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Emergent phenomena in actinides / Phénomènes émergents dans les actinides

Superconductivity in transuranium elements and compounds

Supraconductivité dans les éléments et les composés transuraniens

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ABSTRACT

We present here an overview of the properties of transuranium superconductors, but also of the (non-superconducting) transuranium analogues of uranium superconductors. We briefly review superconductivity in actinide elements and uranium compounds and focus in particular on the $PuTX_5$ ($T = Co, Rh$; $X = Ga, In$) series, the largest superconducting system in actinides and $NpPd_5Al_2$, the so far unique neptunium superconductor. The effects of chemical substitution, ageing and pressure on the properties of transuranium superconductors are also discussed.

RÉSUMÉ

Nous présentons ici un aperçu des propriétés des supraconducteurs transuraniens, mais aussi des analogues transuraniens (non supraconducteurs) des supraconducteurs à base d'uranium. Nous examinons brièvement la supraconductivité dans les éléments d'actinides et les composés d'uranium, puis nous présentons en particulier la famille des composés $PuTX_5$ ($T = Co, Rh$, $X = Ga, In$), la série la plus étendue de supraconducteurs parmi les actinides ainsi que $NpPd_5Al_2$, le seul supraconducteur au neptunium connu à ce jour. Les effets de la substitution chimique, du vieillissement et de la pression sur les propriétés des supraconducteurs transuraniens sont également discutés.

1. Introduction

Uranium was discovered in 1789 by Klaproth [1], eight years after the first observation of the planet Uranus by Herschel [2]. Thorium was the next actinide to be discovered in 1828 by Berzelius [3]. After the discovery of neptunium [4] and plutonium [5] in 1940, other transuranium elements followed after World War II [6].

Superconductivity in actinides was first observed in thorium metal in 1929 [7], then in elemental uranium in 1942 [8], and in uranium compounds in 1958 [9]. A new class of uranium superconductors emerged in the 1980's with the discovery of uranium heavy fermion superconductors [10]. Further surprises came at the beginning of the century with the discovery of ferromagnetic superconductors in uranium systems [11] and the first observation of superconductivity in plutonium [12]

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and neptunium [13] compounds. The actinides (or actinoids) are located at the end of the periodic table ($N = 89$ (Ac) or 90 (Th) to 103 (Lr)). Transuranium elements (or transuranics) are the chemical elements with atomic number (Z) greater than 92 (uranium) and due to their short half-life on a geological timescale, they are essentially synthetic elements. Above $Z = 103$ (Lr), one talks about transactinides (or superactinides) elements. These latter elements have extremely short half-lives and no macroscopic quantity is available for the study of condensed-matter properties.

Actinides are characterized by their uncompleted 5f electronic shell, progressively filled when progressing along the series. The remarkable feature of 5f electrons is their duality: the light actinides (up to plutonium) are itinerant whereas heavier actinides are localized such as curium, berkelium and californium which even present a magnetic order (the ultimate elements of the series—Fm, Md, No, Lr—do not exist in sizable quantity for condensed-matter studies and their ground-state properties are unknown). When combined in alloys and compounds, with different crystallographic structures and inter-atomic distances, all actinide elements can be localized or itinerant and even associate both characters [14]. This peculiar behaviour leads to a broad range of physical properties such as complex magnetism, multi-polar order, heavy quasiparticles, quantum criticality, possibly topological insulation… and of course superconductivity, the object of the present review.

The discovery of superconductivity in Hg by Kamerlingh Onnes in 1911 [15] has triggered a huge effort on basic research at low temperatures. Rapidly, it appeared that superconductivity was a rather common phenomenon in metallic materials and that the purity of the sample was important. Progress was then made in the purification of the elements and preparation of materials. After World War II, the results—half of the elements of the Mendeleiv Table superconduct at low temperature, some when pressurized—were extended to several thousands of alloys and compounds, for applied research and for industry, and leading to a phenomenological approach [16] before the appearance of a microscopic theory based on phonon coupling, called BCS theory [17].

Superconductivity is characterized by the complete loss of the electrical resistance, the expulsion of magnetic field and the opening of a gap at the Fermi energy below a critical temperature, T_{sc}, which is a characteristic of the material [18]. Superconductivity is destroyed by the application of a magnetic field that exceeds a critical value $H_c(T)$, a decreasing function of temperature T, and vanishes at $T = T_{sc}$. Superconductors fall into two classes: Type-I and Type-II. Materials of Type-I possess a single well-defined critical field, H_c, below which the sample is totally superconductor, whereas Type-II materials have two critical fields: a lower one, H_{c1}, which defines the field below which the magnetic flux is completely expelled from the material (Meissner–Ochsensfeld effect [19]). For fields above H_{c1} but below an upper critical field H_{c2}, the superconductor is in the mixed state: the magnetic flux penetrates it in the form of vortices, but supercurrent (i.e. electrical current with zero resistance) can still flow, without energy dissipation, which opens possibilities for industrial applications. The critical fields of superconductors vary widely, from rather low values in pure elemental superconductors, usually of Type-I (typically 10 mT), up to very high values (of the order of 100 T), for the high-T_{sc} ceramic materials.

In the BCS theory [17], superconductivity is described by the electron–phonon interaction characterized by a specific temperature θ_D, the Debye temperature, at the origin of the coupling of the superconducting Coopers pairs. This formalism gives basically a simple relation between T_{sc} and θ_D, $T_{sc} \sim \theta_D e^{-1/N_0 V_0}$, with N_0 the density of states at the Fermi level and V_0 the Coulomb repulsion term. The superconducting state is then described by an isotropic order parameter with a s-symmetry ($L = 0$). However, some superconductors, called “anomalous” or “unconventional”, exhibit properties that deviate from the BCS theory: d-symmetry ($L = 2$) or p-symmetry ($L = 1$), coexistence with magnetic order, “heavy” electrons, Cooper pairs not bound by phonons. Note that Cooper pairs with $S = 0$ (spin singlet) must have symmetric orbital wave functions ($L = 0, 2 \ldots$), whereas those with $S = 1$ (spin triplet) must have antisymmetric orbital wave functions ($L = 1, 3 \ldots$), all showing anisotropic features.

2. Interest and difficulties of actinides

Research on actinide superconductors does not simply contribute to the—important but restricted—field of actinide science, but also provides unique underpinning to the theory of superconductivity and more widely to the study of electron correlations: rare-earth and actinide intermetallic superconductors have allowed the first observations of unconventional superconductivity, with symmetry properties (p-wave or d-wave) of the order parameter different from those in usual metal superconductors (s-wave) and with electron-pairing mechanisms different from the case of classical electron–phonon interactions. Most actinide superconductors exhibit heavy-fermion characteristics, i.e. the effective mass of conducting electrons are considerably enhanced in these systems—typically 100 to 1000 times the free electron’s mass—reflecting their strong interactions with 5f electrons. Uranium superconductors also provide multiple model examples of magnetically ordered superconductors and the only few cases of coexistence of ferromagnetism and superconductivity observed so far. Finally, actinide superconductors offer the first possibility of experimental observation of another type of unconventional superconductivity predicted in 1964 [20], called FFLO and characterized by the spatial modulation of the order parameter.

Uranium does not pose any serious experimental problem, but difficulties are rapidly increasing when moving to heavier actinides. Transuranics are extremely difficult to investigate, due to their scarceness and radioactivity that does not only pose safety and security problems but also experimental problems such as self-heating (limiting access to low temperatures), self-damage (sample ageing), self-decay (sample purity). Table 1 lists the half-life, and self-heating power of most common actinide isotopes, from actinium to curium. Ac and Th have no 5f electrons and present low interest in the physics of actinides. Pa is rare and difficult to handle. The self-heating power drastically increases from ^{238}U to ^{237}Np. Then, it further increases (roughly by a factor of 100) for ^{239}Pu and again for ^{241}Am and ^{244}Cm. Therefore, to reach low temperatures with

Table 1
Radioactivity parameters of selected actinide isotopes.

Isotope	^{227}Ac	^{232}Th	^{231}Pa	^{238}U	^{237}Np	^{239}Pu	^{242}Pu	^{241}Am	^{243}Am	^{244}Cm	^{248}Cm
Half-life (y)	21.79	$14 \cdot 10^9$	32,760	$4.47 \cdot 10^9$	$2.14 \cdot 10^6$	24,110	374,000	432	7370	18.1	348,000
Power (mW/g)	36.3	$2.66 \cdot 10^{-6}$	1.441	$8.51 \cdot 10^{-6}$	$2.06 \cdot 10^{-2}$	1.93	0.117	114	6.43	2840	0.512

Source: Nucleonica (2014).

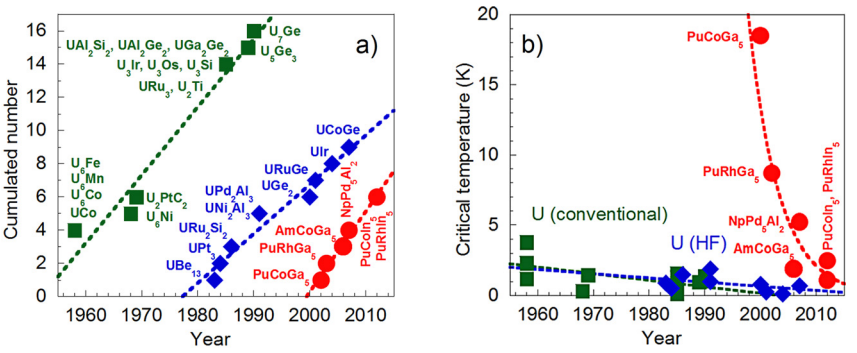

Fig. 1. (Color online.) Total number (a) and critical temperature (b) of conventional uranium (U) superconductors (squares), uranium heavy-fermion (HF) superconductors (diamonds) and transuranium superconductors (circles) as a function of their discovery year.

such materials, one must use very small samples and, whenever possible, a less active—but also less available—isotope (e.g. ^{242}Pu, ^{243}Am, or ^{248}Cm). Safety constraints increase when moving from uranium to neptunium, plutonium, americium and further, whereas the availability of the materials decreases in parallel.

This explains why condensed matter fundamental physics studies in transuranics are essentially limited to neptunium and plutonium. In the case of superconductivity, the challenge in transuranics is even bigger, since critical temperatures of actinide elements and uranium compounds are typically of the order of 1 K and physical measurements below 2–4 K are not routinely achieved in transuranium compounds. Nevertheless, since 2002, six transuranium superconductors compounds have been discovered (five of which belonging to the same "1 : 1 : 5" system). Some of them appear to have rare-earth analogues that also exhibit superconductivity. On the contrary, no superconductivity has yet been observed in any transuranium analogue of uranium superconductors.

Fig. 1a shows what we currently know about half as many transuranium superconductors as uranium heavy fermion superconductors. However, the rate of discovery of transuranium superconductors is comparable to—or even larger than—uranium superconductors and the difference in the current total of superconductors is due to the late discovery of the first transuranium superconductor (2002). Fig. 1b shows a more pessimistic trend: the average critical temperature of the new superconductors is decreasing over the years, gently in uranium but exponentially in transuranics, where it now reached the experimental limit (1–2 K) under which measurements on plutonium or heavier actinides become very challenging.

3. Superconductivity in actinides elements

The natural elements Th and U were produced in the early 1930's in pure phases, and even as single crystals for thorium [7]. At that time, the actinides concept had not yet been proposed as such and rather, these metals were associated with transition metals such as Hf or W [7]. It is only in the 1960 that actinide elements and specifically transuranium ones were produced in sufficient quantity and purity to ensure reliable studies at low temperature. The clarification of the superconducting properties of Pa has been realized when very pure metal was prepared at ITU [21] and first measurements below 1 K have been performed in a collaboration between ITU and LANL [22]. Efforts on actinide metals preparation [23] lead to the possibility to study the heat capacity of protactinium [24] and americium [25]. Several key parameters of the normal and superconducting state of Th, Pa, U and Am are given in Table 2. When positioning superconducting elements in a H_c vs. T_c diagram (Fig. 2a), one can notice that thorium and protactinium display a similar behavior to other BCS elements, while americium and the different phases of uranium metal present an enhanced critical field. Heavy actinides, starting from Cm up to Lr, are expected to be more localized and "magnetic" and non-superconducting. However, the coexistence of superconductivity and magnetic order has been later observed in numerous uranium compounds. Up to now, superconductivity has been observed in four actinide metals, listed hereafter.

Thorium is an ordinary BCS superconductor [26]. Although it belongs to the actinide series, thorium does not possess 5f electrons and its physical and chemical behavior, dominated by 6d electrons, is similar to that of transition metals such as Ti, Hf, or Zr [27]. Th crystallizes into a highly symmetric structure (fcc), in contrast with the other light actinides (Pa to Pu) that present structures with lower symmetry (see Table 2). Thorium metal is paramagnetic ($6d^27s^2$) with $\chi = +96 \cdot 10^{-6}$ mol $\cdot$ emu^{-1} at room temperature [28]. Its magnetic susceptibility is nearly temperature-independent, but depends on the amount of impurities [29]. Thorium metal is a weak BCS Type-I superconductor below $T_{sc} = 1.374$ K,

Table 2
Structural, electronic and superconducting parameters of actinides elements.

	Structure[a] space group	Lattice parameters (Å)	Density ($g \cdot cm^{-3}$)	γ[b]	θ_{Debye}[c] (K)	T_{sc} (K)	Type-I or II	H_c[d] (mT)	$\rho_{300\,K}$ ($\mu\Omega \cdot cm$)	dT_c/dp[g] ($K \cdot GPa^{-1}$)
Th	(C) $Fm\bar{3}m$	$a = 5.084$	11.72	4.3	160	1.374	I	16.2	15.8	−0.07
Pa	(T) $I4/mmm$	$a = 3.925$ $c = 3.238$	15.37	5.0	195	0.43	II	5.5	18	> 0
α-U	(O) $Cmcm$	$a = 2.854$ $b = 5.869$ $c = 4.955$	19.05	9.9	222	[0.1–0.78][f]	II	[30–74][f]	28	+1.3
β-U (Pt-stabilized)	(T) $P4_{2/mmm}$	$a = 5.656$ $b = 10.759$ $c = 10.759$	20.66	15	153[e]	0.8	–	–	–	–
γ-U (Mo-stabilized)	(C) $Im\bar{3}m$	$a = 3.524$	18.06	15.8	139	2.1	II	5000	67	–
α-Am	(H) $P6_3/mmc$	$a = 3.468$ $c = 11.241$	13.67	2.0	121	0.79	I	56	74	+0.25
γ-Am	(C) $Fm3m$	$a = 4.894$	13.67	–	–	1.1	–	–	–	+0.2

References indicated in the section *Superconductivity in actinide elements.*
[a] (C) stands for cubic, (T) for tetragonal, (H) for hexagonal, and (O) for orthorhombic.
[b] Unit for γ is in $mJ \cdot mol^{-1} \cdot K^{-2}$.
[c] Deduced from the low-temperature fit of heat capacity.
[d] $H_c(0)$ for Type-I superconductors, H_{c2} for Type-II superconductors.
[e] Deduced for β-U (Pt stabilized) from data in [71].
[f] Ranges of values obtained at ambient pressure.
[g] Slope values determined around 0 GPa in the linear regime of $T_c(p)$.

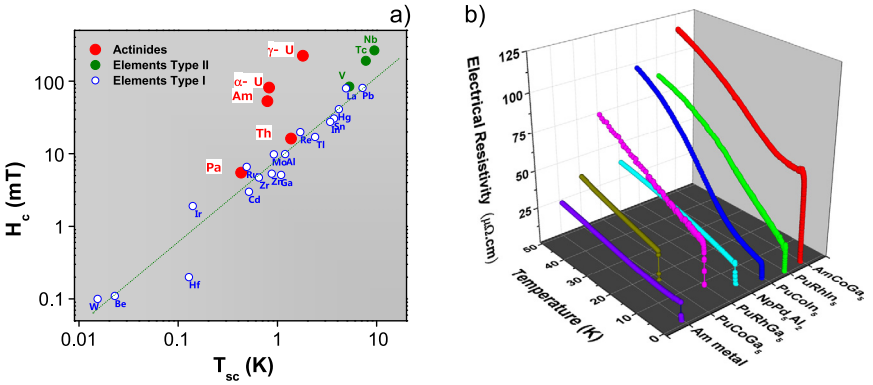

Fig. 2. (Color online.) (a) H_c vs. T_{sc} diagram for superconducting elements at ambient pressure. For Type-II elements, H_{c2} is used. There is no data for the β-U or γ-Am stabilized phases. (b) Superconducting transition determined by electrical resistivity of the transuranium superconductors [12,104,182,211, 218,219,228]. The intermetallic compounds have been sorted by decreasing T_{sc}.

with a critical field $H_c = 15.92$ mT [30]. It exhibits a complete Meissner effect and a critical field curve $H_c(T)$ with a parabolic temperature dependence. The specific heat anomaly at T_{sc} has been reported by several authors to be around 8.4 $mJ \cdot mol^{-1} \cdot K^{-1}$ [31–35]. The energy gap for Th when $T \to 0$ K is 3.53 $k_B T_{sc}$, and the discontinuity in the specific heat at T_{sc}, is 1.42 γ T_{sc} [36], in excellent agreement with BCS theory. Calculations on electron–phonon coupling have been also reported to be in good agreement with experimental data [37–40]. Some questions remain about a possible very weak f character deeply embedded in the electronic structure, as suggested for example by the non-linear dependence of T_{sc} on pressure [41,42] and the unexpectedly large metallic radius [43]. The calculated [44] wide empty 5f band lies just above the Fermi level, and could possibly hybridize with the conduction band, as supported by photoemission and Bremsstrahlung isochromat spectroscopy (PES and BIS) [45].

Protactinium with the bct structure is the first element of the actinide series presenting a real 5f character, as illustrated by its magnetic [46,47], thermodynamic [48,49], electronic and transport properties [50,51]. Pa is paramagnetic with $\chi = 190 \cdot 10^{-6}$ $emu \cdot mol^{-1}$ [46] and early data indicated superconductivity below 1.4 K [52]. It appeared later that pure metal presents a clear superconducting collapse only below $T_{sc} = 0.43$ K [22]. Despite its very low $H_{c2} \approx 5.6$ mT, Pa is reported as a Type-II superconductor. The initial discrepancy on T_{sc} has been attributed to a possible filamentary superconductivity or a stress effect on the crystallographic structure during metal preparation. It has then been suggested that Pa metal should

presents an increased T_{sc} when pressurized [53,54]. Stewart et al. [49] measured the heat capacity of a single crystal in the temperature range 4.9–18 K, temperature range limited by the self-heating of Pa and were not able to observe the heat capacity jump at T_{sc}. Interestingly, the unit cell volume of Pa metal first decreases when cooled down and then increases again at low temperature [55].

Uranium metal presents three distinct crystallographic phases (α, β and γ) below the melting point, preventing to grow easily single crystals: a transition from the "low-temperature" orthorhombic α-phase to the tetragonal β-phase at 668 °C and a transition from the β-phase to the body-centered-cubic γ-phase at 775 °C [56–58]. Uranium metal is weakly paramagnetic and exhibits a temperature-independent paramagnetism, with $\chi = 390 \cdot 10^{-6}$ emu $\cdot$ mol^{-1} [59]. The occurrence of superconductivity in the α-structure of uranium is still under debate. Superconductivity is observed at low temperature (< 1 K), but questions remain on its intrinsic nature to the α-structure. A wide range of T_{sc} has been observed at ambient pressure (from 0.1 to 1.3 K) [60–64] and recent works point out the presence of impurity defects or stresses at the origin of superconductivity [65,66], as suggested by the reduced superconducting parameters when samples are significantly purified [66,67]. Charge density waves have been observed in α-uranium [68]—which is quite unique for an element—and clearly compete with superconductivity in the metal, especially under pressure [69,70]. β- and γ-uranium are both superconductors [9,71], with sizable T_{sc} and enhanced related critical fields. These phases do not exist at room temperature and must be stabilized by a dopant (from 1% for β-U to 10–15% for γ-U!). β-U superconductivity was reported initially on Pt and Cr stabilized samples with $T_{sc} \approx 1$ K. γ-U has been more extensively studied, as T_{sc} is greatly enhanced and reaches 2.2 K using Mo or Nb as stabilizing element [9,72]. This value is similar to γ-U when pressurized (see below). Finally, we can mention an extended work on doping effect of γ-uranium by Tkach et al. [73] and the observation of superconductivity in splat cooling samples stabilized by Mo. The H_{c2} critical field is dramatically increased and reaches 5 T, which suggests that U metal lies in the dirty limit induced by the structural disorder by the Mo—U substitution [73].

Americium metal is indeed the first transuranium superconductor. The significant self-heating (see Table 1) restricted for a long time the temperature range for measurements of electrical resistivity [74], magnetization [75,76] and heat capacity [25]. The nature of the superconductivity of americium is somewhat puzzling. This element is located immediately after the "Mott transition" in the actinides series, and despite the fact that its 5f electrons are fully localized, americium's electronic configuration $5f^6$ ($J = 0$) leads to a paramagnetic state with a magnetic susceptibility almost temperature independent [76], without sizable local magnetic moment, but slightly enhanced at Room Temperature with $\chi = 780 \cdot 10^{-6}$ emu $\cdot$ mol^{-1} [76]. This situation suggested that americium metal could be superconducting [53], but its scarcity, radio-activity and self-heating were an experimental challenge to observe its rather small critical temperature $T_{sc} = 0.78$ K. Americium metal in the dhcp structure (room temperature) is a Type-I superconductor [77,78] with a rather large critical field $H_c = 56$ mT for an element and a so low T_{sc}. Interestingly, the high-temperature phase, when quenched (fcc) presents also a superconducting state with a similar critical temperature, $T_{sc} \approx 1$ K [77,79]. Heat capacity measurement at low temperatures [25,78] shows a very small electronic coefficient, $\gamma \approx 2$ mJ $\cdot$ mol^{-1} $\cdot$ K^{-2}, but the specific heat jump could not be observed, due to the self-heating.

Other transuranics: despite predictions of superconductivity [80] in α-Np and α-Pu metals, no clear signature of superconducting transition has been observed down to 0.5 K [81]. Smith et al. [82] mention the detection of superconductivity in Np metal at very low temperature, but it has not been confirmed by any further work.

Plutonium presents six allotropic phases. No hint of superconductivity has been observed down to 0.5 K in α-Pu [81]. The low temperature properties of "high temperature" phases such as β and ζ ones have been investigated by stabilizing the structure with doping by Os and U, respectively [83,84]. The β-phase stabilized by Os has been examined at low temperature, but did not show any hint of superconductivity down to 2 K [83]. For the δ-phase, Ga, Al, Ce or Am have been used [85–87], but here also, no hint of superconductivity has been observed.

Cm, Bk and Cf order magnetically. Cm is antiferromagnetic in the dhcp phase ($T_N = 64$ K) and ferromagnetic for the cubic (fcc) phase ($T_C \approx 200$ K). Bk was reported to be antiferromagnetic at $T_N = 25$ K and Cf, ferromagnetic at $T_C = 52$ K [88]. None of them are considered as potential superconductors since their magnetic ground state is analogue to that of rare-earth elements (Gd and beyond).

Superconductivity study of actinide elements under pressure from Th to Am: The pressure dependence of the critical temperature of thorium has been measured up to 20 GPa [42,89–91] and calculated [92,93]. T_{sc} decreases linearly with pressure down to a minimum around 7.4 GPa, followed by a slight increase up to 10 GPa, and again a decrease at higher pressures. This non-linear dependence is surprising and questions on a possible slight 5f character of carriers developing under pressure far from the structural transition reported at 80 GPa [94]. The pressure dependence of H_c [41] has been investigated, showing a direct correspondence between T_{sc} and H_c under pressure. The crystallographic structure of protactinium metal has been examined up to 130 GPa [95], showing a change of structure at 80 GPa. The itinerant nature of the 5f carriers explains the weak effect of pressure. As mentioned before, early low-temperature works on Pa suggest a possible increase of T_{sc} with pressure [53].

Pressure effects on uranium metal increase superconducting parameters, even stabilizing the superconducting state when initially not observed and heat capacity measurements at 1 GPa [96] have confirmed an enhancement of the 5f carriers' contribution. The maximal T_{sc} value for uranium is 2–2.3 K at pressure $\approx$ 1.0–1.1 GPa [97,98]. Recent work shows the strong pressure dependence of the electron–phonon coupling, whereas the Fermi-surface nesting is independent of pressure [70].

Excepted structural studies, no work has been reported yet on the properties of neptunium metal under pressure, especially at low temperature. Slight pressurization seems to be determinant to "enhance" the superconductivity state in 5f

actinide elements (U, Am, and probably Pa metals). Studies under pressure of high-purity Np metal at very low temperature should help clarifying the observation of Smith et al. [82].

Several phases of plutonium metal have been examined under pressure and at low temperature. For instance, the Am-stabilized δ-Pu phase, which, despite noticeable electronic correlations, does not show any hint of superconductivity [99]. Other phases such as β-Pu and ζ-Pu phase [83,84] would be interesting candidates because they present important electronic coefficients $\gamma \sim \delta$-Pu and will require very low self-heating isotopes for these fundamental studies.

The crystallographic structure of americium is extremely sensitive to pressure [100,101] and a complex structural diagram under pressure has been reported [102,103] with drastic collapses of the atomic volume cell. Considerable effort has been undertaken in the last decade to understand the link between structure and superconductivity and especially to access the very low temperature range below 1 K. When pressurized, T_{sc} of americium increases from 0.8 up to 2.3 K (6 GPa) [79] with an enhanced critical field [104] from 0.05 up to 1 T. The non-monotonic variations of T_{sc} vs. pressure and the probable change of superconductivity from Type-I to Type-II suggested by the drastic change of order of critical field [105] are reminiscent of superconducting state in U metal under pressure.

4. Uranium superconductors and their transuranium analogues

The first superconductors discovered in uranium compounds in the sixties and later can be described by "conventional" superconductors (Table 2). Rather sparse and vague information is available for transuranium analogues of conventional uranium superconductors: ^{237}Np Mössbauer spectroscopy showed that Np_6Fe does not order magnetically down to 1.9 K [106] and ^{57}Fe Mössbauer spectroscopy suggested the absence of magnetic order in Pu_6Fe down to 16 K [107]. The authors also mention [private communication from Adamson and Mortimer] that the resistivity behavior versus temperature of this compound is similar to that of pure Pu, with a higher residual resistivity. This indicates that Pu_6Fe is not superconducting in this (unknown) temperature range. $NpRu_3$ has been synthesized, but its magnetic properties have not been reported.

In the 1980's, a new class of uranium superconductors has emerged in the wake of the first heavy-fermion superconductor discovered in the rare earth cerium, $CeCu_2Si_2$ [108]: UPt_3, UBe_{13}, URu_2Si_2, etc., have critical temperatures similar to those of the previous uranium superconductors, but much larger effective masses (as illustrated by the Sommerfeld coefficient of the specific heat given in Tables 3 and 4). Furthermore, with the exception of UBe_{13}, these compounds also order magnetically ($T_{ord} > T_{sc}$) and both phases coexist below T_{sc}. Converging experimental and theoretical evidences indicate that 5f electrons are responsible for both superconductivity and magnetism, *d*- or *p*-wave symmetry of the order parameter and the pairing mechanism is thought to be mediated by magnons rather than phonons. A recent extensive review of actinide heavy-fermion superconductors has been made by C. Pfleiderer [109], completing a work by G.R. Stewart [110].

UGe_2, UIr, URhGe and UCoGe have the peculiarity to order ferromagnetically. The full coexistence of superconductivity and ferromagnetism, carried by the same electrons, has up to now only been observed in these few uranium systems, where spin-triplet (*p*-wave) superconductivity is probably realized. We should note that superconductivity and ferromagnetism have been observed in several rare earth-based compounds (Chevrel-phase $HoMo_6S_8$ ($T_{sc} \approx 1.8$ K, $T_C \approx 0.7$ K) [111], $ErRh_4B_4$ ($T_{sc} \approx 8.7$ K, $T_C \approx 0.9$ K) [112], $ErNi_2B_2C$ ($T_{sc} \approx 11$ K, $T_N \approx 6$ K, $T_C \approx 2.3$ K) [113]), but in these cases they are competing: the onset of ferromagnetic ordering rapidly destroys the superconducting state. The perovskite $RuSr_2GdCu_2O_8$ shows the coexistence of superconductivity and ferromagnetism ($T_{sc} \approx 16$ K, $T_C \approx 133$ K) [114] but Cooper pairs originate from the CuO_2 planes, while ferromagnetic order is associated with the Ru moments, in different layers.

Y_4Co_3 and its Co-rich variation Y_9Co_7 exhibit both ferromagnetism and (spin-singlet) superconductivity ($T_{sc} \approx 2.5$ K, $T_C \approx 4.5$ K) [115], but both physical phenomena compete, although their coexistence is made possible (at least in the range 1 K to 2.5 K) by some spatial separation of atom sublattices responsible for different phenomena [116].

The superconductivity initially reported in the ferromagnet $ZrZn_2$ [117] was later shown to be due to remnant of a superconducting layer induced by spark erosion [118].

Neptunium and plutonium analogues of practically all uranium heavy fermion superconductors have been investigated, while americium analogues are extremely rare, but none has been found to be superconducting. Instead, antiferromagnetic order is almost systematically observed, with a few exceptions (paramagnetism or ferromagnetism). However, it should be noted that measurements below 2–4 K are not common in transuranium compounds, i.e. the ground state of many of them might not yet be fully established. We review below each system.

AnBe$_{13}$: Superconductivity in UBe_{13} had been first observed in 1975 by Bucher et al., but ascribed to U filaments [129]. A decade later, Ott et al. established the presence of intrinsic, bulk superconductivity in this compound, with a critical temperature $T_{sc} \approx 0.85$ K [10]. Its important critical field (see Table 2) presenting a positive upturn at low temperature suggested for long time a possible FFLO state [130]. UBe_{13} also exhibits a huge Sommerfeld coefficient of the specific heat ($\gamma = 1100$ mJ $\cdot$ mol^{-1} $\cdot$ K^{-2}), the largest among actinide heavy fermions but, contrary to other uranium heavy-fermion superconductors, does not order magnetically. Its isostructural homologue $NpBe_{13}$ also displays a huge quasi-particle mass enhancement $\gamma \approx 900$ mJ $\cdot$ mol^{-1} $\cdot$ K^{-2} and orders antiferromagnetically at Néel temperature $T_N = 3.4$ K. This compound does not show any sign of superconductivity down to 0.080 K [131]. The low-temperature properties seem to be sensitive to the Be content: samples with excess Be have been found not magnetic, whereas optimal samples had $T_N = 4.9$ K [132]. ^{237}Np Mössbauer spectroscopy, magnetic susceptibility and neutron experiments evidenced a complex, modulated magnetic structure with wave vector $\mathbf{q}[1/3, 0, 0]$, moments perpendicular to the propagation direction, and two sublattices with moments perpendicular to each other. The magnetic moments carried by Np take two different values: 1.12 and 0.97 μ_B [133].

Table 3
Conventional uranium superconductors.

		UCo	U_6Fe	U_6Mn	U_6Co	U_6Ni	URu_3	U_3Ir	U_3Os	U_3Si[a]	U_2Ti	U_5Ge_3[b]	U_7Ge[b]	UAl_2Si_2	UAl_2Ge_2	UGa_2Ge_2	U_2PtC_2[c]
T_{sc} (K)		1.22	3.78	2.31	2.33	0.33	0.15	1.24	0.16	0.56	0.38	0.99	1.40	1.34	1.60	0.87	1.47
γ[d]		–	26	17	22	15	12.4	22	–	23	–	36	16	28	–	–	75
Structure[e]		(C)	(T)	(T)	(T)	(T)	(C)	–	(T)	–	(H)	(H)	–	(C)	(C)	(C)	(T)
Space group		$I2_13$	$I4/mcm$	$I4/mcm$	$I4/mcm$	$I4/mcm$	$Pm\bar{3}m$	–	–	–	$P6_3/mmm$	–	$P6_3/mcm$	$Pm\bar{3}m$	$Pm\bar{3}m$	$Pm\bar{3}m$	$I4/mcm$
Lattice parameters	a (Å)	6.356	10.303	10.312	10.323	10.390	3.977	–	–	–	4.828	8.56	–	4.145	4.219	4.218	3.52
	c (Å)		5.235	5.255	5.191	5.156					2.487	5.83					12.54
Ref.		[119,120]	[9,119]	[9,119]	[9,119]	[119,121]	[120,122]	[123]	[123]	[123,124]	[123,124]	[125]	[125]	[122]	[122]	[122]	[126,127]

[a] Three different crystallographic structures exist [124], the crystallographic structure of the superconducting phase [123] could not be identified.
[b] The existence of U_5Ge_3 and U_7Ge is questioned by Boulet et al. [128] (mixtures of U_5Ge_4 and U metal?).
[c] U_2PtC_2 is intermediate between heavy-fermion and less anomalous superconductors [127].
[d] Unit for γ is in $mJ \cdot mol^{-1} \cdot K^{-2}$ per U_{at}.
[e] (C) stands for cubic, (T) for tetragonal and (H) for hexagonal.

Table 4
Non-conventional uranium heavy fermion superconductors.

		UPt_3	UBe_{13}	URu_2Si_2[d]	UPd_2Al_3	UNi_2Al_3[e]	UGe_2[f]	UIr[f]	URhGe	UCoGe
T_{sc} (K)		0.54	0.9	1.5	1.9	1.0	0.8	0.14	0.27	0.8
H_{c2}[a] (T)		2.8/2.1	13	12/2.8	3.3/3.9	0.8/0.3	2.8/2.6/4.8	0.026//[101]	2.5/2/0.7	> 30/18/0.6
Order parameter		**p**	**p**	**d**	**d**	**p**	**p**	–	**p**	**p**
Symmetry										
T_{ord} (K)		5	–	17.5	14.5	5	53	46	9.5	3
Order type		AF	–	HO	AF	AF	F	F	F	F
μ_{ord} (μ_B)		0.01	–	0.03	0.85	0.24	1.5	0.5	0.42	0.05
γ[b]		440	1100	70	150	120	35	49	160	55
Structure[c]		(H)	(C)	(T)	(H)	(H)	(O)	(M)	(O)	(O)
Space group		$P6_3/mmc$	$Fm\bar{3}c$	$I4/mmm$	$P6/mmm$	$P6/mmm$	$Cmmm$	$P2_1$	$Pnma$	$Pnma$
Lattice parameters	a (Å)	5.764	10.248	4.128	5.382	5.207	3.997	5.62	6.875	6.845
	b (Å)						15.039	10.59	4.331	4.206
	c (Å)	4.899		9.592	4.189	4.018	4.807	5.60 $\beta = 98.9°$	7.507	7.222
d_{An-An} (Å)		4.12	5.13	4.1	4.0	4.0	3.8	3.3	3.5	3.5
Ref.		[139]	[168]	[169]	[147]	[170]	[171]	[172,173]	[159,174]	[161,174]

[a] H_{c2} values are given along the a, b and c axes, successively.
[b] Unit for γ is in $mJ \cdot mol^{-1} \cdot K^{-2}$ per U_{at}.
[c] (C) stands for cubic, (T) for tetragonal, (H) for hexagonal, (O) for orthorhombic and (M) for monoclinic.
[d] Reported as a possible multiband double gap system [169].
[e] UNi_2Al_3: measurements on thin films suggest isotropic H_{c2} and spin-singlet pairing [175].
[f] UGe_2 and UIr are pressure-induced superconductors. Superconducting parameters (T_{sc} and H_{c2}) are given for the optimal pressure—other parameters are given for ambient pressure.

Magnetic correlations around this wave vector have been sought in UBe_{13}, but in this compound, short-range antiferromagnetic correlations have been actually found at $\mathbf{q}[1/2, 1/2, 0]$ [134]. In $PuBe_{13}$ also, no superconductivity was detected (ac-susceptibility down to 0.4 K). An anomaly, attributed to a Kondo peak, is observed in the specific heat around 11 K [131]. *dc*-magnetic susceptibility shows a Curie–Weiss behavior with effective moment $\mu_{eff} = 0.74\ \mu_B$, which is consistent with electronic configuration $5f^5$ [135].

AnPt$_3$: UPt_3 becomes superconducting below $T_{sc} = 0.54$ K [136]. In addition, weak antiferromagnetic order sets in below $T_N = 6$ K with an ordered moment $\mu_U = 0.02\ \mu_B$ [137]. A remarkable feature of this compound is the existence of two distinct superconducting phases, plus a third one under applied magnetic field [138,139]. $NpPt_3$ crystallizes into the $TiNi_3$-type structure, which is different from the case of UPt_3, but belonging to the same space group $P6_3/mcm$ [140]. The compound shows a complex magnetic phase diagram, with the onset of two successive antiferromagnetic phases at $T_N = 30$ K and $T^* = 20$ K, with respective wave vectors $\mathbf{q}[1/3, 1/3, 0]$ and $\mathbf{q}[1/2, 0, 0]$. The ordered magnetic moments measured at 4.2 K amount to $\mu_{Np_1} = 1.85\ \mu_B$ and $\mu_{Np_2} = 0.5\ \mu_B$ [141]. It should also be noted that the low-temperature antiferromagnetic phase is destroyed by the application of a magnetic field (≈ 3 T). $PuPt_3$ crystallizes into the $AuCu_3$-type structure ($Pm\bar{3}$) and orders antiferromagnetically at $T_N = 40$ K [142].

AnRu$_2$Si$_2$: URu_2Si_2 is a heavy-fermion superconductor with $T_{sc} = 1.5$ K and $\gamma = 180\ mJ \cdot mol^{-1} \cdot K^{-2}$, which has a mysterious ground state below $T_o = 17.5$ K with a dipolar moment $\mu_U = 0.02\ \mu_B$. The order parameter has not been identified yet, and the phase has been called "hidden order" (HO). Under pressure, the ground state switches from the HO phase to an antiferromagnetic phase with "large" moment ($\mu_U = 0.4\ \mu_B$), whereas superconductivity is suppressed [143]. The application of high magnetic fields restores the HO phase [144]. The isostructural $NpRu_2Si_2$ counterpart orders at $T_N = 27.5$ K into an incommensurate structure ($\mathbf{q}[0, 0, 0.86]$) with a maximum moment $\mu_{Np} = 1.5\ \mu_B$ aligned along the *c*-axis, and partial squaring at low temperatures [145]. $PuRu_2Si_2$ also crystallizes into the $ThCr_2Si_2$-type structure and behaves as a Curie–Weiss paramagnet down to 1.3 K, with effective moment $\mu_{eff} = 0.75\ \mu_B$ [146].

AnPd$_2$Al$_3$: The largest ordered magnetic moment ($\mu_U = 0.85\ \mu_B$) in uranium heavy fermion superconductors was observed in the antiferromagnet UPd_2Al_3 ($T_N = 14.3$ K, $T_{sc} = 2$ K, $\gamma = 140\ mJ \cdot mol^{-1} \cdot K^{-2}$) [147,148]. The hexagonal isostructural $NpPd_2Al_3$ parent compound is not superconducting down to 1.2 K [149], but orders at $T_N \approx 40$ K into an incommensurate modulated magnetic structure with a wave vector $\mathbf{q}[1/, 1/3, 0.36]$. Below 25 K, a commensurate magnetic phase with $\mathbf{q}[1/, 1/3, 1/2]$ appears and becomes dominant [150]. The maximum ordered moment amounts to $\mu_{Np} = 1.67\ \mu_B$ [151]. $PuPd_2Al_3$ is paramagnetic down to 1.2 K [149].

AnNi$_2$Al$_3$: UNi_2Al_3 is an itinerant antiferromagnet ($T_N = 4.5$ K) with a very small magnetic moment ($\mu_U = 0.2\ \mu_B$) and an enhanced Sommerfeld coefficient of 120 $mJ \cdot mol^{-1} \cdot K^{-2}$, that becomes superconducting at $T_{sc} = 1$ K [152,153]. As UPt_3, but in contrast to UPd_2Al_3 and other antiferromagnetic heavy-fermion superconductors which are thought to be spin-singlet *d*-wave superconductors [154], ^{27}Al Knight shift measurements strongly suggest the occurrence of spin-triplet superconductivity in UNi_2Al_3 [155]. Resistivity measurements show that $NpNi_2Al_3$ orders at $T_{ord} = 23$ K, but no superconductivity was found down to 1.2 K [149]. To our best knowledge, the plutonium equivalent $PuNi_2Al_3$ has not been reported yet.

AnGe$_2$: UGe_2, with the base-centered orthorhombic $ZrGa_2$ crystal structure ($Cmmm$), is a ferromagnet with Curie temperature $T_C = 54$ K and an ordered magnetic moment $\mu_U = 1.48\ \mu_B$. When high pressure is applied to the system, the Curie temperature decreases and eventually vanishes around 1.7 GPa, but the most striking feature is the appearance of

superconductivity—and of its coexistence with ferromagnetism—between 1 and 1.7 GPa [11]. The Sommerfeld coefficient of the specific heat $\gamma = 35$ mJ · mol^{-1} · K^{-2} indicates that UGe_2 is a correlated metal, but the electron interactions are relatively weak. The $NpGe_2$ phase has not been reported, but the defect stoichiometry $NpGe_{2-x}$ phases have been obtained, crystallising either in the tetragonal $ThSi_2$-type ($I4_1/amd$) or hexagonal AlB_2-type ($P6/mmm$), for $x = 0.28$ and $x = 0.41$, respectively [156,157]. Both modifications of $NpGe_{2-x}$ order ferromagnetically at $T_C = 121$ K and $T_C = 157$ K and exhibit ordered magnetic moments $\mu_{Np} = 2.17\ \mu_B$ and $\mu_{Np} = 2.01\ \mu_B$, respectively. $PuGe_2$ crystallizes into the $ThSi_2$-type structure and is also a ferromagnet, with a Curie temperature $T_C = 35$ K [158].

AnRhGe: In the wake of UGe_2, URhGe displays the intriguing coexistence of ferromagnetism ($T_C = 9.5$ K, $\mu_U \approx 0.2\ \mu_B$) and superconductivity ($T_{sc} = 0.25$ K)—for the first time at ambient pressure [159]. URhGe crystallizes into an orthorhombic structure and has strongly correlated 5f electrons ($\gamma = 160$ mJ · mol^{-1} · K^{-2}). On the contrary, no superconductivity was observed down to $T = 1.8$ K in NpRhGe, but it orders antiferromagnetically at $T_N = 21.5$ K with an ordered moment $\mu_{Np} = 1.14\ \mu_B$ and shows a large $\gamma = 195$ mJ · mol^{-1} · K^{-2} [160]. PuRhGe was shown to remain a paramagnet down to 2 K [160].

AnCoGe: UCoGe also exhibits the coexistence of ferromagnetism and superconductivity ($T_{sc} = 0.8$ K) at ambient pressure [161]. The Curie temperature and magnetic moment are very small ($T_C = 3$ K, $\mu_U = 0.03\ \mu_B$) and UCoGe might be close to a quantum critical point. The linear term in the electronic specific heat, $\gamma \approx 57$ mJ · mol^{-1} · K^{-2}, is comparable to that observed in UGe_2. NpCoGe exhibits below $T_N \approx 13$ K a modulated, possibly elliptical, antiferromagnetic structure with an average ordered magnetic moment $\mu_{Np} = 0.80\ \mu_B$ [162]. The Sommerfeld coefficient amounts to $\gamma \approx 170$ mJ · mol^{-1} · K^{-2}.

"**1-1-1-1**": Finally, we can mention the transuranium analogues of the so-called "oxypnictides", a recent family of rare-earth superconductors. Some Fe- and Ni-based oxypnictides are superconducting at low temperature, like, e.g., LaOFeP ($T_{sc} = 4$ K) or LaONiAs ($T_{sc} = 2.75$ K) [163]. The critical temperature can be greatly enhanced by doping: $LaO_{1-x}F_xFeAs$ ($T_{sc} = 28$ K enhanced to 43 K under pressure ($P = 3$ GPa)) [164], $SmO_{1-x}F_xFeAs$ ($T_{sc} = 43$ K) [165]. These systems represent, together with the copper oxide "High-T_c" superconductors, the only class of superconductors with $T_{sc} > 40$ K. The neptunium analogue NpFeAsO was found to order antiferromagnetically ($\mu_{Np} = 1.70\ \mu_B$) at $T_N = 57$ K. This compound exhibits an Invar behaviour between 5 and 20 K and represents the first actinide-based antiferromagnet in which negative thermal expansion has been observed [166]. The Sommerfeld coefficient ($\gamma = 75$ mJ · mol^{-1} · K^{-2}) is in the lower range of heavy fermion values. PuFeAsO also shows an antiferromagnetic ground state ($T_N = 50$ K) [167]. The Sommerfeld coefficient is small ($\gamma \approx 4$ mJ · mol^{-1} · K^{-2}) and reflects the fact that the density of states near the Fermi energy has mostly Fe-*d* character. Both systems do not show any hint of superconductivity down to 1.8 K.

5. Transuranium superconductors

Until 2002, no superconductivity had been observed in transuranium-based compounds, except in neptunium Chevrel phase $Np_{1+x}Mo_6Se_8$ ($T_{sc} = 5.6$ K) [176]. However, this compound does not show bulk superconductivity (complete diamagnetic expulsion and zero resistivity). Morevover, in the rare-earth-based Chevrel phases [177], there is a clear distinction between the electrons carrying magnetism (4f) and the electrons at the origin of superconductivity (d) [178]. This microscopic distinction has also macroscopic effects with the coexistence at macroscopic scale of separated magnetic and superconducting domains. This suggests that the superconducting state observed in $Np_{1+x}Mo_6Se_8$ should probably be due to d electrons of the ligands and not to the 5f electrons of neptunium. Reinvestigating this system or related ones could help answering this problematic. Fig. 2b shows the superconducting transition determined by electrical resistivity for all transuranium superconductors (element and compounds) and Table 5 presents an overview of their superconducting parameters.

$NpPd_5Al_2$ is the first and only neptunium compound presenting clear 5f-electron superconductivity [13] (Type-II), with a critical temperature $T_{sc} = 4.9$ K and a large anisotropic critical field $H_{c2} \approx 16$ T and 4 T along *c* and *a* axes, respectively [13,182]. The 5f nature of the superconducting carriers and the non-conventional type of the ordering parameter have been demonstrated by several experiments such as heat capacity [13,182,183], and Nuclear Magnetic Resonance measurements [181]. The large Sommerfeld coefficient ($\gamma \approx 200$ mJ · mol^{-1} · K^{-2}) and transport properties measurements point to an inherent heavy fermion behaviour probably related to Kondo features [182–184], with strong anisotropic fluctuations [185,186], like for other cerium or uranium heavy fermion superconductors and probably due to the very anisotropic tetragonal crystallographic structure of $NpPd_5Al_2$. Several electronic structure models, considering magnetic spin fluctuations as the mechanism at the origin of superconductivity, have been proposed to explain the appearance of superconductivity in this system and to reproduce anisotropy features [187–189].

$PuCoGa_5$ is the first plutonium-based superconductor reported [12]. The remarkably high critical temperature ($T_{sc} = 18.6$ K) was one order of magnitude higher than all 4f (rare earth) or 5f (actinide)-based superconductors with f-electron Cooper pairs known at that time and especially the isostructural system $CeCoIn_5$ ($T_{sc} = 2.3$ K) [191]. Furthermore, the critical field of $PuCoGa_5$ is so large that it is still only possible to estimate it ($H_{c2} \approx 80$–100 T) [190]. The surprise was important, and synthesis and physical measurements of $PuCoGa_5$ initially performed at LANL and repeated at ITU confirmed the astonishing superconducting properties. To determine the order-parameter symmetry of the superconducting phase, one needs well-oriented single crystals and to access the low-temperature domain $\sim T_{sc}/10$. Such a high T_{sc} allows these extended studies possibilities despite the self-heating of ^{239}Pu. Important efforts have therefore been made to produce high-quality single crystals using gallium self flux [12]. Nuclear Magnetic Resonance [207,208] heat capacity

Table 5
Properties of transuranium superconductors compared to Nb_3Sn and $CeCoIn_5$.

		Nb_3Sn	$CeCoIn_5$	$NpPd_5Al_2$	$PuCoIn_5$	$PuRhIn_5$	$PuCoGa_5$	$PuRhGa_5$	$AmCoGa_5$
Structure		Cubic	Tetragonal	Tetragonal	Tetragonal	Tetragonal	Tetragonal	Tetragonal	Tetragonal
Space group		$Pm\bar{3}n$	P_4/mmm	I_4/mmm	P_4/mmm	P_4/mmm	P_4/mmm	P_4/mmm	P_4/mmm
Lattice parameters	a (Å)	5.293	4.614	4.148	4.574	4.621	4.235	4.301	4.233
	c (Å)		7.552	14.716	7.440	7.460	6.794	6.857	6.823
z			0.306	0.1467	0.3062	0.3021	0.3086	0.3064	0.3106
c/a		1	1.637	2×1.780	1.623	1.614	1.604	1.594	1.612
V_{cell} ($Å^3$)		147.1	160.8	2×126.6	155.7	159.3	121.9	126.9	122.3
T_{sc} (K)		17.8	2.3	4.9	2.5	1.6	18.6	8.7	1.9
H_{c2}[a] (T)		25	11.6, 4.9	3.7, 14	35[d], 9.5	23[d], 6.5	120[d], 100[d]	31[d], 17	N/A, 0.3[c]
$-\frac{dH_{c2}}{dT}$[a] (T/K)		1.6	24, 8.2	6.4, 31	17.6, 13.2	20, 15.8	10.0, 8.0	3.5, 2.0	N/A, 0.25[c]
γ ($mJ \cdot mol^{-1} \cdot K^{-2}$/f.u.)		52	290	200	250	350	77[e]	70[e]	3 <
Order parameter symmetry		s[b]	d	d	d	–	d	d	–
$\frac{dT_c}{dp}$[f] (K/GPa)		−0.14	+0.4	−0.3	–	–	+0.4	+1.1	+0.22
Ageing effect on T_{sc} (K/month)				∼ 0	−0.007	N/A	−0.25	−0.4	∼ 0
Ref.		[179,180]	[243–245]	[13,181,182,232]	[218,219]	[219,227]	[12,190,207,233]	[203,211,213,214,216,233]	[228,229]

[a] Values of H_{c2} and slope at T_{sc} are given along the a and c axes, successively.
[b] Reported as possible double s-gap system.
[c] Initial superconducting parameters determined at moderated pressure 0.35 GPa.
[d] Values extrapolated.
[e] Approximate values due to high T_{sc}, ageing effect and very high critical fields required to access the normal state. Some upper estimation reach twice these values [86].
[f] Slope values determined around 0 GPa in the linear regime of $T_{sc}(p)$.

measurements [190], muon spectroscopy [209], ageing studies [206] and more recently point contact spectroscopy [210] produced convincing evidence of unconventional superconductivity with probably, *d*-symmetry of the order parameter.

Magnetic susceptibility of first single crystals showed Curie–Weiss behaviour and the analogy with cerium analogue; $CeCoIn_5$ suggested that superconductivity would be mediated through spin fluctuations [192,193], and not through phonons [194]. However, the Curie–Weiss behavior and associated local moment were not observed on samples freshly synthesized [195] or made of the less radioactive isotope ^{242}Pu [196]. Instead, weak paramagnetism, reminiscent of δ-Pu metal magnetic features was noticed. Here, we can underline the relation with the tetragonal crystallographic structure of $PuCoGa_5$ which can be viewed as a δ-Pu cell distorted along the *c*-axis by the insertion of $CoGa_2$ layers.

Nevertheless, phonon-mediated superconductivity cannot be definitely ruled out in $PuCoGa_5$ [197]. It is unlikely to occur, considering, e.g., the "too high" critical temperature [198] or the fact that its U analogue $UCoGa_5$ is not superconducting, despite a phonon spectrum very similar to that of $PuCoGa_5$ [199]. More subtle mechanisms for superconductivity based on electron-spin fluctuations have been considered recently, for instance mixed valence or strong electron–phonon coupling [200–202].

The self-decay of ^{239}Pu creates damages and defects in the lattice and affects the physical properties: for instance, the superconducting transition decreases by 0.25 K/month [12,203–205]. Interestingly, this provides an opportunity to follow the collapse of superconductivity as a function of time (i.e. defects) in a single sample and compare it with different models [206].

$PuRhGa_5$ has been discovered shortly after $PuCoGa_5$ [211] and shows some similarities with it: isostructural, Type-II superconductor and high critical temperature ($T_{sc} = 8.7$ K). Its critical field, H_{c2}, is more anisotropic than in $PuCoGa_5$ [190,212] and estimated to be ~ 15 T and 27 T [213] or $H_{c2} \approx 17$ T and 31 T [214] along the *c*-axis and basal plane, respectively. Although the value of H_{c2} along the *c*-axis could fit with the Pauli limit, the linear temperature dependence of H_{c2} is inconsistent with both Pauli and orbital limits, but is well described by a two-band model of superconductivity [214]. Electronic correlations appear more important in $PuRhGa_5$ than in $PuCoGa_5$ [190,213,215,216]. The ageing effect is twice larger in $PuRhGa_5$ (–0.4 K/month) [206] than in $PuCoGa_5$. The superconducting state could be controlled by an anisotropic gap with *d*-symmetry [193,217]. But questions remain also on the Curie–Weiss law in the normal state as the properties of the compounds produced in single crystals vary rapidly with disorder [206]. Electronic structure calculations and models similar to those developed for $PuCoGa_5$ have been applied to $PuRhGa_5$ and suggest a stronger anisotropy of the Fermi surface, a rather localized aspect with Kondo-like interactions that could explain the larger sensitivity to disorder in the material [192,200–202].

$PuCoIn_5$ is isostructural to $PuCoGa_5$ and is a Type-II superconductor with $T_{sc} = 2.5$ K [218]. This critical temperature is much lower than in $PuCoGa_5$, but very similar to the case of $CeCoIn_5$ ($T_{sc} = 2.3$ K). The upper critical field H_{c2} amounts to 10 T along the *c*-axis and is estimated at ≈ 33 T in the basal plane. The Sommerfeld coefficient of the specific heat ($\gamma \approx 200\ \mathrm{mJ \cdot mol^{-1} \cdot K^{-2}}$) is larger than in $PuRhGa_5$ and $PuCoGa_5$. Single crystals of $PuCoIn_5$ have been obtained at LANL and ITU by indium self-flux. Heat capacity, electrical resistivity and magnetic susceptibility measurements have confirmed bulk 5f superconductivity in this material [218,219]. One might wonder why it took a decade to explore this direct analogue of $PuCoGa_5$ and $CeCoIn_5$ superconductors. In fact, early attempts to produce $PuCoIn_5$ by arc melting failed [220]. Even with the flux-grown single crystals, only repeated experiments and a careful analysis allowed us to identify the intrinsic properties of $PuCoIn_5$. Indeed, a magnetic anomaly was observed around 14 K and finally identified as the antiferromagnetic transition of $PuIn_3$ [219,221], present as an impurity in some samples. $PuIn_3$, previously reported as paramagnetic [223], has been produced in single crystals and revised as an itinerant antiferromagnet [221,222]. Finally, recent ^{115}In nuclear quadrupolar resonance experiments indicate *d*-wave superconductivity and the presence of strong spin fluctuations in $PuCoIn_5$ [224]. All these results point to a strong analogy with the 4f heavy-fermion systems such as $CeCoIn_5$ with Kondo features and localized f electrons [225]. Surprisingly, the self-decay of ^{239}Pu in the material does not shift significantly the T_{sc} with time (-0.007 K/month [226]).

$PuRhIn_5$ has been reported shortly after $PuCoIn_5$ [219]. It also crystallizes into the same tetragonal structure as other $AnCoX_5$ (X = Ga, In) compounds. $PuRhIn_5$ is a strong Type-II heavy-fermion superconductor with $T_{sc} \approx 1.6$ K [219]. There is much less information on this compound due to the experimental difficulties to reach such low temperatures in a plutonium-based compound. $PuRhIn_5$ seems to have the heaviest quasiparticles of all transuranium superconductors ($\gamma \approx 350\ \mathrm{mJ \cdot mol^{-1} \cdot K^{-2}}$) [227].

$AmCoGa_5$ becomes superconducting below $T_{sc} \approx 1.9$ K, with a critical field estimated at 0.3 T [228]. As for americium metal, the critical temperature is not significantly affected by sample ageing. Electrical resistivity measurements at such low temperatures in an americium compound were made possible using the ^{243}Am isotope and very small samples (both polycrystalline and single crystals) [228]. Unfortunately, due to the difficulty to cool the material down to very low temperatures, only transport measurements have shown the superconducting state. Heat capacity and magnetization experiments confirm the absence of magnetic order and indicate the absence of heavy quasiparticles ($\gamma < 3\ \mathrm{mJ \cdot mol^{-1} \cdot K^{-2}}$) [229]. Electronic structure calculations suggest a strong localization of 5f electrons in this system as for americium metal [230,231].

The effect of **pressure** has been examined for $NpPd_5Al_2$, $PuCoGa_5$, $PuRhGa_5$, and $AmCoGa_5$. The critical temperature of $NpPd_5Al_2$ collapses down to zero when the applied pressure increases up to 6 GPa [232]. On the contrary, in both Pu-based systems, T_{sc} increases with pressure and reaches a maximum value, around $p \approx 10$ GPa ($T_{sc} \approx 21$ K in $PuCoGa_5$ and $T_{sc} \approx 17$ K in $PuRhGa_5$) [233]. When the pressure is further increased to ≈ 20 GPa, the critical temperature of $PuCoGa_5$ remains stable, whereas in $PuRhGa_5$, T_{sc} decreases back to its ambient pressure value. An analogy can be drawn with the

Ce-115 counterparts, suggesting common mechanisms driving the superconductivity in Pu and Ce systems [234], but on a much higher pressure scale, taking into account the wider extension of the 5f orbitals by comparison to the 4f orbitals. Finally, $AmCoGa_5$ shows a pressure dependence similar to that observed for Am metal [228]; the critical temperature increases continuously with pressure, from 1.9 to 2.6 K at $p = 3.2$ GPa. The critical field also increases from 0.3 to 0.8 T at the maximum pressure achieved.

6. Actinide or rare-earth analogues of transuranium superconductors

The transuranium superconductors have so far no actinide analogue also showing superconductivity; however, some rare-earth analogues (mostly cerium-based ones) do exhibit superconductivity.

$AnPd_5Al_2$: the study of $NpPd_5Al_2$ has been extended to other actinides (Th, U, Pu, Am) [235–238]. Some of these isostructural compounds order magnetically, but none exhibit superconductivity down to 2 K. For example, $PuPd_5Al_2$ orders antiferromagnetically with a Néel temperature ($T_N = 5.6$ K) comparable to the critical temperature of $NpPd_5Al_2$ [237]. Curiously, the tetragonal 1–5–2 structure cannot be obtained with other transition metals [239]. UPd_5Al_2 has a non-magnetic ground state and an effective moment ($\mu_{eff} = 3.4\ \mu_B$) close to the free-ion value ($\mu_{eff} = 3.6\ \mu_B$) [235], while its electronic specific heat coefficient amounts to $\gamma \approx 20\ \text{mJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$. $AmPd_5Al_2$ does not show any sign of magnetic nor superconducting transition down to 2 K [238]. Surprisingly, only few rare-earth representatives [240] of this family had been reported before. In particular, $CePd_5Al_2$ [241] was reported after the $NpPd_5Al_2$ discovery. $CePd_5Al_2$ is an antiferromagnet showing two successive transitions at $T_{N_1} = 4.1$ K and $T_{N_2} = 2.9$ K [241] and the onset of superconductivity ($T_{sc} = 0.57$ K) under applied pressure ($9 < p < 12$ GPa) [242].

$AnTIn_5$: **$CeRhIn_5$** is a heavy fermion ($\gamma \approx 400\ \text{mJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$) antiferromagnet ($T_N = 3.8$ K). The application of high pressure (1.5 GPa) induces a superconducting state with $T_{sc} = 2.1$ K [243]. **$CeCoIn_5$** stands for the archetype of the 1–1–5 family. It is not ordered magnetically, but superconducting at ambient pressure ($T_{sc} = 2.3$ K) [244], which was a record at that time. The large value of $\gamma \approx 290\ \text{mJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$ indicates substantial mass renormalization. **$URhIn_5$** orders antiferromagnetically at $T_N = 98$ K and its Sommerfeld specific heat coefficient amounts to $\gamma = 50\ \text{mJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$ [245], whereas **$UCoIn_5$** is an antiferromagnet with $T_N = 98$ K [245].

$AnTGa_5$: the cerium analogues do not exist. $UCoGa_5$ and $URhGa_5$ are paramagnets [246] and mixed valence was reported in $UCoGa_5$ [247]. $NpCoGa_5$ [248–250] and $NpRhGa_5$ [249,251] are anisotropic Type-I antiferromagnets (**q**$[0, 0, 1/2]$) with magnetic moments pointing along the c-axis and ordering temperatures $T_N = 47$ K and $T_N = 37$ K, respectively. $NpRhGa_5$ undergoes a second magnetic transition—corresponding to the reorientation of the magnetic moments from the c-axis to the basal plane—at $T^* = 32$ K. In both compounds, the antiferromagnetic phase is found to be more fragile when the magnetic field is applied along the c-axis, which is analogous to the field behaviour of the superconductivity in $PuCoGa_5$ and $PuRhGa_5$. $NpCoGa_5$ and $NpRhGa_5$ carry ordered magnetic moments with reduced values ($\mu_{Np} = 0.84\ \mu_B$ and $\mu_{Np} = 0.96\ \mu_B$, respectively) compared to the Np^{3+} free ion ($2.4\ \mu_B$). The Sommerfeld specific heat coefficient of both compounds is moderately enhanced ($\gamma = 64\ \text{mJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$ and $\gamma = 50\ \text{mJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$, respectively).

Chemical substitution induces slight changes in the lattice parameters and electronic structure of the investigated materials. In the case of heavy-fermion superconductors, it allows one to tune the system along the "Doniach diagram" [252], resulting in the variation—in extreme cases, the appearance or disappearance—of the superconductivity or magnetic order. When the magnetic ordering temperature is approaching the absolute zero, thermal fluctuations are vanishing and the properties of the system are driven by quantum fluctuations. This region of the Doniach diagram is called "quantum critical point" and unconventional superconductivity usually appears in the vicinity of quantum criticality.

Substitution in uranium superconductors have been performed with neptunium all the way up for URu_2Si_2 and UPd_2Al_3 to pure $NpRu_2Si_2$ and $NpPd_2Al_3$. Unfortunately, the critical temperature of these uranium superconductors is very small and cannot be easily observed, but seems to rapidly decrease with the Np substitution [149,253]. The magnetic ordering temperature is much more accessible and can be monitored on the whole substitution range. Neptunium compounds have much higher ordering temperatures than their uranium counterparts, in the same order of magnitude as the enhancement of the de Gennes factor from U to Np (1.7). For example, the ordering temperature increases monotonously from URu_2Si_2 to $NpRu_2Si_2$, although the "hidden order" phase ($\mu_U = 0.02\ \mu_B$) is immediately (already in $U_{0.95}Np_{0.05}Ru_2Si_2$) replaced by the "large-moment" ($\mu_U \approx 0.4\ \mu_B$) phase induced by the ordered moment carried by neptunium ($\mu_{Np} \approx 1.5\ \mu_B$) [254–256]. In $U_{1-x}Np_xPd_2Al_3$, the situation is different, although both pure U and Np compounds are antiferromagnetic: the Néel temperature globally increases from 14 K in UPd_2Al_3 to 40 K in $NpPd_2Al_3$, but shows a pronounced minimum ($T_N \approx 4.5$ K) for $x \approx 0.3$. This unusual behaviour arises from the magnetic anisotropy with different easy axis in UPd_2Al_3 (moment in the *ab* plane) and $NpPd_2Al_3$ (moment along the c-axis). Contrary to the constant neptunium moment observed in $U_{1-x}Np_xRu_2Si_2$, the magnetic moment carried by neptunium in $U_{1-x}Np_xPd_2Al_3$ remains very weak ($\mu_{Np} \approx 0.2\ \mu_B$) from $x \approx 0.1$ to $x \approx 0.3$ and then increases up to $\mu_{Np} = 1.7\ \mu_B$ for $x = 1$ [254].

Substitution in $PuCoGa_5$, the transuranium superconductor with the highest critical temperature have been performed by actinide and transition metals [257]. In all cases, the critical temperature decreases with chemical substitution. However, the isoelectronic substitution (preserving the electrons count) is the least destructive for superconductivity. Indeed, non-isoelectronic and in particular actinide substitution (substitution of Pu by U or Np) dramatically affects the critical parameters—much more than the simple volume effect—which suggests that 5f electrons are crucial in driving the

superconductivity. The latter vanishes so rapidly that no coexistence with magnetic order (expected for larger Np amounts) could be observed.

7. Conclusions and perspectives

The last three decades have been particularly fertile in the field of superconductivity with key discoveries: organic superconductors [258] high-T_{sc} cuprates [259], nano-superconductors [260] and in the field of actinides: heavy-fermion superconductors, ferromagnetic superconductors, and finally transuranium superconductors, of which six have been discovered to date. Their critical temperatures are relatively high, at the exception of the last-discovered ones, $PuCoIn_5$ and $PuRhIn_5$, close to the experimental limit for plutonium and $AmCoGa_5$. Transuranium superconductors known so far do not require very high sample purity (as indicated for example by the room-temperature Residual-Resistivity Ratio), unlike, e.g., URhGe. However, transuranium superconductors with low critical temperature and high sensitivity to purity and damages might exist, but will be extremely hard to detect. Due to the peculiar nature of their main constituent, actinide-based superconductors might not have applications. However, their unique properties challenge and boost models and theory about superconductivity. Actinide elements are not only the frontier of the periodic table, but also the "elements of surprise" and we can wonder what will come next. Another actinide superconductor with higher critical temperature than $PuCoGa_5$ ($T_{sc} = 18.5$ K, record for f-electron superconductivity) would certainly be an important discovery and allow extensive studies. In transuranics, much might still remain to be discovered: for example, no pressure-induced superconductivity has been reported. Surprisingly, the coexistence of superconductivity with magnetic order has also not been yet observed in neptunium, plutonium or americium systems, although it is so common in uranium. Considerable experimental and theoretical efforts have been made to understand the superconductivity of transuranics. The symmetry of the order parameter has been identified with a reasonable evidence, despite the lack of measurements at very low temperature ($< T_c/20$), the pairing mechanism is still under debate ($PuCoGa_5$, $PuCoIn_5$) [218]. While spin fluctuations have been observed in uranium superconductors, e.g., UPd_2Al_3 [261–263] and suggested for $NpPd_5Al_2$ [185,186], they have failed to be detected in plutonium compounds. However, these experiments are really demanding and hard to perform in transuranics requiring large single crystals for neutron inelastic scattering and special isotopes. Indeed, the most common isotope ^{239}Pu is not the best candidate as it strongly absorbs neutrons. Experimental research has explored neptunium and plutonium analogues of uranium and rare-earth superconductors (recent examples are the "1 : 1 : 1" [162] and "1 : 1 : 1 : 1" [166] systems), but none of these attempts have led to the discovery of new transuranium superconductors, although other interesting properties have been found (spin spiral magnetism, Invar effect). Up to now, only the "1 : 1 : 5" and "1 : 5 : 2" families have shown superconductivity in both the rare-earth and transuranium analogues ($PuCoIn_5/CeCoIn_5$, $NpPd_5Al_2/CePd_5Al_2$). Moreover, when substituting an actinide by another one in a transuranium superconductor, the properties of the system are dramatically affected, apparently more than what could be expected from a slight electronic or volume change. There are clearly many finely balanced energy differences in these systems, leading to completely different resulting ground states. This point should be taken into account by theories aiming at predicting the behavior of analogue systems with actinide substitution. Finding the right parameter(s) would be a major advance in condensed matter physics. Finally, very few condensed mater physics at low temperature have been performed on transamericium elements and compounds such as Cm, Bk, and Cf (alloys or oxides). Much remains to be done in the exploration of the basic properties of these materials as they are present (and problematic) in irradiated fuels and nuclear wastes.

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