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LATTICE EFFECTS IN THE LIGHT ACTINIDES

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INTRODUCTION

The light actinides show a variety of lattice effects that do not normally appear in other regions of the periodic table. Following the pioneering work of Johansson and his collaborators (Johansson et al., 1981) we know that these effects arise from interactions involving the 5f electrons. Progress in theory is bringing these effects within the range of detailed theoretical calculation: we have prepared this article to summarize the experimental situation.

The article will cover the crystal structures of the light actinides, their atomic volumes, their thermal expansion behavior, and their elastic behavior as reflected in recent thermal vibration measurements made by neutron diffraction. A discussion of the melting points will be given in terms of the thermal vibration measurements. Pressure effects will be only briefly indicated.

To indicate the range of exotic effects that are encountered in the light actinides, Fig. 1 shows the linear thermal expansion of Pu metal plotted vs. temperature. This figure shows a succession of six distinct crystallographic phases as the material is heated from room temperature to the melting point. There is a wide range of atomic volumes and thermal expansions, including a range of *negative* thermal expansion, for two of the phases, δ and δ' . The thermal expansion of Fe is shown for comparison: by this standard the thermal effects in Pu are quite large. The melting point of 613°C is anomalously low, either by rare earth or by transition metal norms.

Fig. 1. Linear thermal expansion of the six phases of Pu metal. The thermal expansion of Fe is shown for comparison.

Fig 2. Schematic phase diagram of the light actinides. Reprinted from J. Less-Common Metals 90, "Magnetism and bonding: a nearly periodic table of transition elements," p. 83, Copyright: (1983), with permission from Elsevier Science.

With so many distinct phases involved, it is necessary that some of the phases are crystallographically complex. This has been found to be the case, and the structures are in fact the most complex found for any metal. The situation is nicely presented in the schematic phase diagram of Kmetko and Smith, 1983, which is shown as Fig. 2. This diagram shows that as one proceeds from Ac through to Am, there is a progression of complex crystal structures that is correlated with a deep depression of the melting point. These features, together with other symptoms of 5f electron instability, are the subject of this article.

STRUCTURES

The light actinides are famous for their idiosyncratic structures. At normal pressures, Th has two polymorphs, U and Np each have three, while Pu has six. As one gets to Am in the periodic system there are again only two polymorphs. The structures of the various polymorphs are summarized in Table 1. The general trend with atomic number is from simple to complex to simple again. The general trend with temperature is from complex to simple. These trends are evident also in Fig. 2.

Table 1. Polymorphs of the light actinides.

Element	phase	crystal system	Stability Range (K)	space group	N _{cell}	N _{type}	ref.
Th	α	cubic	0-1673	Fm3m	4	1	1
	β	cubic	1673-2115 (M)	Im3m	2	1	1
Pa	α	tet.	ambient	I4/mmm	2	1	1
	β	cubic	high T	Fm3m	4	1	1
U	α'	CDW	0-43				2
	α	ortho.	43-935	Cmcm	4	1	3
	β	tet.	935-1045	P4 ₂ /mmm	-30	5	4
Np	γ	cubic	1045-1405 (M)	Im3m	2	1	1
	α	ortho.	0-553	Pmcn	8	2	5
	β	tet.	553-850	P4 ₂ /mmm	4	2	6
Pu	γ	cubic	850-910 (M)	Im3m	2	1	1
	α	mono.	0-395	P2 ₁ /m	16	8	7
	β	mono.	395-479	I2/m	34	7	8
	γ	ortho.	479-592	Fddd	8	1	9
	δ	cubic	592-724	Fm3m	4	1	1
	δ'	tet.	724-749	I4/mmm	2	1	10
Am	ϵ	cubic	749-913 (M)	Im3m	2	1	1
	α	hex.	ambient	P6 ₃ /mmc	4	2	1
	β	cubic	high T	Fm3m	4	1	1
Cm	α	hex.	ambient	P6 ₃ /mmc	4	2	1

References for Table 1

1 Donohue (1982)	6 Zachariasen, (1952b)
2 Lander et al. (1994)	7 Zachariasen and Ellinger, (1963b)
3 Jacob and Warren (1937)	8 Zachariasen and Ellinger, (1963a)
4 Lawson et al., (1988)	9 Zachariasen and Ellinger, (1955)
5 Zachariasen, (1952a)	10 Ellinger, (1955)

Fig 3. Crystal structures of the phases of U and Np. Blocks of 2x2x2 unit cells are shown. The atoms are numbered when there is more than one atom type. For β -U, the atom at the origin is shaded.

Fig 4. Crystal structures of the phases of Pu. Blocks of 2x2x2 unit cells are shown. The atoms are numbered when there is more than one atom type. For β -Pu, the atom at the origin is shaded.

The main point of interest is the complexity of the low temperature of U, Np and Pu. The α -form of Pu metal is the most complex known for any metallic element: it is monoclinic, with 16 atoms per primitive cell, and there are 8 unique atom types. Zachariasen's solution of this structure from powder data is one of the outstanding achievements in all of crystallography. Figs. 3 and 4 illustrate this structural complexity. The figures, which are drawn to scale, show blocks of eight unit cells ($2 \times 2 \times 2$) in order to convey some idea of the packing.

Fig. 5 shows the atomic volumes of the light actinides plotted vs. atomic number. These volumes are obtained from crystallographic data, and there is a point shown for each polymorph. The points corresponding to the polymorph of lowest volume (always the low temperature structure) are connected to illustrate the trend. The wide range of values for Pu is taken by Johansson et al. (1981) as evidence for a metal-insulator transition among the 5f electrons.

Fig. 5. Atomic volumes for the various light actinide phases.

THERMAL EXPANSION

Thermal expansion can also be determined from crystallographic data. Fig. 6 shows the atomic volume plotted vs. temperature for the light actinides. To a good approximation, the volume vs. temperature behavior has a quadratic dependence, and the coefficient of thermal expansion can be obtained by numerical differentiation. A factor of 1/3 has been applied to obtain a coefficient of linear expansion, and the results are plotted in Fig. 7. Schonfeld and Tate (1996) give further information on a range of thermal expansion measurements.

Fig 6. Atomic volume vs. temperature for some of the light actinide phases. Data are from Lawson et al. (1994a,b) and Lawson et al. (1996).

Fig. 7. Coefficient of linear thermal expansion for some of the light actinide phases.

The temperature dependence of the sequence $\delta \rightarrow \delta' \rightarrow \epsilon$ is shown in detail in Fig. 8. The sequence is really quite unusual, as it connects two phases of cubic symmetry: the BCC ϵ -phase and the FCC δ -phase. The intermediate tetragonal δ' -phase can be construed as a distortion either of the ϵ -phase or of the δ -phase, and its space group, $I4/mmm$ is a subgroup of both $Im\bar{3}m$ (ϵ) and $Fm\bar{3}m$ (δ). In Fig. 8 we have applied appropriate geometric factors to the lattice constants in order to clarify the metric relationships between the phases.

The approximate continuity of the normalized lattice constants of Fig. 8 suggests that the $\delta \rightarrow \delta'$ transition could be second order, while the $\delta' \rightarrow \epsilon$ transition is first order. But the situation is obscured by impurity effects: in actual experiments, the δ - and δ' -phases are separated by a two-phase region where δ and ϵ -phases are observed simultaneously. The behavior of this region – indeed whether δ' is observed at all – is extraordinarily sensitive to impurity content: 200 ppm of metallic impurities is known to place a significant role.

Ordinarily, the δ -phase of Pu is stable only over a narrow temperature range 592–724 K. The stability range can be considerably extended by the addition of certain solutes, including Al, Ga, Zn and Ce. The lattice constants of several Pu-Ga alloys plotted vs. temperature are shown in Fig. 9. This figure shows that the addition of only small amounts of solute gradually changes the high temperature thermal expansion from negative to positive. At the same time, the lattice constant is reduced. Similar x-ray data for a wide variety of solutes is available from Lee et al. (1960) and Elliott et al. (1960). Fig. 10 shows the lattice constants measured at 573 K for a variety of Pu alloys, and the effect of all solutes – except Ce – is to reduce the lattice constant. The effect of these solutes on the high-temperature thermal expansion (573–673 K) is to increase the thermal expansion coefficients from negative to positive, as shown in Fig. 11. Again, Ce is anomalous; otherwise the effect is roughly independent of the solute.

Fig. 8. Normalized lattice constants vs. temperature for δ -, δ' - and ϵ -phases of Pu metal. After Ellinger (1956).

Fig. 9. Lattice constant vs. temperature for some Pu-Ga alloys.

Fig. 10. Lattice constant at 573 K vs. concentration for some Pu-X alloys. "Al-2" data are from Lee et al. (1960), and the other data are from Elliott et al. (1960).

Fig. 11. Coefficient of linear thermal expansion over the temperature range 573-673 K. "Al-2" data are from Lee et al. (1960), and the other data are from Elliott et al. (1960).

ELASTIC PROPERTIES

Neutron diffraction offers an interesting method for determining the elastic properties of materials via the Debye-Waller factors. The basic idea is that diffraction measures the mean-square vibrational amplitude of atomic vibration, $\langle u^2 \rangle$, and the atomic spring constants can be inferred from these quantities. The effect of increasing vibrational amplitude on the appearance of the diffraction pattern is quite strong: this is shown in Fig. 12. Precise numerical values for $\langle u^2 \rangle$ can be extracted from the diffraction patterns by Rietveld refinement.

$\langle u^2 \rangle$ is a strong function of temperature. For an ideal Debye solid, $\langle u^2 \rangle$ would be a linear function of temperature at high enough temperatures, and the slope of the $\langle u^2 \rangle$ vs. temperature curve would be the inverse of the atomic spring constant, κ_{atom} according to

$$\kappa_{\text{atom}} \langle u^2 \rangle = k_B T \quad (1)$$

where k_B is the Boltzmann constant. κ_{atom} is related to the Debye-Waller temperature by the formula

$$\kappa_{\text{atom}} = \frac{mk_B^2 \Theta_{DW}^2}{3\hbar^2} \quad (2)$$

Fig. 12. Neutron diffraction patterns for $\text{Pu}_{0.95}\text{Al}_{0.05}$ at 13 and 260 K. The data have been fit by Rietveld analysis. '+'s are the data, the line is the Rietveld fit, and the lower curve is the residual.

Fig. 13. $\langle u^2 \rangle$ vs. temperature for $\text{Pu}_{0.94}\text{Ga}_{0.06}$ and for Th. The data have been fit with $\Theta_{DW} = \Theta_{DW,0} + cT$.

This is just a definition of the Debye-Waller temperature in terms of atomic spring constants. For any temperature,

$$\langle u^2 \rangle = \frac{3\hbar^2 T}{mk_B \Theta_{DW}^2} \left[\frac{\Theta_{DW}}{4T} + \frac{T}{\Theta_{DW}} \int_0^{\frac{\Theta_{DW}}{T}} \frac{u du}{e^u - 1} \right] \quad (3)$$

Fig. 13 shows the measured values of $\langle u^2 \rangle$ plotted vs. temperature for thorium metal and for δ -stabilized $\text{Pu}_{0.94}\text{Ga}_{0.06}$, and the behavior of these two isostructural materials is quite different. The difference has two causes: first, Th and δ -phase Pu have different Debye-Waller temperatures, and second, the Θ_{DW} 's have different temperature dependencies. Plotting the data in dimensionless form can separate these effects, and this is done in Fig. 14. In this figure the dimensionless displacement, $mk_B \Theta_{DW} \langle u^2 \rangle / \hbar^2$ is plotted versus the dimensionless temperature, T/Θ_{DW} for several δ -phase Pu alloys. The ideal Debye behavior (Eq. 2) is also shown in this graph. On this graph it is easier to see that the curvature of the $\langle u^2 \rangle$ vs. T plot is much greater for Pu than for Th, and that the deviation for Pu from the ideal behavior is quite significant. The data have been fitted by assuring the validity of Eq. 3, but with the additional model assumption that Θ_{DW} is linear in temperature:

$$\Theta_{DW} = \Theta_{DW,0} + cT. \quad (4)$$

Fig. 14. Dimensionless plot of $\langle u^2 \rangle$ vs. temperature for some Pu-Ga alloys and for Th. The ideal Debye behavior is also shown. The upper curve shows the linear thermal expansion data, $\Delta a/a$ vs $T/\Theta_{Dw,0}$.

Fig. 15. Θ_{DW} vs. temperature for some of the light actinides and for Pb. The points with error bars show the limits of our measurements. The solid line shows the limit of the solid phase.

Fig. 16. Anharmonicity parameter, c , for the light actinides and for Pb. The anharmonicity has two parts, Grüneisen and intrinsic, as explained in the text.

In this equation, Θ_{DW} is the temperature dependent Debye-Waller temperature, $\Theta_{DW,0}$ is the fitted Debye-Waller temperature at $T=0$, and c is a fitted parameter we may call the anharmonicity parameter.

In the quasi-harmonic model of lattice dynamics, we assume that Θ_{DW} changes according to the Grüneisen equation of state

$$\frac{\Theta}{\Theta_0} = \left(\frac{\Omega}{\Omega_0} \right)^\gamma \quad (5)$$

In this equation, Ω is the atomic volume and γ is a materials constant, the Grüneisen parameter. This would imply that the fitted constant c should be proportional to the thermal expansion. The data of Fig 14 are in direct contradiction to this conclusion, as the variation of thermal expansion with Ga concentration is quite large, while c hardly changes at all. In the case of Pu-Ga alloys, we conclude that Θ_{DW} is *explicitly* (or *intrinsically*) dependent on temperature (Grimvall, 1986).

We have completed neutron diffraction experiments on many of the light actinide phases. The temperature dependence of the Θ_{DW} 's is shown in Fig. 15. The linear behavior is a consequence of the model assumption of Eq. 4. $\Theta_{DW,0}$ is the intercept and c the slope of the straight line plots of Fig. 15. Error bars of the fits are shown for the high and low temperature limits of the experimental data. The extrapolation of Θ_{DW} to the melting point is shown with a full line, and beyond the melting point with a dashed line. The size of these error bars depends strongly on the temperature range over which measurements can be performed, as Θ_{DW} and c are strongly correlated. The temperature range is of course limited by phase stability, and this is a serious limitation for the actinides. For example, we have not so far been able to say very much about the temperature dependence of Θ_{DW} in ϵ -phase Pu.

In the case of stabilized δ -phase Pu, we can compare our results to the ultrasonic measurements of Taylor et al. (1967), and we find very good agreement. Their data also support the model assumption of Eq. 4; in particular, it seems that it is Θ_{DW} , rather than K_{atom} , that has a linear dependence on temperature. The linear variation of Θ_{DW} with temperature seems to be well established for δ -phase Pu because of its large stability range; this is less well established experimentally for some of the other phases.

We have plotted the measured anharmonicities, c , for the light actinides in Fig. 16. We have estimated the relative contributions of Grüneisen and intrinsic anharmonicity by calculating the expected change in Θ_{DW} from thermal expansion data as assuming reasonable values for γ . If reliable experimental data are not available, we have assumed $\gamma=2$. The intrinsic contributions to the anharmonicity are very large for U and Np and somewhat smaller for Pu. For comparison, data for Pb are also shown on this graph.

Diffuse Scattering

A curious feature of the neutron diffraction patterns of δ -phase Pu is the diffuse background scattering that is barely perceptible in Fig 12. This scattering becomes quite pronounced at high temperature, as shown in Fig 17, which shows a diffraction pattern for *unalloyed* δ -phase Pu metal at 440°C. The observation of this scattering in unalloyed material, together with the determination that the scattering is proportional to the temperature suggests that the lattice vibrations alone cause the scattering. The inset to this

feature shows the calculated thermal diffuse scattering (TDS) using a formula devised by Warren and modified by Borie (Warren, 1969). The scattering can be approximated by Rietveld refinement using a special background function of the form

$$B_{TDS}(k) = \sum_j C_j \frac{d^2}{dR^2} \left(\frac{\sin(kR_j)}{kR_j} \right) e^{-2W_j} \quad (6)$$

Fig. 17. Diffuse scattering observed in unalloyed δ -phase Pu at 440°C. In the upper curve, the diffuse scattering has not been fit. In the lower curve, the diffuse scattering has been fit as explained in the text. The inset shows the calculated Warren-Borie scattering for appropriate parameters.

In this expression, k is the scattering vector, R_j is an interatomic distance for the FCC structure, C_j is a parameter fitted to the diffraction data, and $\exp(-2W_j)$ is a Debye-Waller factor. The function $\sin(kR)/(kR)$ is just the Debye scattering function that describes the diffraction patterns of liquids and amorphous materials. In practice, a successful fit can be made with a small number of interatomic distances (seven for Fig. 17). A comparison of the fitted values of C shows that the observed scattering can be accounted for by the elastic continuum described by Debye's theory of lattice vibrations.

This type of scattering is not often observed with neutrons, because the velocities of the scattered neutrons are much less than the sound speeds in most materials. Then the static approximation, on which the Warren-Borie formula depends, is invalid. Pu is an exception, as its low Debye temperature means that the static approximation is valid and the diffuse scattering can be observed and interpreted. The diffuse scattering is unusual but has no special significance.

Elastic Anisotropy

Ledbetter and Moment (1976) measured single crystal elastic constants for δ -phase Pu. Their results show that δ -phase Pu is the most elastically anisotropic of all the face centered cubic metals. The anisotropy is characterized by the so-called Zener anisotropy defined by

$$Z = \frac{2c_{44}}{c_{11} - c_{12}} \quad (7)$$

For $Z = 1$, the surface representing the Young's modulus is spherical. The available data for the FCC metals are shown in Fig. 18. Among the FCC metals, δ -phase Pu is much more anisotropic than its nearest competitor, γ -Ce.

MELTING POINTS

As shown in Fig. 2, there is a dramatic depression of the melting point near Pu. This fact has long been regarded as one of the essential features of Pu physics and metallurgy and has received extensive discussion in the literature, for example, by Matthias et al. (1967) and by Kmetko and Hill (1976). It turns out that the melting behavior of the light actinides is explained rather naturally by the Lindemann rule if the temperature dependence of the Θ_{pw} shown in Fig. 14 is taken into account.

The Lindemann rule (Lindemann, 1910) was originally put forward to provide a means of estimating the Einstein temperature of a solid from its measured melting point. Lindemann's idea was that a solid should melt when the vibrational amplitude of the atoms is large enough that they bump into each other. Gilvarry (1958) reinterpreted the Lindemann rule to say that a solid melts when the vibrational amplitude is a fixed fraction of the interatomic distance, the Lindemann constant of about 10%. The Debye temperature determines the vibrational amplitude, so we can use our measured values of

Fig. 18. Zener anisotropy ratio for various FCC metals. Data are from Hearmon (1979, 1984) and from Ledbetter and Moment (1976).

Fig. 19. Measured and calculated melting points for Pb and the light actinides. The calculations are made with the Lindemann rule, both including and neglecting the temperature dependence of Θ_{DW} .

Θ_{DW} to predict the melting point. However, it is necessary to use the Θ_{DW} evaluated at the melting point, or the vibrations will be underestimated.

If the Lindemann rule is applied without correction for the temperature dependence of Θ_{DW} , the estimated melting points are in serious disagreement with experiment, as shown in Fig. 19. If Θ_{DW} is evaluated at the melting point, then the agreement is much better. This is also shown in Fig. 19, using a Lindemann constant of 8.3%. With a look back at Fig. 2, one can say that depression of the melting points observed in the actinide series is not really anomalous, given the behavior of Θ_{DW} .

The elastic properties of the actinides vs. position in the periodic system are shown in Fig. 20. The trend in $\Theta_{DW,0}$ is actually *opposite* to that of the melting point. However, it is more than compensated by the trend in the anharmonicity, and this gives the observed depression of the melting points with the minimum at Pu.

PRESSURE EFFECTS

We can only suggest the range of pressure effects in the light actinides by showing the pressure-volume-temperature diagram determined by Morgan (Fig. 21). Clearly this is a fertile area for exploration, but the experimental difficulties are considerable.

Fig. 20. Melting points for the actinides are rare earths and measured values for Θ_{DW} and anharmonicity, c .

Fig. 21. Pressure-volume-temperature diagram for plutonium metal from Morgan (1970). The ζ -phase appears only at high pressure and its structure is not known. Reprinted with permission of the AIME.

SUMMARY AND DISCUSSION

The physical properties of the light actinides are dominated by 5f-electron interactions. These interactions are manifest in idiosyncratic structures, a singular dependence of volume on atomic number and on anharmonic behavior that dominates the thermal expansion and elastic properties. The latter effects are large, and it is necessary to take the effect of temperature into account explicitly, as the materials properties are highly temperature dependent.

Theoretical progress has been quite rapid during the past decade. There is now a good explanation of the structural trends, given in a series of papers by Wills and Eriksson (1992), Eriksson et al. (1993), and Söderlind et al. (1995, 1997, 1998). The general idea is that structural complexity is to be expected when narrow, delocalized f-bands cross the fermi level as is believed to occur in the light actinides. These theories are not yet predictive of structural details, and they do not yet address the tendency for Pu to form "self-intermetallic compounds" in those cases where there is more than one crystallographically unique Pu atom. (Lawson et al, 1996).

There have been some attempts to calculate the thermal expansion behavior of the light actinides by Söderlind et al. (1990) and Söderlind and Johansson (1993). It is difficult to compare with experiment, because the experimental results are dominated by the temperature dependence of the expansion behavior (Fig. 7). In the same way, calculations of elastic constants (Söderlind et al., 1993) will also have to include the temperature dependence before a meaningful comparison with experiment (Fig. 15) can be made.

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