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ACTINIDES: FROM HEAVY FERMIONS TO PLUTONIUM METALLURGY

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ABSTRACT

The actinide elements mark the emergence of 5f electrons. The f electrons possess sufficiently unusual characteristics that their participation in atomic binding often result in dramatic changes in properties. This provides an excellent opportunity to study the question of localization of electrons; a question that is paramount in predicting the physical and chemical properties of d and f electron transition metals. The transition region between localized (magnetic) and itinerant (often superconducting) behavior provides for many interesting phenomena such as structural instabilities (polymorphism), spin fluctuations, mixed valances, charge density waves, exceptional catalytic activity and hydrogen storage. This region offers most interesting behavior such as that exhibited by the actinide compounds UBe_{13} and UPt_3. Both compounds are heavy-fermion superconductors in which both magnetic and superconducting behavior exist in the same electrons. The consequences of f-electron bonding (which appears greatest at Plutonium) show dramatic effects on phase stability, alloying behavior, phase transformations and mechanical behavior.

Key Words: Heavy Fermions, Plutonium Metallurgy, Metallic Bonding, Actinides, Structural Instabilities.

INTRODUCTION

The actinide elements mark the emergence and filling of the 5f-electron shell. Because the heavier actinide elements have extremely short half-lives, the dominant feature of the physical properties of the best known actinide elements is the gradual onset of localized behavior. While the 3d-electron elements never do show local moment behavior, the 4f-electron lanthanide elements almost go through the complete localization process at cerium, the first element. Thus, the actinides, which switch over to local moment behavior at americium, are the ideal series in which to observe the effects of the onset of localization and loss of bonding. When f electrons do form an energy band, their lower symmetry is generally believed to be responsible [1] for the complex crystal structures and reduced melting points seen in the elements, again making the localization (or loss of the f-electron energy band) easier to follow.

In Fig. 1 we show the d- and f-electron elements arranged according to the onset of localization [2]. The d- and f-electron wavefunctions shrink across their series and may finally fall within the inert gas core, which allows an atomic-like local moment in the condensed phases. This happens at different rates for each series, and Fig. 1 merges all of this into a table. In the superconductivity region, hybridization of the d- or f-electron energy levels forms them into energy bands, and superconductivity is the usual ground state at low temperatures. In the magnetism region, local moments occur and tend to order magnetically at low temperatures. In the diagonal, crossover region the electrons are on the edge of localization leading to uncertainty in the ground state. This uncertainty makes the detailed electronic properties sensitive to perturbations.

This sensitivity to perturbations shows up in many physical properties. The number of allotropic crystal structures is a maximum for elements at the diagonal. Generally, structural instabilities are more common in materials made of elements from this region. Catalysis and hydrogen storage also occur more readily in materials whose electrons are on the edge of localization because their bonding properties are easily modified. Uncertainty in the ground state also leads to charge density waves, spin density waves, itinerant electron magnetism, and some forms of mixed valence at low temperatures. In other words, it is the nature of electrons (as fermions) to attempt to lower their energy (as a Fermi gas) at low temperature. The most simple and common ground states are superconductivity and magnetism, which lower the densities of states of electrons at the Fermi energy. When the details of the electronic structure makes these phenomena unfavorable, more complex (to observe and to explain) behavior occurs. At the crossover region this is aggravated by the presence of a partially filled energy band that is thus both relatively narrow and pinned to the Fermi energy. This, in fact, offers a wide range of charge, spin, or ion position instabilities as possible ways to lower this high density of states. Different scientific disciplines study aspects of this phenomena without acknowledging the greater generality. More important, this is the part of the periodic table where new science (new types of behavior) should be sought. It seems obvious that more exotic electronic behavior will be found at this diagonal than elsewhere. The new heavy fermion materials, which we briefly discuss later, are the most recent demonstration of this. It must be emphasized that Fig. 1 is only a qualitative guide to electronic behavior. It explains general trends, not detailed behavior.

PHYSICS OF ACTINIDE COMPOUNDS

Figure 1 shows only elements. The application of pressure to elements near the diagonal shifts their behavior towards superconductivity because of increased band formation. On the other hand dilution of these elements in alloys or compounds generally interferes with hybridization, increases the lattice spacing, and makes the materials more magnetic. Because there are many different atoms that can be added and many crystal structures that can be formed, the formation of compounds offers the greatest avenue to study hybridization and explore new behavior, which is a goal of this conference.

In 1970, Hill [3] demonstrated that for cerium, uranium, neptunium, and plutonium there was a critical spacing that marked the division between superconductivity and magnetism, or the point of f-electron localization. This was seen only as a necessary condition in that these ground states did not always occur. Also if other atoms remained magnetic, then magnetism could occur in the superconductivity region. The importance of this was that it demonstrated that these four elements were not like the heavy rare earths with local moments, but had behavior determined by hybridization of their f electrons. The most surprising aspect of the plots of f-electron atom spacing versus behavior was that they showed behavior largely independent of the other atoms in compounds, which is to say, independent of the first nearest neighbor atoms. In the intervening years these ideas have been essentially confirmed by many later measurements. However, the number of exceptions, which are understandable on an individual basis, has also been slowly climbing. The class of heavy fermion superconductors now represent behavior that is incomprehensible within the context of these plots. That magnetism could be destroyed by some extra hybridization with other atoms when the f-electron atoms were far apart is consistent with a necessary but not sufficient condition for magnetism. That some of these compounds would become superconducting is truly an exception. Indeed, this is simply one indication that the superconductivity may be a new type and result from a spin-spin interaction, rather than the usual phonon-mediation.

Thus, the most surprising behavior found for actinide compounds in recent years is the superconductivity of UBe_{13} [4] and UPt_3 [5] which have uranium spacings exceeding 5.1 Å and 4.1 Å, respectively. The compound $CeCu_2Si_2$ was found to be superconducting five years ago [6], but variations in properties among samples slowed general acceptance of the superconductivity. The superconducting and normal state properties of UBe_{13} are remarkably similar to those of $CeCu_2Si_2$, while in most other respects the compounds are very different (for example cubic versus tetragonal, 5f-electron versus 4f, congruent melting versus peritectic, p-band versus d-band, binary versus ternary). Thus UBe_{13} clearly demonstrated that, taken with $CeCu_2Si_2$, there was indeed a new class of very strange superconductors. Next, UPt_3 was added to this class, and yet it had rather different properties [7]. These compounds have been dubbed "heavy fermion superconductors" because their most dominant low temperature property is an effective mass of the superconducting electrons (fermions) that is about 200 times heavier than free electrons. This is deduced from the extremely large electronic heat capacity that is seen to occur below about 10 K. The f electrons seem almost localized (infinite effective mass) near room temperature because they exhibit a Curie-Weiss magnetic susceptibility, but at low temperature an extremely narrow band develops showing that the f electrons are just barely delocalized. While electronic band structure calculations cannot account for all of this behavior, they do suggest that the relatively small amount of hybridization occurs because other bands have been filled, which moves them below the Fermi energy, while the f band is of course pinned there because it can contain fourteen electrons [8]. Several questions are obvious: how does such a narrow band form; is that mechanism the same for 4f and 5f electrons; what is the mechanism for superconductivity; and is the superconducting electron pairing different from most superconductors? These questions have been posed by others in far more detail at this conference [7,9,10], while the answers will likely remain a subject of controversy for a long time.

Another interesting development is the discovery of U_2Zn_{17}[11], $NpBe_{13}$ [12], and UCd_{11} [13] as heavy fermion magnets. They are very similar to the superconductors in their properties except that instead of becoming superconducting below 1 K, they order magnetically below 10 K. The precise nature of the magnetic state is not even known yet. The intriguing thought is that because all of these compounds are caught on the edge of localization, their electronic properties are scaled up by more than two orders of magnitude, and yet, at low enough temperatures the same division into ground states is seen as is shown in Fig. 1. It is a strong confirmation of our fundamental understanding of fermions and suggests that there are more compounds to be found that will be interesting.

PHYSICS OF ACTINIDES

The gradual formation of an f-electron band near protactinium, its narrowing through to plutonium, and its loss from the onset of localized f-electrons just past plutonium can be easily identified in Fig. 2 where binary alloy phase diagrams have been connected [2]. As is discussed later, the reduction of symmetry in crystal structures, the larger number of these structures, and the lowering of the melting points are the signatures of the filling and gradual localization of the f-electron band [1]. Superconductivity at the beginning of the series disappears at neptunium and plutonium, because of the increase of magnetic correlations in the narrowed f band. The next element, americium, is shown to have localized f electrons not because it is magnetic, but rather because its unique $5f^6$ configuration gives a precisely nonmagnetic $J=0$ ground state. The exactness of this is clear from the superconductivity that is observed in americium [14], which strangely and uniquely is thus a demonstration of localized f electrons. The properties of the remainder of the actinides are understandable by their similarities to the lanthanides beyond cerium and will not be discussed.

The generalization to compounds and other alloys of the band properties shown for actinide alloys in Fig. 2 is less obvious and is most easily seen by consideration of atomic radii or volumes within the materials. It should be noted that our use of the concept of energy bands is quite general (as is obvious from the ideas derived from Fig. 2) and is not limited to the concepts of one-electron potentials used for energy band calculations. So the formation of f bands and their contribution to bonding in compounds and alloys is seen as a reduced volume for the f electron atoms [15]. An equivalent point of view is that hybridized f electrons are less efficient at screening nuclear charge than are localized f electrons, and they lead to a general contraction of the electron orbitals. Thus in Fig. 1, the nonmagnetic elements are smaller than they would be without hybridization, while the magnetic elements show only the general contraction of atoms as a shell is filled. This is the general contraction that leads to the crossover to the localized behavior.

It is true that in compounds that have localized f electrons, the corresponding 4f- and 5f-element compounds have the same lattice parameters (of course, assuming they are isostructural) [15]. This permits the ready identification of actinide compounds with hybridized 5f electrons by a simple comparison with the same lanthanide compounds. It is sometimes thought that the situation for alloys is more complex. For example, in Pu-Aℓ alloys which have aluminum atoms located substitutionally on sites in the α-phase (a nonequilibrium situation), the lattice is expanded by their presence, even though the aluminum atoms are smaller than the α-Pu atoms [16]. However, all of the size considerations become simpler if it is appreciated that the foreign atoms must interfere locally with the f-electron hybridization and make the neighboring plutonium atoms larger. This can even be quantified by arguing that four atoms are so modified in α-Pu. This raises interesting questions about local symmetry, diffusion rates, and the identification of aluminum as a "substitutional" impurity.

The resistivity of metallic materials as a function of temperature is an important parameter. For actinide materials these curves are reasonably well known, but there is little detailed understanding [17]. We will not review this but will comment on the impact of heavy fermion materials on this situation. Generally actinide materials in which the f electrons are hybridized have high resistivities and show a great deal of low-temperature electron scattering. This is clearly due to relatively narrow energy bands, but there is no generally accepted model that offers detailed insight. Heavy fermion materials have significantly narrower bands, which are evidenced by more electron scattering to even lower temperatures and by the presence of sharp features at low temperatures. This means that heavy fermion materials are the limiting case for narrow bands and, in some sense, are thus the opposite of a free electron gas. Limiting cases are simpler to calculate than more general cases. Thus heavy fermion materials may now provide the key to theoretical descriptions of materials at the crossover (which are of great technological importance) because their dramatic yet simpler properties will stimulate theoretical descriptions that can then be generalized to the wider class of narrow band materials.

Two more properties that are important to tracking the physics of metallic materials are specific heat and cohesive energies. It is specific heat that quantifies the narrowness of bands at low temperatures and made possible the identification of the extreme nature of heavy fermion materials. It is the cohesive energies of materials that put the behavior shown in Fig. 1 on a quantitative basis and permit more accurate comparisons of materials. Stewart [18] has recently reviewed heat capacity of heavy fermion and related materials, and Ward, Kleinschmidt, and Peterson [19] have discussed thermodynamic properties in detail.

Clearly the narrow energy bands in the actinides dominate the physical properties. Heavy fermion materials have highlighted this, and their unexpected properties have stimulated thinking along these lines and demonstrated how little work has actually been done on actinide compounds. Alloys tend to pose different problems and are discussed next.

PHYSICAL METALLURGY OF ACTINIDES WITH EMPHASIS ON PLUTONIUM

The physical metallurgy of the d-electron transition elements has been studied extensively because these elements or their alloys represent a large number of materials of significant commercial interest for structural, electronic and magnetic applications. The metallurgy of 4f elements has received much less attention. These elements are of interest commercially for magnetic applications and as dilute alloying elements, but they are not used as structural materials by themselves. Some of the consequences of f-electron bonding have been noted in cerium where an isostructural transformation occurs under pressure from one fcc phase to another with a 15 percent volume change [20]. In the 5f elements, localization occurs gradually over a number of elements, thereby making it possible to study the metallurgical consequences of 5f-electron bonding. Although the actinides are of interest primarily for their nuclear properties, uranium and plutonium are also important structural materials in these applications for which chemical, physical and mechanical properties play a vital role. Uranium has three allotropes and some very interesting metallurgical properties [21]. However, the most complex and fascinating of the actinides is plutonium. We will confine our remarks in this paper to the metallurgy of plutonium.

The most interesting and important manifestation of f-electron bonding in plutonium is the existence of six solid-phase allotropes at ambient pressure (a seventh appears with the application of modest hydrostatic pressure). As pointed out above, the f electrons in plutonium are on the verge of localization and are very sensitive to external influences. The ground-state energies for the different phases differ only slightly and, hence, small external perturbations such as temperature changes cause changes in crystal structure. Since the actinides involve turning the bonding of f electrons on or off, the consequences of such crystallographic transformations are dramatic. In plutonium one finds that decreasing the f bonding causes transformations from low-symmetry crystal structures to cubic (high-symmetry) structures, often with enormous volume changes. Such changes in crystal structure affect the resulting properties of plutonium significantly.

The thermal instability of plutonium is best illustrated by a dilatometer trace, a plot of length change during heating [22]. Figure 3 compares the behavior of plutonium with that of iron. Transformations in plutonium occur readily with increasing temperature. Most are accompanied by large length and thus volume changes. Such volume changes are difficult to accommodate in solids at relatively low temperatures without loss of physical integrity, which would be catastrophic for a structural material. The volume change upon melting is negative contrary to that of most elements. This results from the fact that f electrons bond quite easily in the liquid because its less rigid structure increases rotational freedom [1]. This enhancement of bonding promotes the stability of the liquid, lowers the melting point, and promotes a high-density liquid phase. The volume expansion upon freezing is advantageous during casting since solidification is not accompanied by the usual shrinkage defects.

The thermal expansion coefficients of the α, β, γ, and ε phases are abnormally large because the normal expansion due to lattice vibrations is accompanied by increasing atom sizes caused by decreasing f bonding with increasing temperature. The expansion coefficients of the α, β, and γ phases are also highly anisotropic. The average coefficient of the α phase is $54 \times 10^{-6}/°C$, or five times that of iron. This causes special compatibility problems wherever plutonium is in contact with other metals in a varying temperature environment. Figure 3 also illustrates another peculiarity of plutonium, the negative thermal expansion coefficients of the δ and δ'phases.

The crystal structures and corresponding densites of the six plutonium allotropes are listed in Fig. 3. The three crystal structures closest to room temperature are low symmetry, with α being simple monoclinic which is highly unusual for a metal. The cubic structures that are typical of most metals appear only at high temperatures where the 5f-electron bonding is overwhelmed by thermal effects. The low-symmetry structures (especially the α phase) exhibit very directional bonding. The α-phase monoclinic structure is essentially covalently bonded. Its unit cell contains 16 atoms with eight different bond lengths ranging from 2.57 to 3.71 Å [23]. Consequently, most of its physical properties are also very directional.

The low symmetry and nearly covalent nature of the bonding greatly affects the mechanical properties, which more nearly resemble those of covalently bonded minerals than most metals. The α phase is strong and brittle because the low symmetry controls the nature and motion of defects. In contrast, the fcc δ phase behaves much more like a normal metal with strength and malleability similar to that of aluminum. One must remember, however, that at δ-phase temperatures the 5f bonding is essentially gone. The δ phase is also the lowest density phase in spite of the fact that it is the only one with a traditional close-packed stacking arrangement. The higher densities in the other phases result from the directional bonding which provides for efficient packing of nonspherical atoms. In the δ phase the lack of 5f bonding (greater localization) provides more screening from core electrons and hence larger atoms. In addition, the lack of 5f character results in spherical atoms. The δ phase displays more conventional, metal-like properties than the α, β, and γ phases.

The δ phase is easily retained to room temperature by alloying. Trivalent elements such as Al, Ga, In, Ce, Sc, and Am are particularly effective. Figure 4 demonstrates the complexities of the Pu-Aℓ binary system in which almost every kind of solid-state reaction can be observed [24]. A look back at Fig. 2 reminds one that plutonium is right on the verge of localization since the addition of very little americium favors the fcc structure. Likewise, it takes only a few atomic percent aluminum to stabilize the δ-phase structure. The resulting alloys (like the pure δ phase) are much more like normal fcc metals with low strength, high ductility, smaller thermal expansion coefficients and larger ranges of temperature stability (a Pu-5 at% Aℓ alloy is stable to above 500°C). In addition, δ-phase alloys are also more compressible, more corrosion resistant and better conductors than pure α-phase plutonium.

The low melting point of pure plutonium has several other important consequences besides the obvious lack of solid-phase stability. The liquid phase must be contained during processing. This is very difficult because it is highly reactive and corrosive. It also has the greatest known surface tension and viscosity among metals (again a consequence of 5f bonding).

An important metallurgical consequence arises from the fact that most rate processes in solids depend upon homologous temperature, that is, temperature relative to the absolute melting point. Hence, diffusion and other thermally activated processes in plutonium are quite rapid at room and slightly elevated temperatures. This greatly affects the physical and mechanical properties of plutonium and its alloys. In fact, the α phase can exhibit significant ductility at room temperature at very small grain size (on the order of microns) and low strain rates so that sufficient diffusional flow occurs to accommodate plastic deformation by a grain boundary sliding mechanism.

The kinetics are very important in the transformation behavior of plutonium and its alloys. Transformations above 100°C are diffusion controlled. Only those near room temperature and below are martensitic (displacive). Transformations occur readily by changing temperatures, applying pressure, or any other kind of mechanical work. Martensitic transformations have been observed at low temperatures in Pu-Aℓ alloys [25]. These alloys tranform by an isothermal martensitic mechanism (diffusion controlled nucleation followed by a displacive) transformation at temperatures as low as that of liquid nitrogen. Hydrostatic pressure collapses the low-density crystal phases quite easily. In pure plutonium [26] the δ phase disappears at pressures less than 1 kbar. In alloys the low-density phases are also easily transformed. The solutes trapped in a martensitic transformation from the δ phase to an α-like phase are typically quite mobile. However, their role during and after the transformation is not well understood. The monoclinic α and β phases typically exhibit no equilibrium solubility for the δ-phase stabilizing elements.

It is now clear that the peculiar behavior and properties of plutonium and its alloys can be understood only by recognizing the consequences of 5f bonding. These "electronic" effects are maximal at plutonium. A proper understanding here may lead to new insight to the behavior of other elements near the diagonal (Fig. 1) which marks the crossover from bonding to localized electrons.

SUMMARY

The actinide elements, their compounds and alloys offer a most fascinating area of fundamental research. Quite recently heavy fermion materials have served to focus attention on their most important physical characteristic, the narrow energy bands. These materials have raised many new questions, and yet the extreme narrowness of their bands offers the possibility of progress on these questions. The resulting understanding will be very useful for many aspects of all actinide materials and even for the other series in the periodic table. The metallurgical behavior of plutonium and other actinides can be used as a sensitive probe to study bonding and localization. The onset of f bonding is quickly and easily recognized because the consequences are dramatic; low-symmetry crystal structures with concommitant property changes.

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FIGURE CAPTIONS

Fig 1. A nearly periodic table of the d- and f-electron elements arranged to show where the crossover to localized behavior occurs.

Fig. 2. A somewhat schematic, connected binary-alloy phase diagram of the first half of the actinide series. The solid areas are two phase regions. The hatched areas are estimates. The designations of the phases of pure plutonium are shown.

Fig. 3. A plot of percentage length change as a function of temperature illustrates the dramatic changes that occur with each plutonium phase. The behavior of iron is shown for comparison.

Fig. 4. Plutonium-aluminum phase diagram.

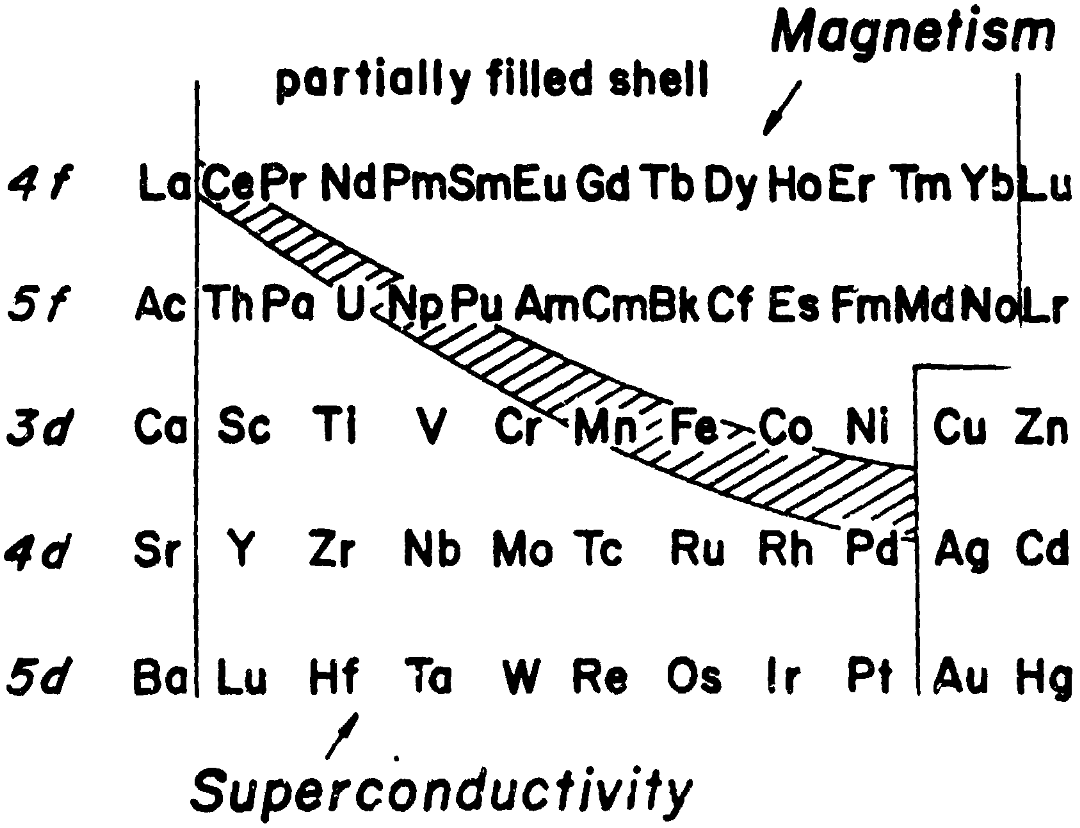
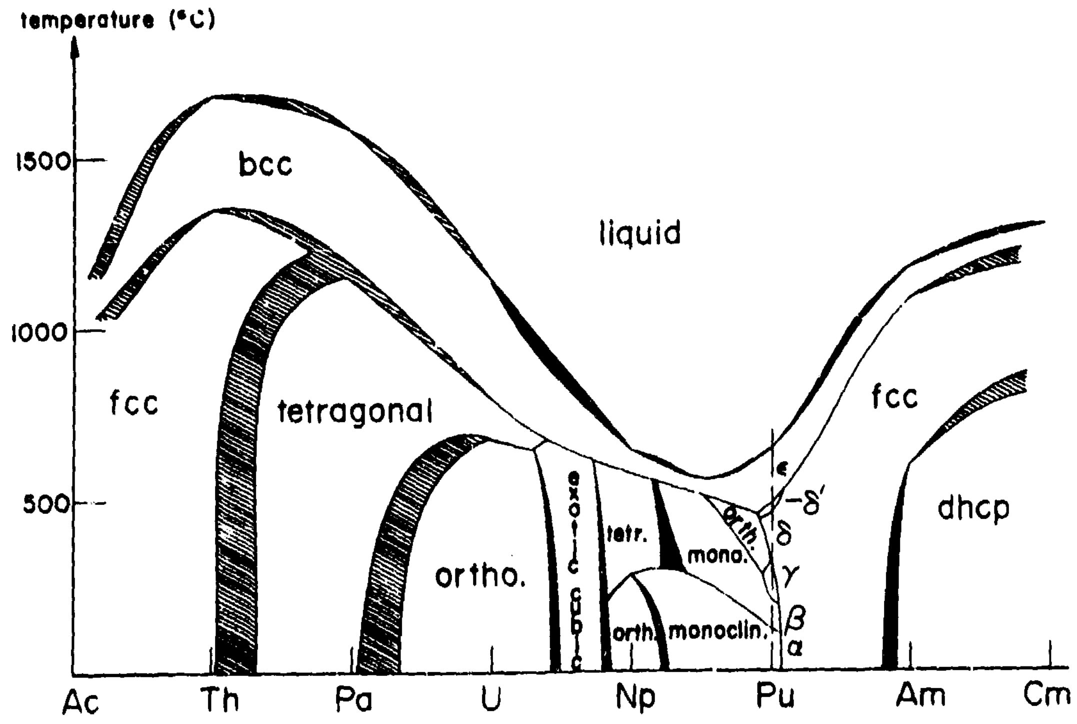

Magnetism
partially filled shell
4f La Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
5f Ac Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr
3d Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn
4d Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Cd
5d Ba Lu Hf Ta W Re Os Ir Pt Au Hg
Superconductivity

Fig. 1

Fig. 2


temperature (°C)
1500
1000
500
bcc
liquid
fcc
tetragonal
fcc
ortho.
exotic cubic
tetr.
orth.
mono.
monoclin.
orth.
ε
δ'
δ
γ
β
α
dhcp
Ac
Th
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U
Np
Pu
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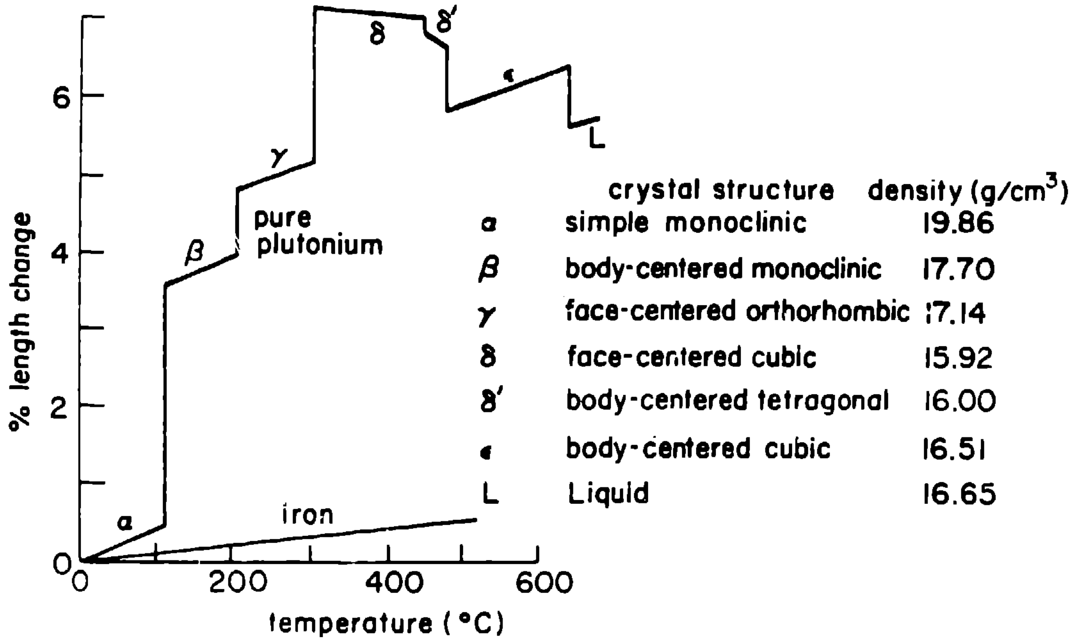
Cm



	crystal structure	density (g/cm^3)
α	simple monoclinic	19.86
β	body-centered monoclinic	17.70
γ	face-centered orthorhombic	17.14
δ	face-centered cubic	15.92
δ'	body-centered tetragonal	16.00
ε	body-centered cubic	16.51
L	Liquid	16.65

Fig. 3

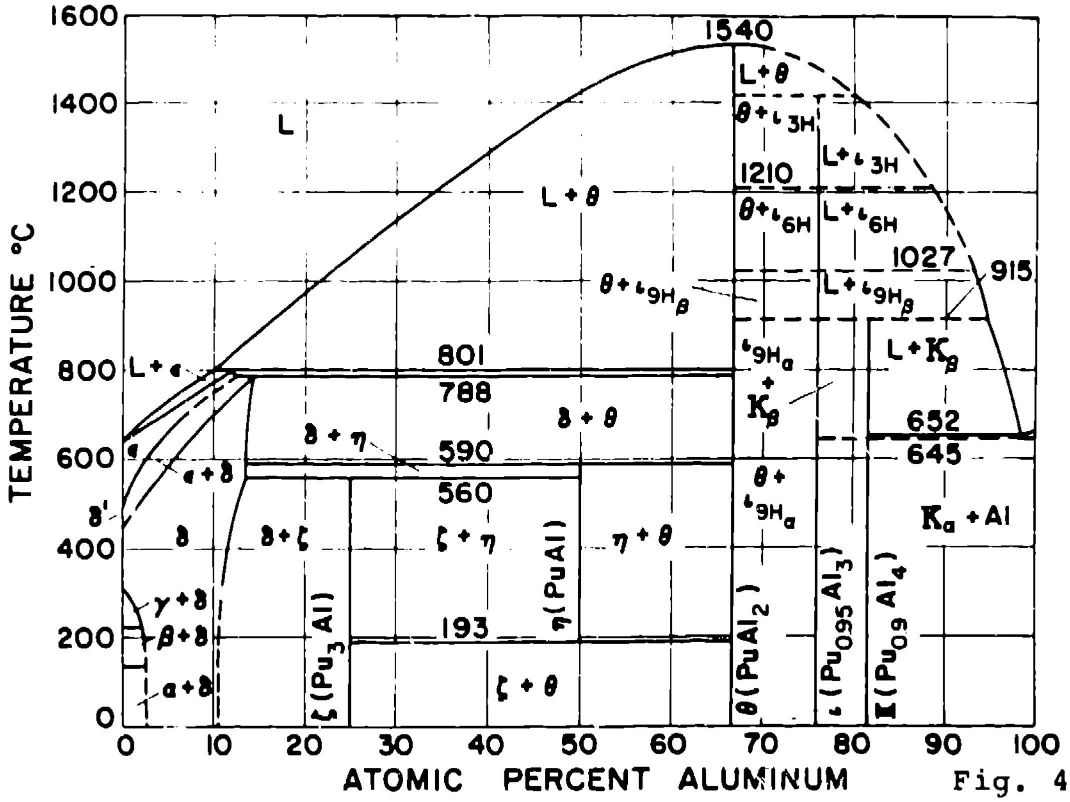


Fig. 4