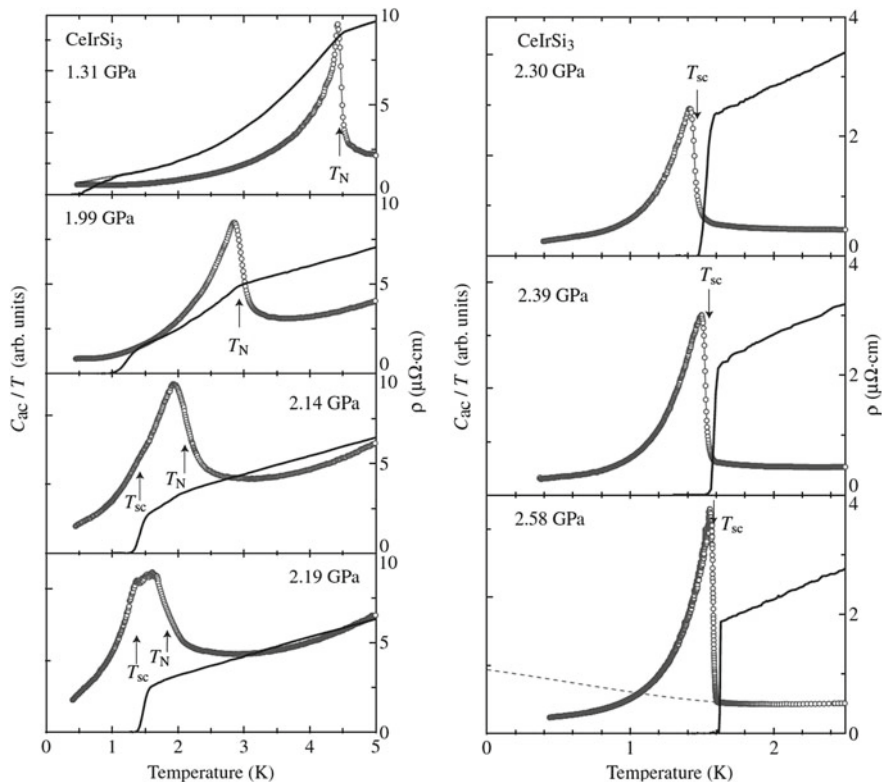


Fig. 2.16 Temperature dependence of the ac heat capacity C_{ac} (circles, left side) and the electrical resistivity ρ (lines, right side) at several pressures in CeIrSi₃. The dotted line in the panel at 2.58 GPa indicates the entropy balance below T_c [75]

BCS model and is probably the highest value among all known superconductors. On the other hand, the jump of the heat capacity associated with antiferromagnetism diminishes when approaching P_1^* , and is no longer observed above P_1^* . Apparently, the entropy gain of the AFM transition is transferred to the superconducting one.

2.2.2.3 Quantum Criticality and Non-Fermi Liquid

Superconductivity in f -electron materials often appears in the vicinity of a quantum critical point (QCP) at which the magnetic ordering temperature is reduced to zero by a nonthermal control parameter such as pressure, magnetic field, or chemical substitution. Often, a QCP accompanies the emergence of a non-Fermi liquid in which the temperature dependence of some physical properties obeys different behavior from that expected in the Fermi-liquid theory. As shown in Fig. 2.17, the electrical resistivity changes from the Fermi-liquid prediction $\rho(T) = \rho_0 + AT^2$ below P_1^* to $\rho(T) = \rho_0 + A'T$ above P_1^* in CeRhSi₃ and CeIrSi₃

[40, 72]. A similar crossover occurs in CeCoGe_3 at 6.9 GPa [43]. The T -linear dependence of the resistivity agrees with the prediction by the 2D spin fluctuation theory as indicated in Table 2.4. On the other hand, in CeIrSi_3 $1/T_1$ -NMR measurements in the normal state reveal a $\sqrt{T}$ dependence at 2.7 GPa for $H \perp c$. This supports a prediction for 3D AFM fluctuations in this system: $1/T_1 \propto T \sqrt{\chi_Q(T)} \propto T/\sqrt{(T + \theta)}$ [78]. Here, $\chi_Q(T)$, the staggered susceptibility with the AFM propagation vector $\mathbf{Q}$, follows the Curie-Weiss law and θ measures the deviation from the QCP. The specific heat divided by temperature C/T of CeIrSi_3 is estimated to be enhanced almost linearly with decreasing temperature from consideration of the entropy balance [75]. The enhancement of C/T is consistent with the spin fluctuation theory, even though from theory we cannot determine the proper dimensionality. The contradiction in the temperature responses of ρ and $1/T_1$ remains unsettled.

In general, it is unclear whether or not a QCP truly exists in CeTX_3 , since the AFM transitions above P_1^* are prevented or masked by superconductivity. For example, the specific heat jump associated with the AFM transition seems to disappear suddenly just above P_1^* in CeIrSi_3 as mentioned above. This suggests that the QCP exists neither at P_2^* nor at higher pressures. However, this does not necessarily indicate that magnetic fluctuations vanish at these pressures. The fact that the maximum T_c and the sharpest resistivity drop take place at P_3^* implies that magnetic fluctuations survive even above P_1^* and rather develop toward P_3^* . Assuming that magnetic fluctuations stabilize the superconducting phase and become stronger at the QCP, P_3^* should be regarded as a *virtual* QCP.

In the vicinity of the QCP some pressure-induced HF superconductors display a strong enhancement of the effective mass of the conduction electrons; namely, via the γ_n value and the coefficient A of the T^2 term of the electrical resistivity [79–81]. In the cases of CeRhSi_3 and CeIrSi_3 , however, such an enhancement is less obvious. The coefficient A in CeRhSi_3 is almost constant up to P_1^* [72] and the γ_n value in CeIrSi_3 is suggested to be unchanged up to P_3^* [75]. On the other hand, the coefficient A in CeCoGe_3 is strongly enhanced from $0.011 \mu\Omega \cdot \text{cm/K}^2$ at ambient pressure to $0.357 \mu\Omega \cdot \text{cm/K}^2$ at 5.4 GPa. In CeIrGe_3 $\rho(T)$ exhibits a complicated pressure dependence (as shown in Fig. 2.18) that makes unclear how A varies with pressure. The drastic changes at certain pressures between 8.6 GPa and 17 GPa may be associated with the step-like decrease of $T_N(P)$ around 13 GPa as seen in Fig. 2.15(d). The residual resistivity becomes maximum at a pressure near 17 GPa. Since such an enhancement of residual resistivity can be a signature of critical valence fluctuations [82], a valence transition or crossover may take place around 13 GPa in CeIrGe_3 .

2.2.3 Superconducting State

2.2.3.1 Anisotropic Upper Critical Field: Two Limiting Fields

Thus far, the most interesting phenomenon in the CeTX_3 superconductors is the extremely high anisotropy in the upper critical magnetic field H_{c2} , with stunningly

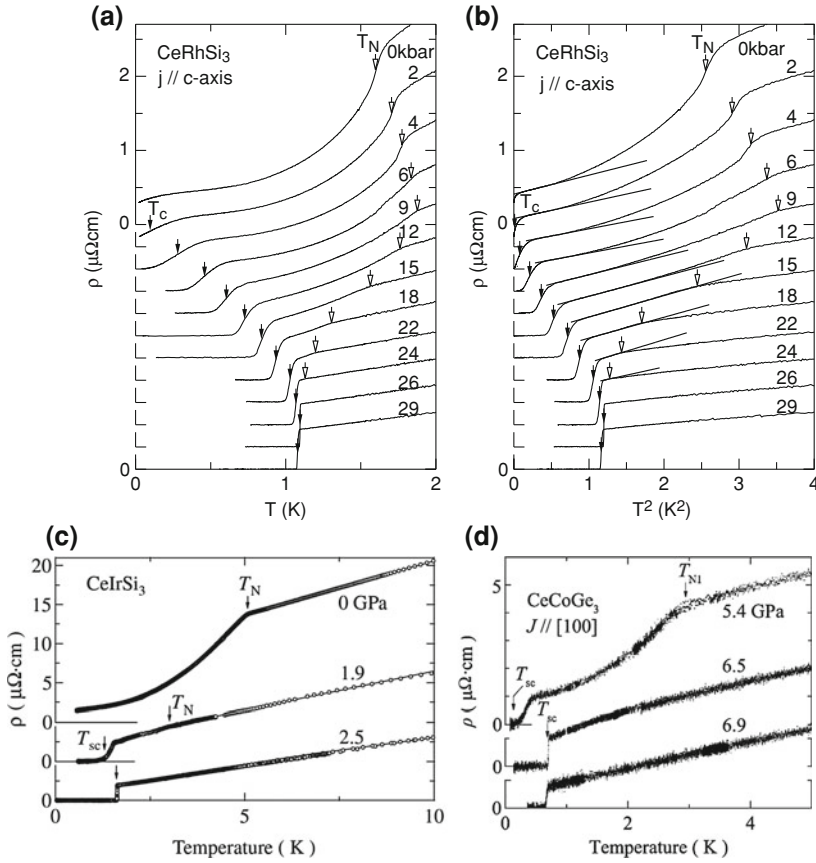


Fig.2.17 Temperature dependence of the resistivity of **a** CeRhSi₃ [72], **c** CeIrSi₃ [40] and **d** CeCoGe₃ [43]. **b** Resistivity against T^2 in CeRhSi₃

Table 2.4 Theoretically predicted quantum critical behavior for 3D and 2D antiferromagnetic fluctuations and corresponding temperature dependence on $CeTX_3$ compounds

	ρ	$1/T_1$	C/T
3D AFM	$T^{3/2}$	$T/\sqrt{T+\theta}$	Const. $-T^{1/2}$
2D AFM	T	$T/(T+\theta)$	$-\ln T$
CeRhSi ₃	T [39]		
CeIrSi ₃	T [40]	$T/\sqrt{T+\theta}$ [78]	$T?$ [75]
CeCoGe ₃	T [43]		
CeIrGe ₃	T [42]		

high values for fields along the c axis. In CeRhSi₃ and CeIrSi₃, for example, H_{c2} exceeds 30 T along the c axis, whereas falls below 10 T along the plane. Considering that in these materials T_c is of the order of 1 K, this leads to very high H_{c2}/T_c ratios not known previously for any *centrosymmetric* superconductors (except for field-

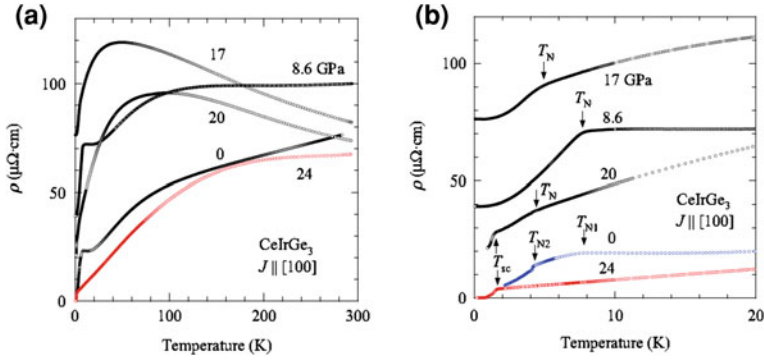


Fig. 2.18 Temperature dependence of the resistivity of CeIrGe_3 at several pressures up to 24 GPa below room temperature (*left*) and below 20 K (*right*) [42]

induced superconductors like URhGe [83] and organic superconductors [84]). The strong field anisotropy and the extremely high H_{c2} s are thought to be understood in terms of the anisotropy of the paramagnetic pair-breaking effect characteristic of non-centrosymmetric (Rashba-type) superconductors.

There are two pair-breaking mechanisms for Cooper pairs under magnetic fields: the paramagnetic and the orbital. The former mechanism is attributed to the spin polarization due to the Zeeman effect, which competes with the antiparallel-spin formation of the Cooper pair in spin-singlet superconductors. The influence of the paramagnetic effect depends on the symmetry of the Cooper pairs, as discussed later. On the other hand, the orbital effect is ascribed to the orbital motion in a magnetic field. The influence of this effect is thought to be independent of the pairing symmetry. The magnitude of $H_{c2}(0)$ is consequently restricted by both the paramagnetic (Pauli-Clogston-Chandrasekhar) limiting field H_P and the orbital limiting field H_{orb} [85]. Hereafter, we call H_P the Pauli limiting field for simplicity.

Pauli-Clogston-Chandrasekhar Limit

The paramagnetic effect in spin-singlet and spin-triplet pairing symmetries is illustrated schematically in Fig. 2.19. In centrosymmetric superconductors, the up-spin and down-spin bands of the conduction electrons are degenerate in zero field. The corresponding Fermi surfaces are also degenerate. The presence of a magnetic field inflates and deflates the down-spin and up-spin Fermi surfaces, respectively, due to the Zeeman effect. When the Cooper pair consists of antiparallel spins, e.g. the conventional singlet pair, the paramagnetic pair breaking occurs on the whole Fermi surface. When the Cooper pair comprises parallel spins, namely the triplet pair, the paramagnetic pair breaking does not occur, therefore the Pauli limit is absent. Since the spin direction of the Cooper pair is always aligned to the magnetic field, both cases are independent of the field direction unless the coupling between the orbital and spin parts of the pairing function is present.

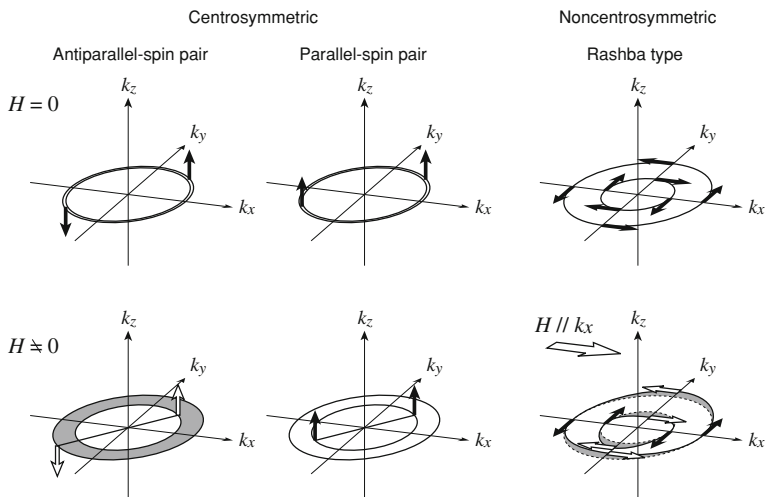
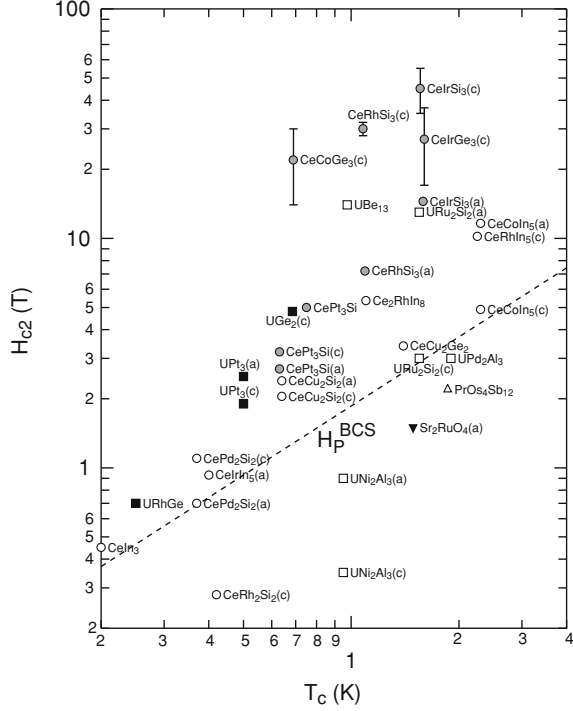


Fig. 2.19 Two-dimensional Fermi surfaces for centrosymmetric and non-centrosymmetric superconductors at zero and finite fields. The centrosymmetric superconductors can be classified into parallel- and antiparallel-spin pairs. In the non-centrosymmetric superconductor, only the Rashba type is displayed. The hollow arrows indicate a pairing no longer allowed. In the antiparallel-spin pair including the conventional singlet pair, the pair breaking occurs on the whole Fermi surface under magnetic fields. On the other hand, in the parallel-spin pair, all the pairing persist. In the Rashba-type superconductor, pair breaking occurs only for the pairs parallel to the applied magnetic field. When the magnetic field is applied along k_z , pair breaking does not occur

In non-centrosymmetric superconductors, the up-spin and down-spin Fermi surfaces are not degenerate even in zero field. In the Rashba-type (tetragonal) superconductors, the spins are perpendicularly aligned with the momenta in the k_z plane (Fig. 2.19) by the spin-orbit coupling, yielding a momentum-dependent effective magnetic field. The presence of an applied magnetic field along the k_x direction inflates and deflates the Fermi surfaces only along the k_y direction, since the spin component perpendicular to the k_x direction is not affected by this field. The paramagnetic pair-breaking effect is thus partial for field along the k_z plane. On the other hand, when the magnetic field is applied along the k_z axis, the paramagnetic pair-breaking effect is absent because all the spins aligned with the k_z plane are perpendicular to the field direction. The strongly anisotropic $H_{c2}(T)$, with high $H_{c2}(0)$ for $H \parallel c$, realized in CeTX_3 compounds is attributed to the anisotropic spin susceptibility expected to appear in non-centrosymmetric superconductors as discussed above in Spin State, Sect. 2.1.3.1.

Figure 2.20 shows $H_{c2}(0)$ versus T_c for CeTX_3 compounds and some well-known HF superconductors. The dashed line indicates H_p^{BCS} (see Eq. 2.4). The $H_{c2}(0)$ of the U-based superconductors UGe_2 , URhGe and UPt_3 exceeds H_p^{BCS} . These superconductors are thought to form a parallel-spin pairing free from the paramagnetic pair-breaking effect. In some spin-singlet superconductors $H_{c2}(0)$ is located above the H_p^{BCS} line, which seems a contradiction. Two possibilities have been proposed:

Fig. 2.20 $H_{c2}(0)$ versus T_c for heavy-fermion and some other well-known superconductors. The broken line represents $H_P^{\text{BCS}} = 1.86T_c$. The circles, squares and upper and lower triangles indicate cerium, uranium, praseodymium and other compounds, respectively. The solid and hollow symbols tag possible triplet and singlet (or unclear) superconductors, respectively. The gray marks label non-centrosymmetric superconductors. The letter *a* or *c* in parentheses denotes the applied field direction and no letter indicates that the result was obtained for a polycrystal

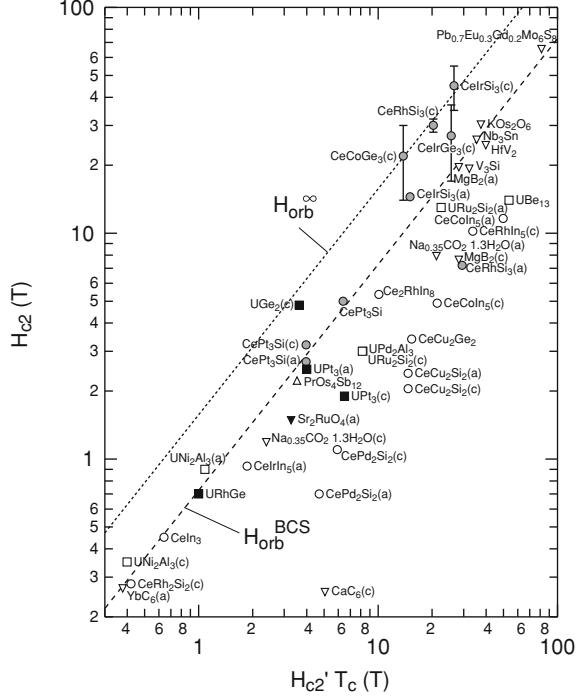


one is a reduction of the g -factor and the other is an enhancement of Δ_0 due to a strong-coupling effect. $H_{c2}(0)$ s of CeTX_3 compounds do not only exceed the H_P^{BCS} line, but also surpass those of all other materials. To understand the high H_{c2} of the CeTX_3 compounds, it is necessary to consider the orbital pair-breaking effect that we discuss next.

Orbital Limit

As we discussed above, the orbital limiting field can be estimated from Eq. (2.6). The value $h(0)$ depends on both the ratio $\xi(0)/l$ and the strong-coupling parameter λ (l : mean free path). In the weak-coupling limit ($\lambda = 0$), namely the BCS model, $h(0) = 0.727$ for $(\xi(0)/l) \rightarrow 0$ (clean limit) and $h(0) = 0.693$ for $(\xi(0)/l) \rightarrow \infty$ (dirty limit). Most HF superconductors satisfy the clean limit. In the strong-coupling limit ($\lambda \rightarrow \infty$), $h(0)$ approaches 1.57 for clean superconductors [86] and increases with λ for dirty superconductors. It is noted that the λ dependence of h is usually derived on the basis of the conventional electron-phonon model, but that in HF systems a corresponding electron-magnon approach should be employed since it is generally believed that the attractive interaction leading to Cooper pairs arises from coupling to spin excitations.

Fig. 2.21 $H_{c2}(0)$ versus $H'_{c2}T_c$ for heavy-fermion and other well-known superconductors. The broken and dotted lines represent $H_{orb}^{BCS} = 0.727H'_{c2}T_c$ and $H_{orb}^{\infty} = 1.57H'_{c2}T_c$, respectively. The circles, squares and upper and lower triangles indicate cerium, uranium, praseodymium and other compounds, respectively. The solid and hollow symbols tag possible triplet and singlet (or unclear) superconductors, respectively. The gray marks label non-centrosymmetric superconductors. The letter *a* or *c* in parentheses denotes the applied field direction and no letter indicates that the result was obtained for a polycrystal



The $H_{c2}(0)$ s in Fig. 2.20 are plotted against $H'_{c2}T_c$ in Fig. 2.21. The dashed and dotted lines indicate the orbital limiting fields at $\lambda = 0$ (weak-coupling limit) and $\lambda \rightarrow \infty$ (strong-coupling limit) for clean superconductors; that is, $H_{orb}^{BCS} = 0.727H'_{c2}T_c$ and $H_{orb}^{\infty} = 1.57H'_{c2}T_c$, respectively. Some parallel-spin, namely triplet, superconductors like UPt_3 and $URhGe$ are located just below H_{orb}^{BCS} . The compounds located far below the H_P^{BCS} line in Fig. 2.20, e.g. UNi_2Al_3 and $CeRh_2Si_2$, are seen in the vicinity of the H_{orb}^{BCS} line in Fig. 2.21. The H_{c2} s of these compounds may be mainly restricted by the orbital limit rather than by the Pauli limit. $CePt_3Si$ is also located near the H_{orb}^{BCS} line. Considering that H_{c2} is almost isotropic in $CePt_3Si$ [10], it may be also mainly constrained by the orbital limit rather than by the paramagnetic one.

The H_{c2} s of the $CeTX_3$ compounds for fields along the *c* axis well exceed the H_{orb}^{BCS} line. They seem to be close to the strong-coupling limit H_{orb}^{∞} . Although the high $H_{c2}/(H'_{c2}T_c)$ is a common feature in the $CeTX_3$ HF superconductors, it is not obvious that such a result can be associated with their non-centrosymmetric crystal structures. Note that UGe_2 also seems to be above the H_{orb}^{BCS} line, but this is due to the jump of the H_{c2} curve attributed to the metamagnetic transition [87, 88]. Therefore, this plot does not necessarily indicate that the intrinsic $H_{c2}(0)$ of UGe_2 exceeds the H_{orb}^{BCS} line.

2.2.3.2 Upper Critical Field for c Axis

In addition to the high values of $H_{c2}(0)$, the upward shape of the temperature dependence of $H_{c2}(T)$ for certain pressures seems to be also a characteristic of the CeTX_3 superconductors [43, 54, 72]. They mostly keep a positive curvature ($d^2 H_{c2}/dT^2 > 0$) down to relatively low temperatures; for example, in CeIrSi_3 down to $T \approx 0.25T_c$ at 2.6 GPa (Fig. 2.23). Interestingly, the curve shapes of CeRhSi_3 and CeIrSi_3 vary with pressure in a complex manner. As for CeIrSi_3 , a positive curvature is seen up to 2 GPa that gradually changes to a negative curvature at 2.4 GPa to a quasi-linear shape at 2.3 GPa. At 2.6 GPa and 2.65 GPa, a positive curvature is recovered that turns to a negative one at higher pressures. Similar behavior is seen in CeRhSi_3 . Below and above 2.6 GPa, the curvatures of $H_{c2}(T)$ are positive. At 2.6 GPa, a quasi-linear change of $H_{c2}(T)$ is observed. Because of the lack of sufficient pressure data, it is not clear whether such a phenomenon is realized in CeCoGe_3 and CeIrGe_3 .

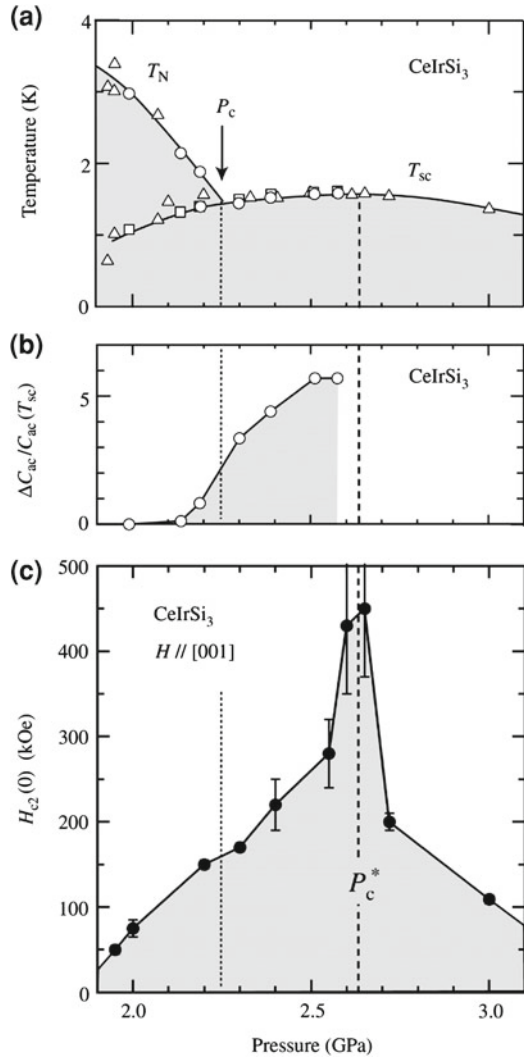
In order to understand the superconducting phase diagram of the CeTX_3 compounds the pressure dependence of $H_{c2}(0)$ will be very helpful. Fig. 2.22(c) shows $H_{c2}(0)$ versus pressure in CeIrSi_3 [54]. $H_{c2}(0)$ increases with pressure and tends to diverge close to 2.65 GPa ($\approx P_3^*$). Above 2.65 GPa it falls steeply to half or less of the value of the maximum $H_{c2}(0)$. It is pointed out that such an acute enhancement of $H_{c2}(0)$ can be interpreted as an electronic instability arising at P_3^* [54]. This instability can cause a mass enhancement of the conduction electrons, giving rise to a reduction in the superconducting coherence length $\xi(0)$. At a first glance, a mass enhancement at P_3^* is consistent with the pressure dependence of the initial slope of the superconducting H - T phase diagram, $H'_{c2} = -dH_{c2}/dT|_{T=T_c}$. From Eqs. (2.5) and (2.6), we can derive

$$H'_{c2}T_c \sim H_{orb} \sim \xi^{-2}(0) \sim (\Delta_0 m^*)^2. \quad (2.10)$$

Here, we use $v_F = \hbar k_F/m^*$. As shown in Figs. 2.23–2.25, H'_{c2} s of CeIrSi_3 and CeRhSi_3 increase with increasing pressure and become maximum at about P_3^* . However, strong pressure dependence of the cyclotron effective mass is not observed in de Haas-van Alphen experiments in CeRhSi_3 [89]. This is consistent with the result of the less obvious pressure dependence of the resistivity coefficient A . A small enhancement of the effective mass is also suggested by heat-capacity measurements in CeIrSi_3 [75].

In order to explain the positive curvature of $H_{c2}(T)$ and the strong pressure dependence of $H_{c2}(0)$, Tada *et al.* considered the temperature and pressure dependencies of the correlation length of the spin fluctuations, ξ_{sf} , [90]. Since ξ_{sf} is expressed as $\xi_{sf}(T) = \frac{\xi_{sf}}{\sqrt{T+\theta}}$, in which $\theta \rightarrow 0$ toward the QCP and the effective pairing interaction is quadratically proportional to ξ_{sf} , the superconducting coherence length $\xi(0)$ is strongly reduced and H_{orb} enhanced at low temperatures. This model can explain the enhancement of the initial slope and is compatible with the weaker enhancement of the effective mass.

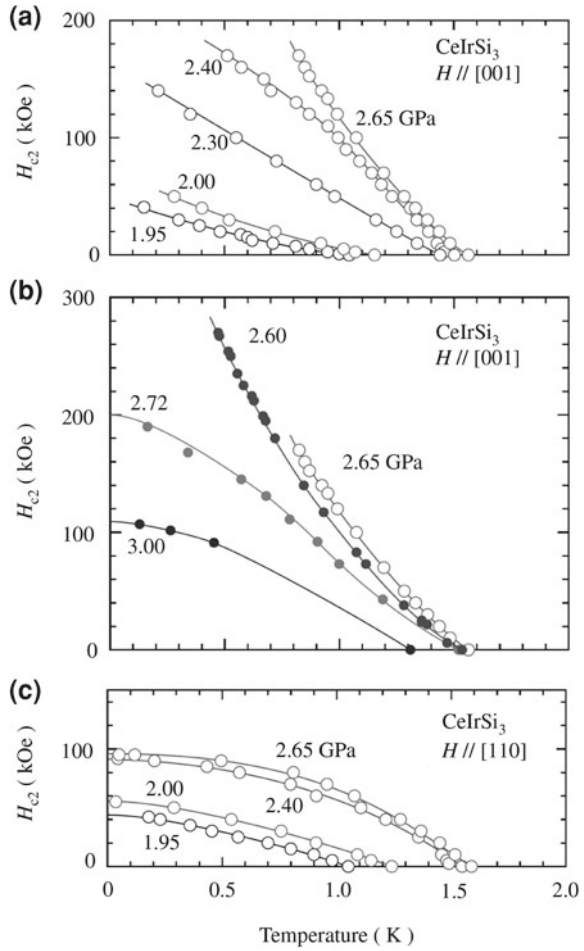
Fig. 2.22 Pressure dependence of **a** T_N and T_c , **b** specific heat jump $\Delta C/C(T_c)$ and **c** $H_{c2}(0)$ for $H \parallel c$ in CeIrSi_3 [54]



2.2.3.3 Superconducting Phase Diagram for Field in the Basal Plane

In contrast to the high $H_{c2}(T)$ for the c axis, $H_{c2}(T)$ in the basal plane is not too high. However, it still significantly exceeds H_P^{BCS} and is situated in the upper part of Fig. 2.20. As discussed in the section above, the high $H_{c2}(0)$ is attributed to the reduced paramagnetic pair-breaking effect which originates mainly from a non-vanishing spin susceptibility. In addition to this, Agterberg et al. pointed out that another characteristic mechanism, the helical vortex state, can also evade the

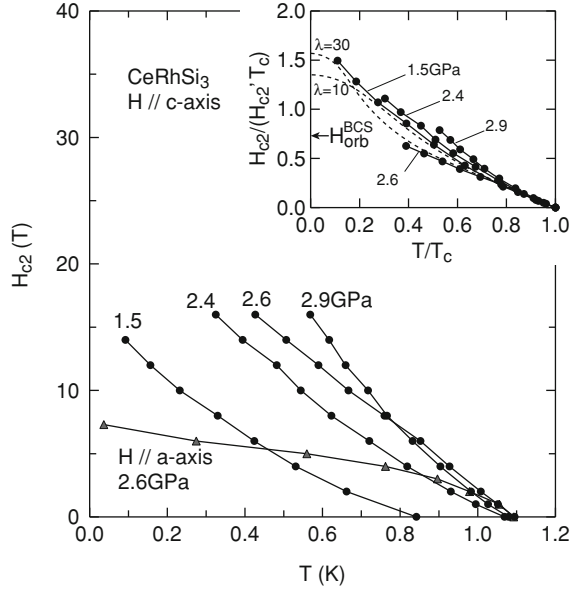
Fig. 2.23 $H_{c2}-T$ phase diagrams of CeIrSi_3 for $H \parallel c$ at $P \leq P_3^* \approx 2.65$ GPa **a**, at $P \geq P_3^*$ **b**, and for $H \perp c$ at $P \leq P_3^*$ [54]



paramagnetic pair-breaking effect when the magnetic field is applied in the basal plane [91, 92].

As discussed above, the paramagnetic pair-breaking effect operates partially, with the center-of-mass momentum of the Cooper pair remaining zero at any k as shown in Fig. 2.26(b). On the other hand, in the helical vortex state, the Fermi surfaces shift toward opposite directions perpendicular to the field direction. An application of a magnetic field does not break the Cooper pairs for all k . In this case, the center-of-mass momenta of the Cooper pairs belonging to each Fermi surface acquire a finite value $\pm q$ (Fig. 2.26(c)). The sign of q depends on the Fermi surface. In the Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state, a Cooper pair with a finite center-of-mass momentum is also realized. However, the pairing takes place in a limited region on the Fermi surface. In other regions, indicated by the shades in Fig. 2.26(c),

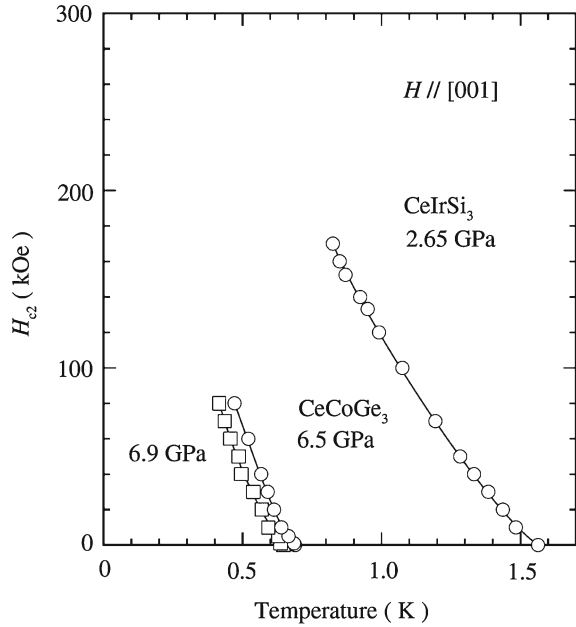
Fig. 2.24 $H_{c2}-T$ phase diagrams of CeRhSi_3 for $H \parallel c$ at several pressures and for $H \parallel a$ at $P_3^* \approx 2.6$ GPa. Inset: $H_{c2}(T)$ curves for $H \parallel c$ normalized by the initial slope. The arrow indicates the orbital limit H_{orb}^{BCS} . The dashed curves are theoretical predictions based on the strong-coupling model using the coupling strength parameter $\lambda = 10$ and 30 [93]



pairing is not allowed. Since both states, helical vortex and FFLO phase, evade the paramagnetic pair breaking, relatively high $H_{c2}(0)$ can be realized.

Although thus far no direct evidence for a helical vortex state has been detected in either CeTX_3 compounds or CePt_3Si , some unusual superconducting properties for the field in the basal plane are reported in CeRhSi_3 [39]. First, there is a concave shape of the $H_{c2}(T)$ curve as shown in Fig. 2.27(a). The rapid change of H_{c2} at low temperatures looks similar to the one observed in a theoretically predicted helical-vortex phase diagram [92]. However, this feature observed at $P < P_1^*$ becomes less obvious at $P_3^* \approx 2.6$ GPa [72]. An influence of the antiferromagnetic order to explain this unusual curve shape cannot be excluded. Second, the ac susceptibility in the superconducting state shows an unusual shape especially below P_1^* . The temperature at which a large drop occurs in the real part of the susceptibility χ' is far below the onset temperature of superconductivity. This might indicate that superconductivity develops gradually in the antiferromagnetic state. On the other hand, the imaginary part χ'' , namely the energy dissipation associated with the dynamics of the superconducting flux, is large even above the temperature at which the large drop occurs in χ' . This contradicts the view of a gradual development of superconductivity. Although the influence of antiferromagnetism is unclear at present, this rare behavior of the flux may be a key feature to verify the helical vortex state.

Fig. 2.25 H_{c2} – T phase diagram of CeCoGe_3 for $H \parallel c$ at 6.5 GPa and 6.9 GPa. The phase diagram of CeIrSi_3 is also displayed [43]



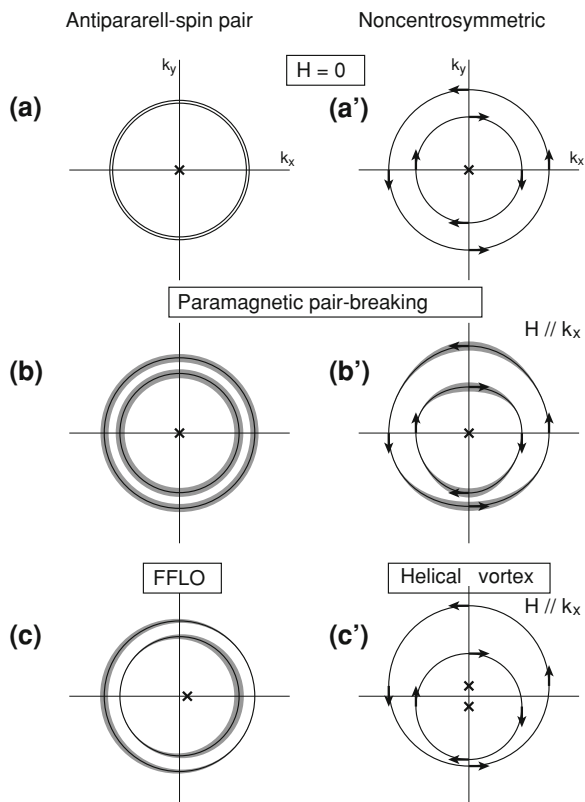
2.2.3.4 Energy Gap Structure and Pairing Symmetry

In CeIrSi_3 the nuclear spin-lattice relaxation rate $1/T_1$ shows a T^3 -like dependence below T_c without a coherence peak, as shown in Fig. 2.28 [78]. The data are well fitted by the line-node gap model $\Delta = \Delta_0 \cos 2\theta$. The fit yields $2\Delta_0/k_B T_c \approx 6$, which is much larger than the BCS weak-coupling value 3.53 and thus suggests strong-coupling superconductivity. Using an extended $s + p$ pairing state within a recent theoretical model a behavior indicative of a line-node gap can be predicted [94]. This $1/T_1$ -NMR measurement is the only one carried out to determine the energy gap structure in CeTX_3 compounds. To identify the pairing symmetry in these materials, other measurements, like the Knight shift, are highly desirable.

2.2.4 Outlook

The high anisotropy and strong enhancement of H_{c2} seem to be unique to CeTX_3 superconductors. Interestingly, other HF and non-HF superconductors without inversion symmetry do not show these properties. The high orbital limiting field inherent to the non-centrosymmetric HF CeTX_3 superconductors discloses the absence of the paramagnetic pair-breaking effect. Conversely, absence of the effect unveils the unconventional nature of the upper critical fields probably associated with the quantum criticality of magnetism. CeTX_3 compounds have the potential to provide a

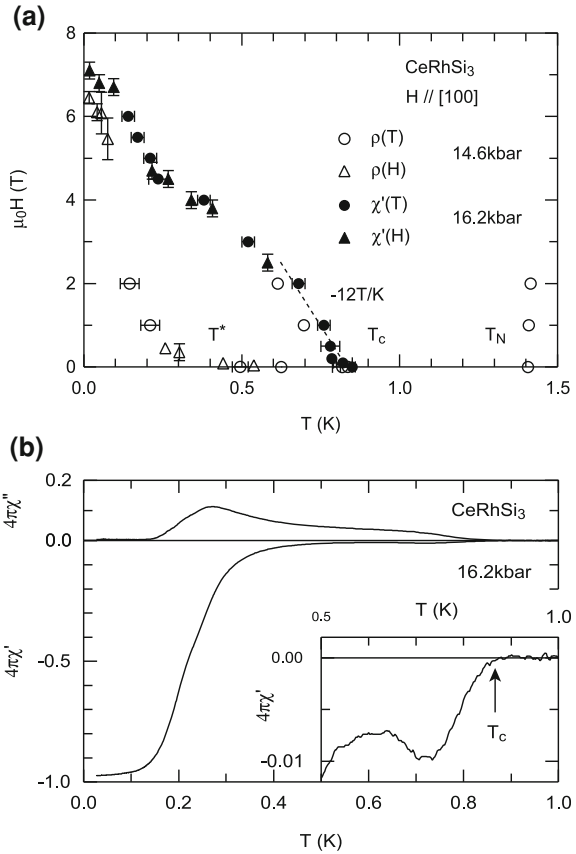
Fig. 2.26 Schematic illustrations of Fermi surface sliced perpendicular to the k_z axis. Pair breaking takes place at the shaded region. The x marks indicate the positions of the center-of-mass momenta of the Cooper pairs



vital clue to the underlying mechanism of superconductivity mediated by magnetic fluctuations.

To consider the relation between magnetism and superconductivity in CeTX_3 , we need to keep in mind that the magnetic ground states of CeCoGe_3 and CeIrGe_3 are different from that of CeRhSi_3 and probably of CeIrSi_3 . CeCoGe_3 seems to display localized f -electron magnetism, while CeRhSi_3 probably exhibits itinerant-electron magnetism. It is very interesting that in these compounds the H_{c2} behaviors are similar in spite of their different magnetism. The mass enhancement at P_3^* is suggested only in CeCoGe_3 . The comparison of the superconducting properties of CeCoGe_3 and CeIrGe_3 with those of CeRhSi_3 and CeIrSi_3 will be important to elucidate the nature of HF superconductors. Identification of the gap structure in each compound is also a challenging issue, which could provide possible evidence for the parity mixing of the superconducting wavefunction. Moreover, some theoretically predicted phenomena, like a helical vortex phase and a novel magnetoelectric effect, remain to be verified in the future.

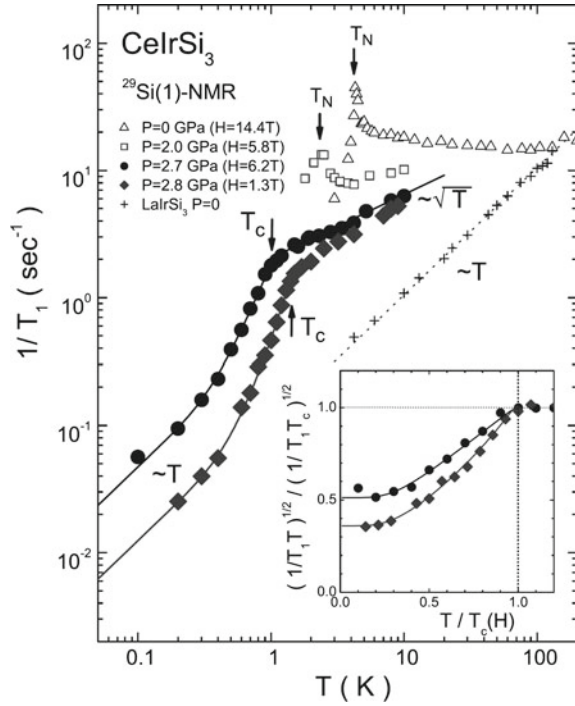
Fig. 2.27 (a) Magnetic field-temperature phase diagram of CeRhSi₃ for $H \parallel a$ below P_1^* . T^* denotes the temperature at which an anomaly is observed in the resistivity and magnetic ac susceptibility measurements. (b) ac susceptibility as a function of temperature at zero field below P_1^* . An enlarged view of the vicinity of T_c is shown in the inset [39]



2.3 UIr

Whereas CeRhSi₃, CeIrSi₃, CeCoGe₃, CeIrGe₃ and CePt₃Si are 4*f*-electron anti-ferromagnets, UIr is a 5*f*-itinerant-electron ferromagnet with a Curie temperature $T_{c1} = 46$ K at ambient pressure [95]. The superconducting state in UIr appears to develop within a higher pressure ferromagnetic phase at a critical temperature $T_c = 0.14$ K in a narrow pressure region around 2.6 GPa [96]. UIr is a moderate heavy-fermion compound with a cyclotron mass $m^* \sim 10 - 30 m_0$ [97]. The coexistence of superconductivity and ferromagnetism imposes several theoretical challenges, such as the mechanism and the state of pairing. The pairing state in superconducting ferromagnets needs to be spin triplet, otherwise the internal exchange field would break the Cooper pairs. On the other hand, a ferromagnetic state has a broken time reversal symmetry. The superconducting BCS ground state is formed by Cooper pairs with zero total angular momentum. The electronic states are four-fold degenerate: $|\mathbf{k} \uparrow\rangle$, $|\mathbf{k} \downarrow\rangle$, $|\mathbf{k} \uparrow\rangle$, and $|\mathbf{k} \downarrow\rangle$ have the same energy $\varepsilon(\mathbf{k})$. The states

Fig. 2.28 Temperature dependence of $1/T_1$ measured by Si NMR for CeIrSi₃ at several pressures. The solid curves below T_c for CeIrSi₃ indicate the calculated values obtained by the line-node gap model with $2\Delta_0/(k_B T_c) \approx 6$ and the residual density-of-states fraction $N_{\text{res}}/N_0 \approx 0.37$ (0.52) in $H = 1.3$ T (6.2 T). The inset shows the plot of $\sqrt{1/(T_1 T)}$ normalized by that at T_c which allows us to evaluate N_{res}/N_0 in the low-temperature limit [78]



with opposite momenta and opposite spins are transformed to one another under time reversal operation $\hat{K}|\mathbf{k} \uparrow\rangle = |-\mathbf{k} \downarrow\rangle$, and the states with opposite momenta are transformed to one another under inversion operation $\hat{I}|\mathbf{k} \downarrow\rangle = |-\mathbf{k} \downarrow\rangle$. The four degenerate states are a consequence of spatial and time inversion symmetries. Parity symmetry is irrelevant for spin-singlet pairing, but is essential for spin-triplet pairing. Time reversal symmetry is required for spin-singlet configuration, but is unimportant for spin-triplet state [98, 99]. In UIr the lack of spatial and time inversion symmetries lifts the degeneracies and, therefore, superconductivity is not expected to occur. Thus, UIr differs from the other two known ferromagnetic superconductors UGe₂ [100] and URhGe [101], in which the spatial inversion symmetry allows degeneracy in the spin-triplet states. Theoretically and experimentally UIr is a very special and challenging superconductor.

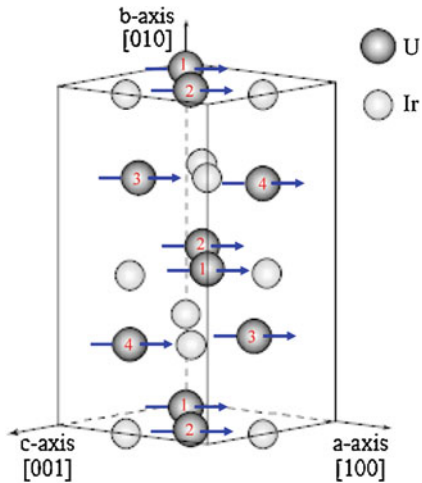
2.3.1 Crystal Structure and Characteristic Parameters

UIr crystallizes in a monoclinic PbBi-type structure (space group $P2_1$) without inversion symmetry [95]. The lattice parameters are given in Table 2.5. The unit cell has eight formula units with four inequivalent U and Ir sites. The absence of inversion symmetry comes from the missing mirror plane $(0, \frac{1}{2}, 0)$ perpendicular to the b axis (see Fig. 2.29). Magnetism is of the Ising type with the ordered magnetic moment oriented along the spin easy axis $[1\ 0\ \bar{1}]$ (Fig. 2.29).

Table 2.5 Normal and superconducting parameters of UIr

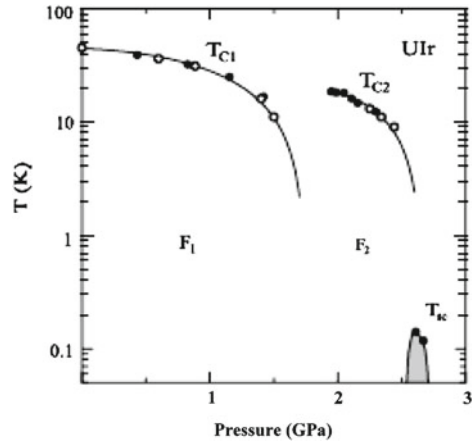
Crystal structure	Monoclinic
Space group	$P2_1$
Lattice parameters	$a = 5.62 \text{ \AA}$ $b = 10.59 \text{ \AA}$ $c = 5.60 \text{ \AA}$ $\beta = 98.9^\circ$
Sommerfeld value of specific heat	$\gamma_n = 40\text{--}49 \text{ mJ/molK}^2$
Effective electron mass (Fermi sheet α)	$m^* \sim 10\text{--}30 m_0$
Mean free path (Fermi sheet α)	$l = 1270 \text{ \AA}$
Ferromagnetic transition temperature (ambient pressure)	$T_{c1} = 46 \text{ K}$
Magnetic propagation vector	$\mathbf{q} = (1, 0, -1)$
Magnetic moment $\mathbf{m}_Q$ along	$[10\bar{1}]$
Saturated moment per U atom	$\mu_s = 0.5 \mu_B$
Superconducting transition temperature	$T_c = 0.14 \text{ K}$
Upper critical field	$H_{c2}(0) = 26 \text{ mT}$
Thermodynamic critical field	$H_c(0) = 8 \text{ mT}$
Ginzburg-Landau coherence length	$\xi(0) = 1100 \text{ \AA}$
Ginzburg-Landau parameter	$\kappa \sim 2$

Fig. 2.29 Crystal and magnetic structures of UIr



Single crystals of UIr have been grown by the Czochralski method in a tetra-arc furnace [102–106]. After annealing, using the solid-state electrotransport technique under high vacuum of the order of 10^{-10} Torr, crystals become of very high quality with residual resistivity $\rho_0 \sim 0.5 \mu\Omega$ and residual resistivity ratio (RRR) $\rho_{300\text{K}}/\rho_0 \approx 200$ at ambient pressure. Interestingly, single crystals of UIr seem to be of the highest quality amongst those of non-centrosymmetric heavy-fermion superconductors.

Fig. 2.30 Temperature–pressure phase diagram of UIr [96]. F_1 and F_2 are ferromagnetic phases



2.3.2 Normal State

2.3.2.1 Phase Diagram and Magnetic Properties

Figure 2.30 shows the temperature–pressure phase diagram as drawn by magnetization and resistivity measurements [96]. The diagram consists of a low-pressure ferromagnetic phase FM1 (F_1 in the figure), a high-pressure ferromagnetic phase FM2 (F_2 in the figure) and a superconducting phase. A third magnetic phase was reported [107], but not further evidence for it has been found. Application of pressure decreases the Curie temperature T_{c1} of the ferromagnetic phase FM1 eventually to zero at the critical pressure $P_{c1} \sim 1.7$ GPa. The FM2-paramagnetic curve appears just below 30 K and about 1.4 GPa, and goes away at a critical pressure $P_{c3} \sim 2.7\text{--}2.8$ GPa. Superconductivity is found in the narrow pressure range 2.55–2.75 GPa below $T_c = 0.14$ K.

The magnetic properties of this compound are governed by a saturation moment along the easy axis $[10\bar{1}]$, as indicated by the magnetization curve of a single crystal at 2 K shown in Fig. 2.31(a) [108]. The ordered magnetic moment goes from $0.5\mu_B/\text{U}$ at ambient pressure in the ferromagnetic phase FM1 to $0.07\mu_B/\text{U}$ at 2.4 GPa in the ferromagnetic phase FM2 [96]. The anisotropy of the magnetization remains at high temperatures, as can be seen in Fig. 2.31(b). The susceptibility data follow a Curie–Weiss law in the high-temperature paramagnetic region, with an effective magnetic moment around $3.57\mu_B/\text{U}$ that is pretty close to the $5f^2$ free-ion value $3.58\mu_B/\text{U}$. The small value of the ordered moment $0.5\mu_B/\text{U}$ has been taken as evidence for the itinerant character of the $5f$ electrons in the ferromagnetic phase, though such a low value could also be due to crystal-field effects [106, 109].

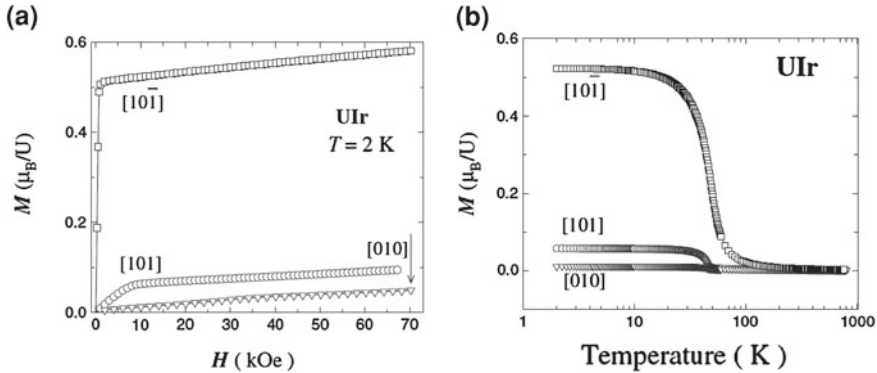


Fig. 2.31 Magnetization as a function of **a** field strength at 2 K and **b** temperature in 10 kOe in a single crystal of UIr [108]

2.3.2.2 Electronic States

Quantum-oscillation and resistivity measurements provide evidence that the low-temperature metallic state of UIr is a Fermi liquid at ambient pressure. The dHvA measurements suggest that the Fermi surface of UIr is two-dimensional and consists mainly of nearly cylindrical sheets [97]. It has an effective mass $m^* \sim 10-30 m_0$ and a mean free path $l \sim 1270$ Å. Such a value of the effective mass of the $5f$ electrons leads to the classification of UIr as a moderate heavy-fermion compound. Since the linear coefficient of the heat capacity $\gamma_n \propto m^*$, summing for all the branches yields the electronic specific-heat coefficient of 40–49 mJ/K² mol [97, 110]. There are no band-structure calculations for this compound.

Figure 2.32(a) shows the variation of the electrical resistivity of UIr as a function of pressure [105, 110]. At low pressure, in the ferromagnetic phase FM1, the resistivity follows $\rho = \rho_0 + AT^n$, with $n \sim 2$ suggesting Fermi-liquid behavior. However, as pressure increases the behavior becomes non-Fermi-liquid like and eventually superconductivity appears in this regime. Figures 2.32(c–e) show the variation in n , A and ρ_0 as pressure increases. The non-Fermi-liquid behavior above ~ 1 GPa may be related to critical fluctuations near the different magnetic transitions.

2.3.3 Superconducting State

Because of its extremely low critical temperature $T_c = 0.14$ K (in most figures in this section this critical temperature is called T_{sc}), there is little information on the superconducting state of UIr. Superconductivity seems to occur inside and near the quantum critical point of the FM2 phase, in the very narrow pressure range of 2.6–2.75 GPa [96]. In the ferromagnet UGe₂ with inversion symmetry the superconducting phase exists inside the ferromagnetic phase as well. Figure 2.33(a) shows the temperature dependence of the resistivity below 10 K and at 2.61 GPa, where

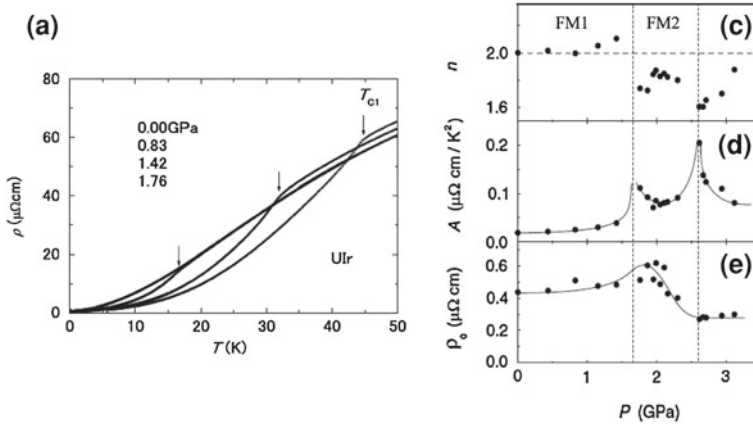


Fig. 2.32 **a** Resistivity as a function of temperature of UIr at different pressures, and **c–e** variations in the parameters n , A and ρ_0 of $\rho = \rho_0 + AT^n$ as pressure increases [108]

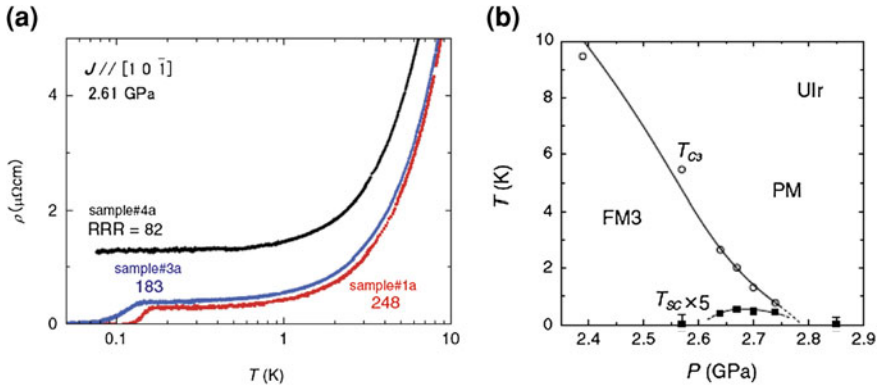
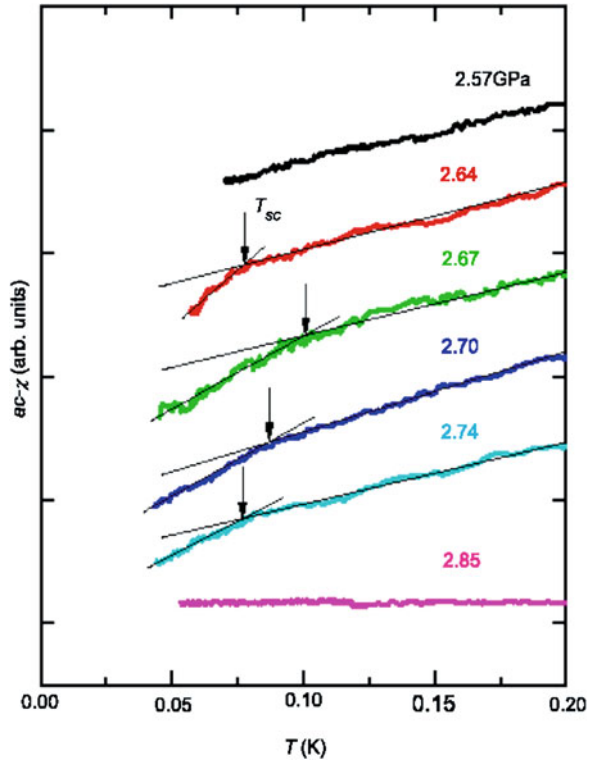


Fig. 2.33 **a** Resistivity below 10 K and at 2.61 GPa of UIr showing a non-Fermi-liquid behavior. The three samples possess different RRR-values: 82, 183 and 248. **b** Low-temperature region of the phase diagram of UIr [107]

it follows a non-Fermi-liquid form $T^{1.6}$ [105]. Figure 2.33(b) is a close-up of the low-temperature region of the phase diagram where superconductivity appears (in this figure, FM3 denotes the ferromagnetic phase FM2).

Up to now experimental indications of the existence of a superconducting phase in UIr come from measurements of resistivity in samples of different qualities. No definite diamagnetic signal has yet been observed in UIr (Fig. 2.34) [107]. The temperature dependence of the resistivity for three different samples is shown in Fig. 2.33(a). The data indicate that superconductivity becomes weaker as the residual resistivity ratio (RRR) of the samples drops. Such a strong suppression of T_c with increasing impurities/defects is typical of unconventional parity-conserving super-

Fig. 2.34 Temperature dependence of the ac susceptibility of UIr below 0.2 K at different pressure. Arrows indicate the onset temperatures [107]

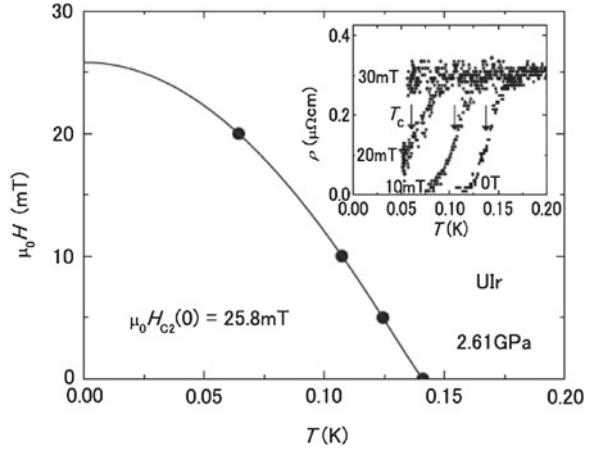


conductors [27]. Recent theoretical works [111, 112] considered impurity effects on the critical temperature of superconductors without inversion symmetry. It was found that impurity scattering leads to a functional form of T_c that, up to a prefactor, is the same as the one for unconventional superconductor with inversion symmetry: $\ln(T_c/T_{c0}) = \alpha \left[\psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} - \frac{\Gamma}{2\pi T_c}\right) \right]$. The suppression of T_c by impurities in non-centrosymmetric UIr agrees with this prediction [111, 112].

The upper critical field $H_{c2}(T)$ in the direction of the easy axis $[1\ 0\ \bar{1}]$ in the high-temperature region was determined by resistivity measurements in a sample with a very high RRR (Fig. 2.35) [105]. T_c was defined as the midpoint of the resistivity drop. By assuming the standard empirical expression $H_{c2}(T) = H_{c2}(0) [1 - (T/T_c)^2]$, the zero-temperature upper critical field $H_{c2}(0)$ was estimated as 26 mT corresponding to a coherence length of $\xi(0) = 1100\ \text{\AA}$. Since this value of $H_{c2}(0)$ is smaller than the paramagnetic limiting field $H_p = 280\ \text{mT}$ orbital depairing is the likely mechanism for the upper critical field in UIr.

It is believed that near a ferromagnetic quantum critical point spin fluctuations lead to Cooper pairing in a spin-triplet channel. There are some possible scenarios for the unexpected realization of superconductivity in this compound. It needs to be confirmed that FM2 is indeed a ferromagnetic phase, and not a canted antiferromag-

Fig. 2.35 Upper critical field $H_{c2}(T)$ along the easy axis $[10\bar{1}]$ in UIr. The critical temperatures were defined by the *midpoints* of the resistivity drops in the inset [105]



netic phase that could yield pairing in the spin-singlet channel. A canted antiferromagnetic phase may be caused by the spin-orbit coupling and the low symmetry of the crystalline structure without inversion symmetry, as discussed by Dzyaloshinsky and Moriya. In this sense, it is important to note that the saturated moment of $0.07 \mu_B/\text{U}$ in the FM2 phase is quite small. Another possibility is the FFLO state, in which at zero magnetic field electrons with momenta $\mathbf{k}$ and $-\mathbf{k} + \mathbf{q}$ can pair with nonzero angular momenta. A mean-field model has been recently proposed for superconductivity in non-centrosymmetric ferromagnets [113], in which the antisymmetric spin-orbit coupling (ASOC) turns out to enhance both superconductivity and ferromagnetism in all spin channels. Future experiments will be absolutely important for the understanding of this unique superconductor.

2.4 Comparison of the Superconducting States of CePt_3Si , CeRhSi_3 , CeIrSi_3 , CeCoGe_3 , CeIrGe_3 and UIr

The superconducting properties of the non-centrosymmetric HF compounds have not been easy to determine. On the one hand, the CeTX_3 and UIr compounds only superconduct under pressure, making technically difficult to study them. On the other hand, CePt_3Si does become superconducting at ambient pressure, but has drawbacks in the crystal quality available. In spite of this, it has been possible to establish some of the characteristics of these materials.

The CeTX_3 materials, with tetragonal space group $I4mm$, and CePt_3Si , with tetragonal space group $P4mm$, have the same generating point group C_{4v} which lacks a mirror plane and a two-fold axis normal to the c axis. Thus, a Rashba-type interaction appears in all these compounds due to the missing of inversion symmetry. In contrast, UIr has a monoclinic lattice.

The Sommerfeld coefficient is much larger in CePt₃Si than in CeTX₃ and UIr. Electron correlations should hence be stronger in CePt₃Si. On the other hand, all compounds turn from Fermi-liquid to non-Fermi-liquid states as pressure is increased and become superconducting in the Fermi-liquid state, with the exception of UIr.

The behavior of the upper critical field H_{c2} in the CeTX₃ systems is very different from that in CePt₃Si. In CeTX₃, $H_{c2\parallel c} > 22$ T and $(-dH_{c2\parallel c}(T)/dT)_{T_c} > 17$ T/K are larger than $H_{c2\parallel c} \approx 3$ T and $(-dH_{c2\parallel c}(T)/dT)_{T_c} \sim 6.3$ T/K in CePt₃Si. Moreover, in CeTX₃ $H_{c2}(T)$ has a positive curvature, unlike in CePt₃Si. In CeIrSi₃, for example, superconductivity is anisotropic ($H_{c2\parallel c}/H_{c2\parallel ab} > 3$), whereas in CePt₃Si it is almost isotropic. There are clear signatures for unconventional superconductivity in CeIrSi₃, CeRhSi₃ and CePt₃Si, including evidence for line nodes in some cases. The fact that these compounds also support strong magnetic features and order suggests strongly that unconventional pairing mechanisms could be at work here. In this context it is particularly intriguing to analyze the role the antisymmetric spin-orbit coupling.

Acknowledgements We are grateful to M. Sigrist, S. Fujimoto, Y. Tada, D. F. Agterberg, K. Samokhin, F. Honda, Y. Matsuda, T. Sugawara, T. Terashima, H. Aoki, F. Lévy and I. Sheikin for helpful discussions. The work of N.K. was supported by KAKENHI (grant numbers 18684018 and 20102002) and partially by MEXT of Japan through Tohoku University GCOE program “Weaving Science Web beyond Particle-matter Hierarchy”. I.B. acknowledges initial support by the Venezuelan FONACIT (grant number S1-2001000693).

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Non-Centrosymmetric Superconductors

Introduction and Overview

Bauer, E.; Sigrist, M. (Eds.)

2012, XI, 357 p. 151 illus., Softcover

ISBN: 978-3-642-24623-4