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Incommensurate/Commensurate Charge-Density-Wave States as a Source for Plutonium Metal Behavior

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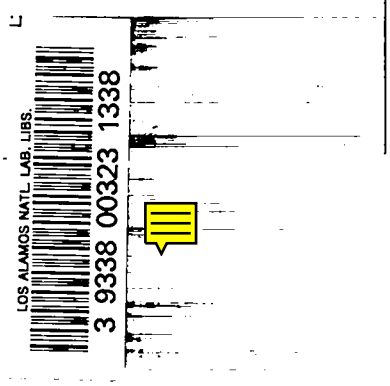

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INCOMMENSURATE/COMMENSURATE CHARGE-DENSITY-WAVE STATES AS A SOURCE FOR PLUTONIUM METAL BEHAVIOR

by

Thomas A. Sandenaw

ABSTRACT

Evidence is reviewed that suggests the possibility of an incommensurate structure in several of the phases of plutonium (Pu) metal. Its phases show the general behavior of investigated incommensurate crystals, i.e., a sample dependence, a time dependence, a dependence on thermal history, a structure memory, and a negative thermal expansion along some axes. All of these effects are empirically found in incommensurate materials.

The space group structure of the alpha phase of Pu ($P2_1/m$) is the same as that of proven charge-density-wave (CDW) systems that have a quasi-one-dimensional structure, i.e., where there is a one-dimensional modulation of the crystal. Chiral symmetry has been shown to be inherent to the incommensurability of such a space group structure, i.e., one that has a twofold screw axis with a center of symmetry. The hysteresis in resistivity vs temperature curve of the alpha phase ($P2_1/m$), as well as Hall coefficient response, suggests that this phase has regions of incommensurate and commensurate CDW states. The reported effect of current density and current direction on Hall voltage is added evidence for these states.

The incommensurate phases of proven incommensurate crystals have chiral symmetry in at least one dimension; i.e., there is a twofold screw axis with a center of symmetry. Space group structures of commensurate phases indicate they are nonchiral. The reported space group structures for all Pu phases, except the gamma, indicate the presence of a screw axis with a center of

symmetry in at least one dimension. Thus all phases, except gamma, have the potential of being incommensurate CDW phases. In incommensurate crystals (not in commensurate ones) the pitch of the screw axes can be varied in a continuous way, permitting the crystal to store energy without linear expansion and thus enabling the material to have zero (or negative) thermal expansion.

The reported internal friction and relative shear modulus results can be analyzed to show an apparent confirmation for incommensurate and/or commensurate CDW states in all Pu phases. Besides this, four of the designated space group structures for Pu phases are listed in a tabulation of superspace groups in (3 + 1) dimensions describing incommensurate structures with a one-dimensional modulation. It is suggested that the CDW modulation of Pu phases may be contributing to f-bonding.

I. INTRODUCTION

The crystal structure for alpha-phase plutonium (α-Pu) was determined to be simple monoclinic[1] and was assigned the space group designation $C_{2h}P2_1/m$. The atoms of the monoclinic cell of the α-Pu structure are arranged in layers at y = 1/4 and y = 3/4 where they form a network of irregular quadrilaterals and/or triangles.[2] Pearson[2] has already classified the distorted-hexagonal α-Pu structure as that of a metallic alloy.

Recent publications indicate that the space group structure for the martensitic phase of the intermetallic compound TiNi is $P2_1/m$[3a,b] or the same as that of α-Pu. The monoclinic structure of TiNi martensite has also been described as hexagonal close-packed.[4] The significance of the $P2_1/m$ space group designation is in a twofold screw axis with a center of symmetry and a mirror plane. The twofold screw axis in this space group structure (No. 11) is single-dimensional.

The transitional-metal trichalcogenides TaS_3[5] and $NbSe_3$[6] have also been shown to have this same space group structure. A general feature of

these materials is a linear stacking of trigonal prisms of chalcogen atoms parallel to the chain axis. Both of these materials undergo charge-density-wave (CDW) transitions. The transitions have been attributed to their quasi-one-dimensional structure.

The fact that the space group structure, $P2_1/m$, is observed with as many different materials suggests not only that it is important for CDW states and solitary excitations (solitons), but also that it may be important in martensitic transformations.

The hysteresis in the resistivity vs temperature curve for α-Pu,[7] as well as the Hall coefficient response,[8] suggests that this Pu phase has regions of incommensurate and commensurate CDW states when comparisons are made with the physical property behavior of known CDW materials.

In a recent paper, Zhao-bin Su and B. Sakita[9] have shown that chiral symmetry and chiral anomaly are inherent in the incommensurability of a quasi-one-dimensional CDW system. Chiral symmetry is present in a screw axis that has a center of symmetry. The incommensurate and commensurate phases of almost every incommensurate crystal have space group structures indicating the presence of screw axes.

Screw diads, screw triads, and even screw tetrads are implied by the space group designations for higher-temperature phases of Pu. In all but one case, the reported space group structures for Pu metal phases indicate the presence of a center of symmetry in the screw axes in at least one dimension. The exception is the gamma (γ) phase. The reported space group structure for the γ-phase indicates the presence of three-dimensional, twofold screw axes but without chiral symmetry.

Scott[10] has suggested that a screw axis may act like a spring when there is chiral symmetry in an incommensurate crystal. Energy may be stored by either compressing the spring or tightening the pitch. Such a compression could account for a lack of thermal expansion along one axis of the incommensurate crystal $BaMnF_4$.[10] The local rotations and internal torques resulting from chiral symmetry of an incommensurate crystal could permit nonequilibrium states.

Thermal contraction along one or more axes is also seen in phases of Pu. Transformations between the many phases of Pu metal have been considered to be martensitic under some conditions.[11] Pu metal shows the

general behavior of investigated incommensurate crystals. The incommensurates show thermal hysteresis (a time dependence), a sample dependence, a dependence on thermal history of the sample, and a structure memory. Structure memory has also been noted between phases of Pu metal, as is the case with some incommensurate crystals. Physical property evidence suggests there may be another incommensurate/commensurate CDW phase region below ∿60 K, as has been suggested previously. Such a phase could be called alpha I (αI).

Explanations for the physical property behavior of α-Pu and higher-temperature Pu phases and the possible tie-in between physical property behavior and space group structure will be presented in greater detail in the following sections.

II. CDW THEORY FOR LAYERED MATERIALS

A CDW has been described as a coupled distortion of the conduction electron density and the underlying lattice.[12] The lattice distortion is considered to be necessary because without it the Coulomb energy of the CDW would be too high. A CDW transition is properly a displacive transition. Such a transition can be martensitic. Transitions to CDW states are seen in many layered transition-metal dichalcogenides. One material in which the different CDW states have been studied is 2H-$TaSe_2$.

Walker and Jacobs[13] have developed a theory of CDW states in a metal dichalcogenide like 2H-$TaSe_2$ through free-energy and symmetry considerations. In investigating the different commensurate states allowed by their theory, they found that there were three distinct commensurate states called Types I, II, and III. They showed that the threefold rotation axes for the three phases (from Type I, II, and III) are in different positions. They found that there are four distinct states that can be formed by stacking Type-I layers. Three are hexagonal ($P6_3/mmc$), and the fourth distinct state is monoclinic ($P2_1/m$) and has six members. Their results show three distinct structures for Type-II states. These are all orthorhombic (Cmcm). There are also three distinct orthorhombic structures, all Cmcm, for Type III states. It should be noted that the space group structures for the three distinct commensurate states (Types I,

II, and III) described by Walker and Jacobs[13] all have twofold screw axes with a center of symmetry in at least one dimension. They thus have chiral symmetry and should show properties of incommensurate CDW (ICDW) phases. There is only a one-dimensional, twofold screw axis in the monoclinic $P2_1/m$ structure. The twofold screw axes in the Cmcm structure are three-dimensional. Two are chiral. There are multiple twofold screw axes in the hexagonal, $P6_3/mmc$ structure. Of the three, only this latter structure shows the presence of screw hexads.

Many of the materials with the $P2_1/m$ space group structure, such as $TaSe_3$, are considered to be one-dimensional conducting systems.[14, 15] Their crystal structure comprises infinitely long chains of distorted prismatic-type columns extending along the monoclinic b axis. In the monoclinic unit cell of $NbSe_3$ there are three inequivalent pairs of trigonal columns. This is supposedly because of the screw axis.[15]

The CDW transitions in TaS_3 and $NbSe_3$ have been attributed to Peierls transitions.[14, 15] Resistivity anomalies found in $NbSe_3$ have also been associated with the openings of gaps at the Fermi surface as the result of CDW formation.[6] In the model of Zhao-bin Su and B. Sakita,[9] the chiral symmetry of an ICDW state is broken to produce the energy gap in the Fermi surface, i.e., a Peierls transition.

The incommensurate to commensurate CDW transition in 2H-$TaSe_2$ (on cooling) is from the hexagonal $P6_3/mmc$ space group structure to the orthorhombic Cmcm space group structure.[16] The $P6_3/mmc$ (No. 194) structure has chiral symmetry in more than one dimension and should be capable of existing as an ICDW state. The space group structure of Cmcm (No. 63) indicates there can be chiral symmetry in two dimensions and nonchiral symmetry in the third. There must be symmetry breaking in the Cmcm structure to give a fully commensurate state, i.e., nonchiral symmetry in three dimensions. This appears to be the case as noted by Simpson et al.[17a] and as discussed by Jacobs and Walker.[17b] Below T_d there were three commensurate CDW (CCDW) vectors. On warming above this temperature there was a hysteresis and only two of the three CDW vectors became incommensurate; the third stayed commensurate. This condition of a mixed incommensurate and commensurate CDW state has been designated as a striped

phase. In this striped phase, 2H-$TaSe_2$ is behaving as it should for its designated Cmcm space group structure.

III. INCOMMENSURATE AND COMMENSURATE CDW STATES IN α-Pu METAL

A. Similarity Between Physical Property Behavior of α-Pu and that of Proven CDW Chalcogenide Materials

The theory of Walker and Jacobs[13] suggests that there should be a CCDW state detectable in the α-Pu because of its space group designation. The electrical resistivity vs temperature curve for α-Pu shows a negative temperature coefficient of resistance from the β→α phase transformation (∿395 K) down to ∿100 K.[18] There is a maximum in the resistivity vs temperature curve of α-Pu at ∿100 K.

The resistivity vs temperature curve of orthorhombic TaS_3 (space group $C222_1$)[5] also shows such a negative temperature coefficient of resistance over a large temperature range.[19] In addition, thermal hysteresis in the resistivity vs temperature curve exists over the temperature range 55 K<T<205 K. The CDW is considered to be incommensurate in this temperature range and then commensurate, at least in the chain direction, below 55 K. The space designation of D_2C222_1 (No. 20) for O-TaS_3 denotes the presence of twofold screw axes in three dimensions but without chiral symmetry in any of the three dimensions. Twofold rotation axes are shown for two dimensions. There must be symmetry breaking in order for any structure designated as $C222_1$ to become incommensurate, i.e., have chiral symmetry. The Peierls transition at ∿55 K must cause O-$TaSe_3$ to revert to the fully nonchiral, commensurate state.

A hysteresis loop has been shown to occur in the resistivity vs temperature curve for α-Pu between ∿160 K and 260-290 K,[7] as mentioned above. It is very much like the hysteresis loop seen in the resistivity vs temperature curve of O-TaS_3 in its supposedly ICDW temperature region. A similar hysteresis loop was seen by Hwang et al.[20a] in the resistivity vs temperature curve of $Ti_{50}Ni_{47}Fe_3$ alloy. Their study[20b] indicated that there was an ICDW structure in part of the temperature region of this hysteresis loop. The resistivity vs temperature loop was higher on the warming cycle, as was the case with α-Pu.[7] This smaller loop seen with

$Ti_{50}Ni_{47}Fe_3$[20a] was completely independent of the much larger loop seen on cooling from the martensitic start temperature (M_s) to the M_f temperature and on warming through the A_s and A_f temperatures.

The resistivity data for α-Pu thus suggest there is an ICDW phase region between ∿290 K and 160 K with the possibility of a CCDW phase region below ∿160 K. A similarity in the Hall coefficient response of 2H-TaSe[21] and α-Pu[8] suggests that α-Pu may even have the CCDW structure down to ∿30 K, although the lower limit is probably 100 K. There is a change in sign of Hall effect of α-Pu at ∿160 K[8] with a minimum at ∿30 K. There is a change of sign at ∿90 K in the case of 2H-$TaSe_2$[21] with a minimum that occurs at 35 K. The 2H-$TaSe_2$ structure is commensurate below 90 K and incommensurate for a short range above this temperature.

C. E. Olson* (private communication) has suggested that the best evidence for CDW states in α-Pu comes from comments of Loree and Pinnick[8] regarding the effects of current density and current direction on Hall voltage. They found that the Hall voltage became nonlinear above a critical current density (j), with an added component of positive sign that increased roughly as the square of the excess current density. The critical j increased with sample thickness and decreased with increasing temperature. The Hall voltage magnitude was only sensitive to current direction below ∿25 K. The behavior noted by Loree and Pinnick[8] is also observed with proven CDW systems.

The ICDW phase of α-Pu should be due to chiral symmetry in one dimension based on the space group designation. The maximum in resistivity vs temperature at 100 K suggests an M_s temperature as is the case with TiNi.[22] A distortion of the lattice parameters below ∿65 K (Pu) as reported by Lee et al.[23] could result in a slightly different structure below a supposedly M_f temperature. The anomaly is the close similarity between the Hall coefficient vs temperature behavior of 2H-$TaSe_2$[21] and α-Pu,[8] which suggests that α-Pu should have three-dimensional CCDW states at least between ∿30 K and 160 K. Any monoclinic space group designation eliminates the possibility of a two- or three-dimensional twofold screw axes.

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An explanation for a similarity in the Hall coefficient behavior of α-Pu and 2H-$TaSe_2$ as a function of temperature is that there is symmetry breaking (a Peierls transition) at 160 K. The Pu structure maintains its one-dimensional, twofold screw axis, but the transition is from chiral symmetry to nonchiral symmetry with a resulting commensurate state. This CCDW state should exist to a postulated M_s temperature of ∿100 K.

There appears to be a precedent for symmetry breaking in a material with the $P2_1/m$ space group structure. The likely structure of the one-dimensional compound TaS_3 was given as monoclinic $P2_1/m$ by Meerschaut et al.[5] This material exhibits a transition at 210 K to a commensurate state that can be explained by a Peierls transition, i.e., symmetry breaking.

B. Comparison of α-Pu and TiNi Space Group Structures

Hwang et al.[20a] have discussed the 'premartensitic' or premonitory phenomena in $Ti_{50}Ni_{47}Fe_3$ just above M_s. They note the difficulty in determining crystal structure and suggest that it might be due to proximity and overlap of premartensitic and martensitic transformation, which makes it difficult to interpret experimental data. They do conclude that the premartensitic effects are not precursory effects but separate electronically driven structural changes.

The intermediate phase of $Ti_{50}Ni_{47}Fe_3$ was recently confirmed by Goo and Sinclair[24] to be rhombohedral $P\bar{3}1m$ (No. 162). This space group does not have chiral symmetry and could give a CCDW state.

A recent publication by Tietze et al.[25] gives the intermediate phase of TiNi as a distorted CsCl structure with monoclinic symmetry (P2/m, No. 10). This monoclinic space group structure has no twofold screw axis. It does not have any chiral symmetry. It is notable that the space group structure reported for α-Pu ($P2_1/m$) and for β-Pu (I2/m, C2/m) are maximal non-isomorphic subgroups of the P2/m (No. 10)[26] structure reported by Tietze et al.[25] for the intermediate phase of TiNi. Low-temperature phases of both TiNi and α-Pu are $P2_1/m$.

It is also notable that the CsCl-type structure given for the TiNi austenite $Pm\bar{3}m$ (No. 221) is a maximal non-isomorphic subgroup of the δ-Pu space group structure, $Fm\bar{3}m$ (No. 225). These two space group structures are also seen with Cu_3Au. $Fm\bar{3}m$ (No. 225) is the disordered phase above 658

K, and $Pm\bar{3}m$ (No. 221) is the ordered phase below 658 K. Sandenaw and Harbur[27] have pointed out that Pu metal and its alloys appear to show the low-temperature physical properties of an antiphase structure, such as is seen in the case of Cu_3Au. This poses a question as to whether Cu_3Au physical properties are due to ICDW (and CCDW) states.

C. Case for an Incommensurate Pu Martensite Below 60 K (αI)

One would suspect that the martensitic phase of Pu metal (below 60 K) should have a Cmcm space group structure. This is based on the similarity in Hall coefficient behavior of commensurate 2H-$TaSe_2$[21] and α-Pu[8] in its phase range from 160 K to $\sim$30 K. Both the $P2_1/m$ (α-Pu) and Cmcm (2H-$TaSe_2$) space group structures should show CCDW states according to Walker and Jacobs' theory.[13] Two of the many maximal non-isomorphic subgroups of the Cmcm structure are $P2_1/m$ and C2/m.[26] These are the α-Pu and β-Pu crystal structures. Also, the Cmcm structure (α-U) is the high-pressure phase of both Ce and Am.

There is the possibility of another space group structure in Pu metal below $\sim$60 K. Evidence favors Pmnm (No. 59). This is the structure of α'-U for the temperature range below 43 K.[28] The Pmnm designation indicates two-dimensional, twofold screw axes with a center of symmetry. This structure is thus chiral in two dimensions. The thermal-expansion results and lattice-parameter results for α'-U[29] and α-Pu[23] below $\sim$60 K suggest that the structure of each metal has two-dimensional, chiral symmetry. The data of Tindall and Steinitz[29] clearly show this for α'-U. There is contraction along two dimensions. The results of Lee et al.[23] show a possible negative thermal expansion in two lattice parameters of αI-Pu, whereas the third is nearly normal.

The similarity in physical property response of U and Pu metals below 50 K has been previously noted[30] in a paper suggesting that a valence-fluctuation model could be applicable to actinide materials. Another similarity in the physical property behavior of α'-U and α-Pu is seen in low-temperature thermal-conductivity measurements. Figures shown by Hall and Lee[31] indicate that every experimentalist gets a different thermal conductivity curve below $\sim$300 K, a variation that must depend upon specimen processing. The greatest difference in results appeared to be below $\sim$60 K. Because of these similarities, Marmeggi et al.[32] have suggested that α-Pu

should have a CDW start at ∿60 K, as does α-U at 43 K. The CDW state in U below this temperature is incommensurate, as it may be for Pu metal below 60 K.

The possibility of not only an ICDW transition in α-Pu at ∿60 K, but of a CCDW transition at ∿30 K, comes from a comparison of the attenuation of longitudinal waves in α-Pu[33] and $NaNO_2$.[34] There is a peak in attenuation at the normal to incommensurate transition in $NaNO_2$;[34] then there is a dip followed by a smaller peak at the commensurate transition. Such a peak in ultrasonic attenuation of α-Pu at 66 K, followed by a dip and hump at a lower temperature, has been reported by Rosen et al.[33] The Hall voltage magnitude was also only sensitive to current direction below ∿25 K, as reported by Loree and Pinnick.[8] A peak in the ultrasonic attenuation of α-U at ∿40 K has also been reported by Rosen.[35]

A survey of the low-temperature physical properties of α-Pu by Lallement and Solente (Pu 1965, p. 147) also suggests a second-order transition around 50-60 K.

IV. CHIRAL SYMMETRY IN PROVEN INCOMMENSURATE CRYSTALS

The space group structures for the incommensurate region of reported incommensurate crystals all show the possibility of chiral symmetry. This is because the reported structures all have a twofold screw axis with a center of symmetry in at least one dimension.

The materials and space group structure for the incommensurate phases of proven incommensurate crystals are as follows: $NaNO_2$ (Immm, No. 71), β-K_2SO_4 (Pnma, No. 62), K_2SeO_4 (Pnma, No. 62), $RbZnCl_4$ (Pnma, No. 62), thiourea (Pnma, No. 62), $BaMnF_4$ [Cmcm(?), No. 63] and 2H-$TaSe_2$ ($P6_3/mmc$, No. 194).

The commensurate phases of these same listed incommensurate crystals have twofold screw axes but without a center of symmetry. The space group structures of these same materials, which lack chiral symmetry, have been reported as: $NaNO_2$ (Imm2, No. 44), β-K_2SO_4 ($P2_1/c$, No. 14), K_2SeO_4 (Pna2, No. 33), $RbZnCl_4$ ($Pna2_1$, No. 33), thiourea ($P2_1ma$, No.26), and $BaMnF_4$ ($Pmc2_1$, No. 26). The commensurate variant of 2H-$TaSe_2$ is Cmcm, (No. 63). The common feature of these commensurate space group structures is that

they have twofold screw axes that do not have a center of symmetry. They can be classified as nonchiral with the exception of 2H-$TaSe_2$ (Cmcm), where there must be symmetry breaking.

Memory effects have been reported for thiourea[36] and deuterated thiourea.[37] It was mentioned above that memory effects have been noted between different Pu metal phases and that there are twofold screw axes (chiral and nonchiral) in all reported structures for Pu metal phases. This section has been introduced as a background for a discussion of the possibility of ICDW and CCDW states in phases of Pu metal occurring above the α-phase.

V. POSSIBILITY OF BOTH ICDW AND CCDW STATES IN Pu PHASES ABOVE α

The space group structures for the Pu phases observed below the melting point can explain much of the hysteresis observed between phases. The space group designations of Pu metal phases indicate the presence of screw diads, screw triads, and screw tetrads. Screw axes with a center of symmetry are required by every Pu space group designation with the exception of the γ phase. Thus, every Pu metal phase except γ has chiral symmetry in at least one dimension. The space group designations for the different Pu phases and the indication of the number of twofold screw axes for each are shown in Table I.

TABLE I

Pu Phase	Space Group No.	Space Group Designation[38]	Structure	Screw Axes[26]
α	11	$P2_1/m$	mono.	Diads (1-dimen.), C. of S.[a]
β	12	I2/m (C2/m)	bc mono.	Diads (1-dimen.), C. of S.
γ	70	Fddd	fc ortho.	Diads (3-dimen.), no C. of S.
δ	225	$Fm\bar{3}m$	fcc	Diads, Tetrads, C. of S.
δ'	139	I4/mmm	bc tetrag.	Diads, Tetrads, C. of S.
ε	229	$Im\bar{3}m$	bcc	Diads, Tetrads, C. of S.

[a]With center of symmetry.

The physical properties of Pu metal phases seem to be very sensitive to impurities. This is evident from the differing results reported by experimentalists for the same measurement. Gruner and Zettl[39] point out the importance of local deformations caused by impurities. They note that as a consequence of these deformations, long-range CDW order should not exist in less than four dimensions. Their comment suggests that the phases of Pu metal, if incommensurate, may have to be considered as supercrystals with superspace group symmetry. That is, their characteristic x-ray or neutron-diffraction patterns would have to be considered as those of an n-dimensional crystal having a d-dimensional modulation. Four-dimensional space would be required, i.e., 3n + d.

DeWolff, Janssen, and Janner[40] have presented a complete list of (3 + 1) dimensional space groups. These groups describe the symmetry of incommensurate crystal structures with a one-dimensional modulation. That modulation may result from CDWs or spin-density waves. In their tabulation of superspace groups, describing incommensurate structures with a one-dimensional modulation,[40] are listed the space group structures assigned to the α, β, γ, and δ' phases of Pu. These are: $P2_1/m$, No. 11 (α-Pu); B2/m, No. 12 [β-Pu (Published as I2/m)]; Fddd, No. 70 (γ-Pu); and I4/mmm, No. 139 (δ'-Pu). The space group structures for these listed Pu phases are thus recognized as having the possibility of being superspace groups, i.e., of being incommensurate phases.

The resistivity behavior of the higher-temperature phases of Pu metal suggests CDW phases for each phase. The resistivity vs temperature curves for the α-, β-, γ-, δ-, δ'-, and ε-phases are nearly temperature-independent between ∿300 K and the melting point.[18] (See Fig 1.) The α-phase above ∿300 K and the β-phase show slightly negative temperature coefficients of resistance. The temperature coefficients are slightly positive in the δ and δ' phases. In making the resistivity measurements over the temperature range of the many phases of Pu, the self-heating property of the metal was utilized in the measuring apparatus.[18] Heating of the surrounding furnace was such that furnace wall temperature followed that of the specimen. The furnace temperature did not

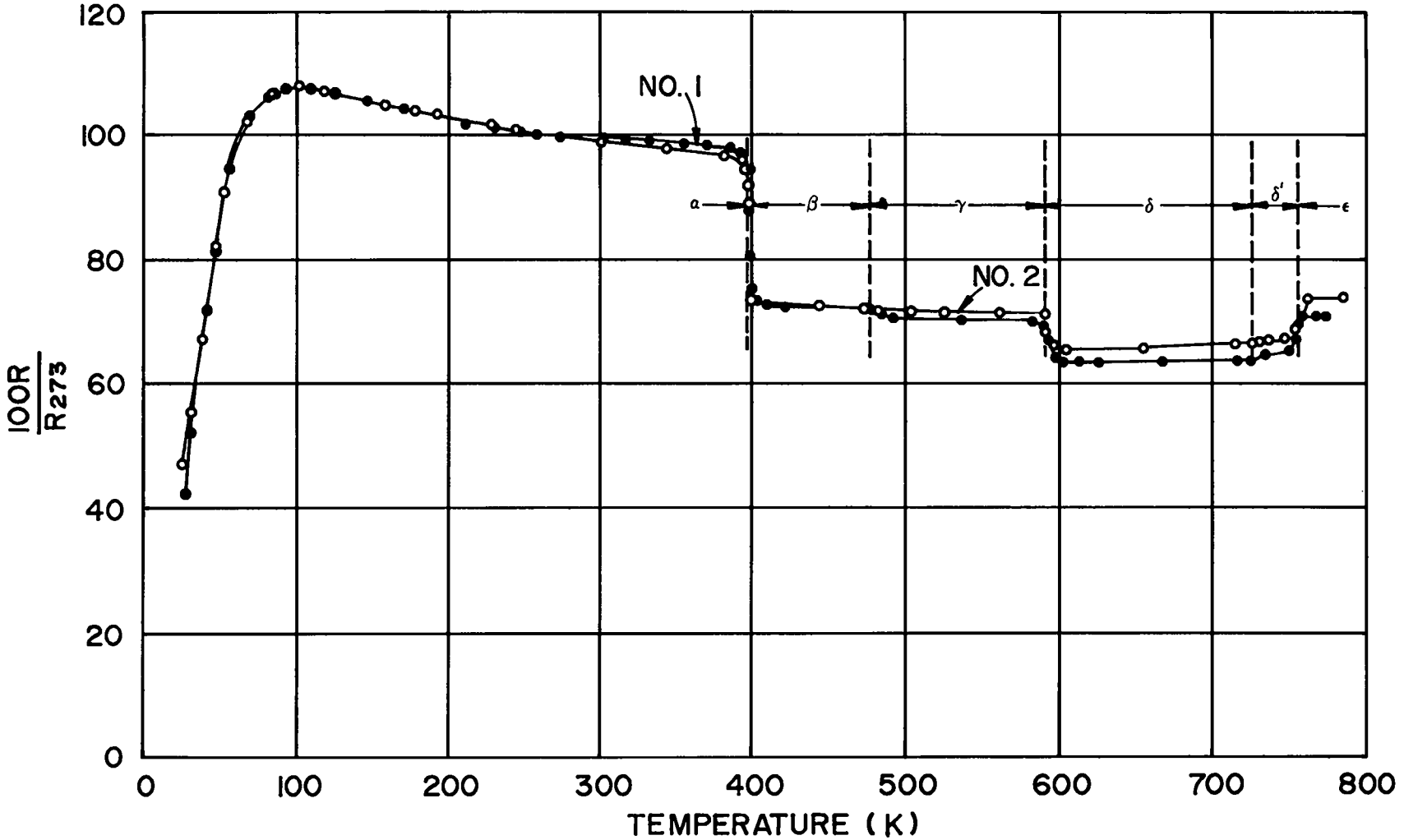

FIG. 1. ELECTRICAL RESISTANCE PLOTTED AS $\frac{100R}{R_{273}}$ VS ABSOLUTE TEMPERATURE.[18]

rise during phase transformations, so each transformation could go to completion before any increase in furnace temperature. Any forming ICDW or CCDW phase within a phase could form completely.

The possibility that a slightly negative temperature coefficient of resistance, or a temperature-independent coefficient, could be the effect of a CCDW phase comes from the study of Hwang et al.[20a,b] on $Ti_{50}Ni_{47}Fe_3$. The resistivity vs temperature curve for that material was relatively flat between the M_s temperature (173 K) and 243 K, or in the major portion of the CCDW range of the material.

The hysteresis noted in phase transformations between all Pu phases must be due to transformations that involve screw axes (and possibly CDW states) because they are all common to Pu phases and because the metal appears to be acting like an incommensurate crystal in all phases as mentioned above. Several phases are seen in some incommensurate crystals.

The existence of CDW phases could explain the hysteresis loops seen in heating and cooling of a Pu specimen between the α-phase and the ε-phase. An illustration of the hysteresis loop is shown by Fig. 1 of a paper by Hocheid et al.[41] and by Fig. 15 of a survey by Goldberg and Massalski.[11] These figures are a plot of $\Delta\ell/\ell$ vs temperature. It is a study of the expansion or contraction occurring in a specimen on heating or cooling from phase to phase. A reproduction of Fig. 1 of Hocheid et al.[41] is shown as our Fig. 2.

The large change in relative elongation between phases, as shown in Fig. 2, suggests there should be a considerable change in physical properties when a material moves from phase to phase. The largest change in resistivity, as shown in Fig. 1, appears to be between the α- and the β-phases. There appears to be no change in resistivity between the β- and γ-phases for the case of specimen no. 2. There appears to be little difference in hardness between the β-, γ-, and δ-phases as reported by different workers.[38] (p. 74.) The internal-friction and shear-modulus study of Selle and Focke[42] shows a considerable change in values, with hysteresis, at the $\beta \rightleftarrows \gamma$ transformation. (See Figures 3 and 4.)

An argument that Pu metal is acting like an incommensurate crystal also comes in observed memory effects (structural memory). Spriet[43] reported a discovery of a memory effect at the transformation interface of

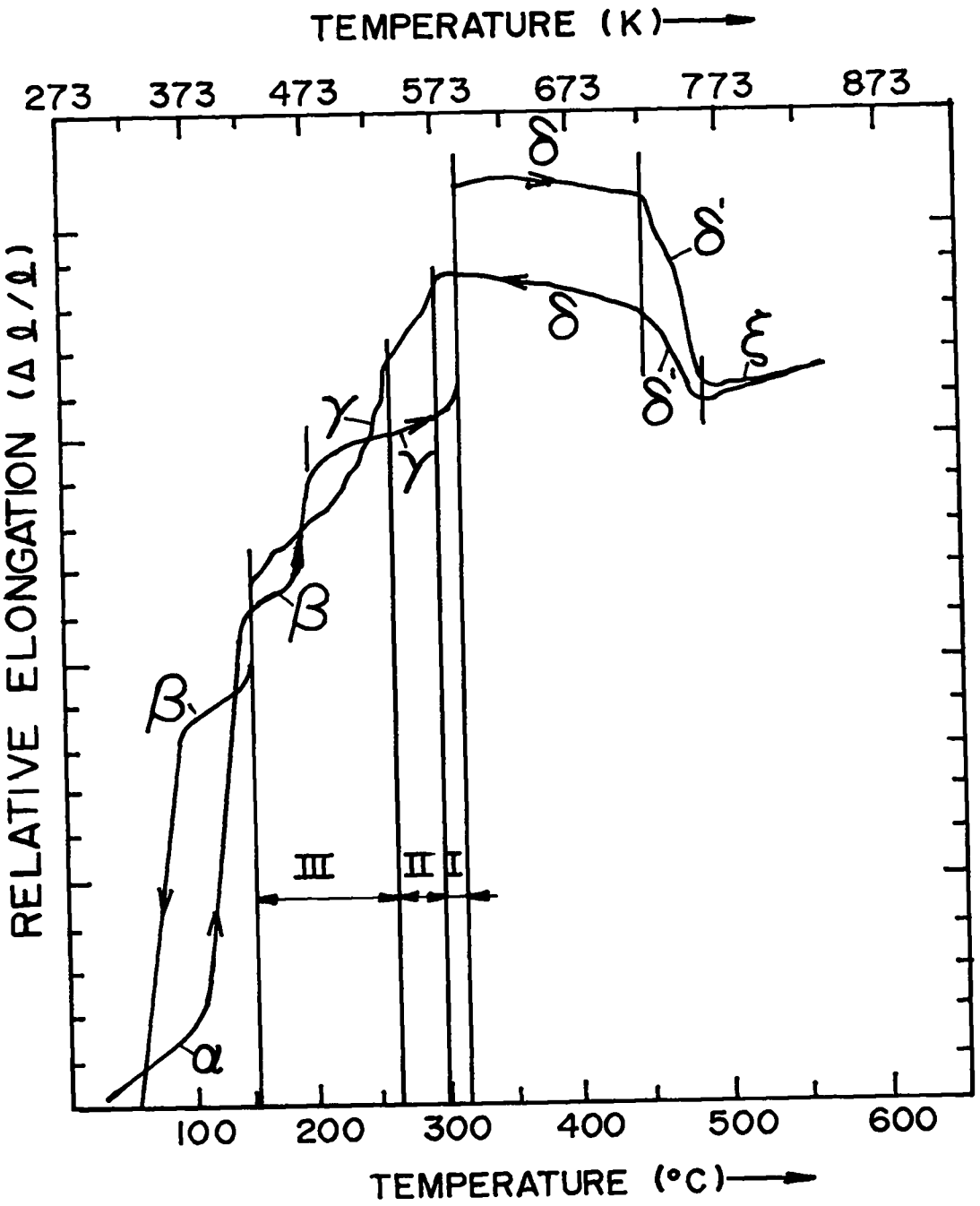

FIG. 2. HYSTERESIS IN PHASE TRANSFORMATIONS OF Pu METAL AS SHOWN BY DILATOMETRY STUDIES.[41]

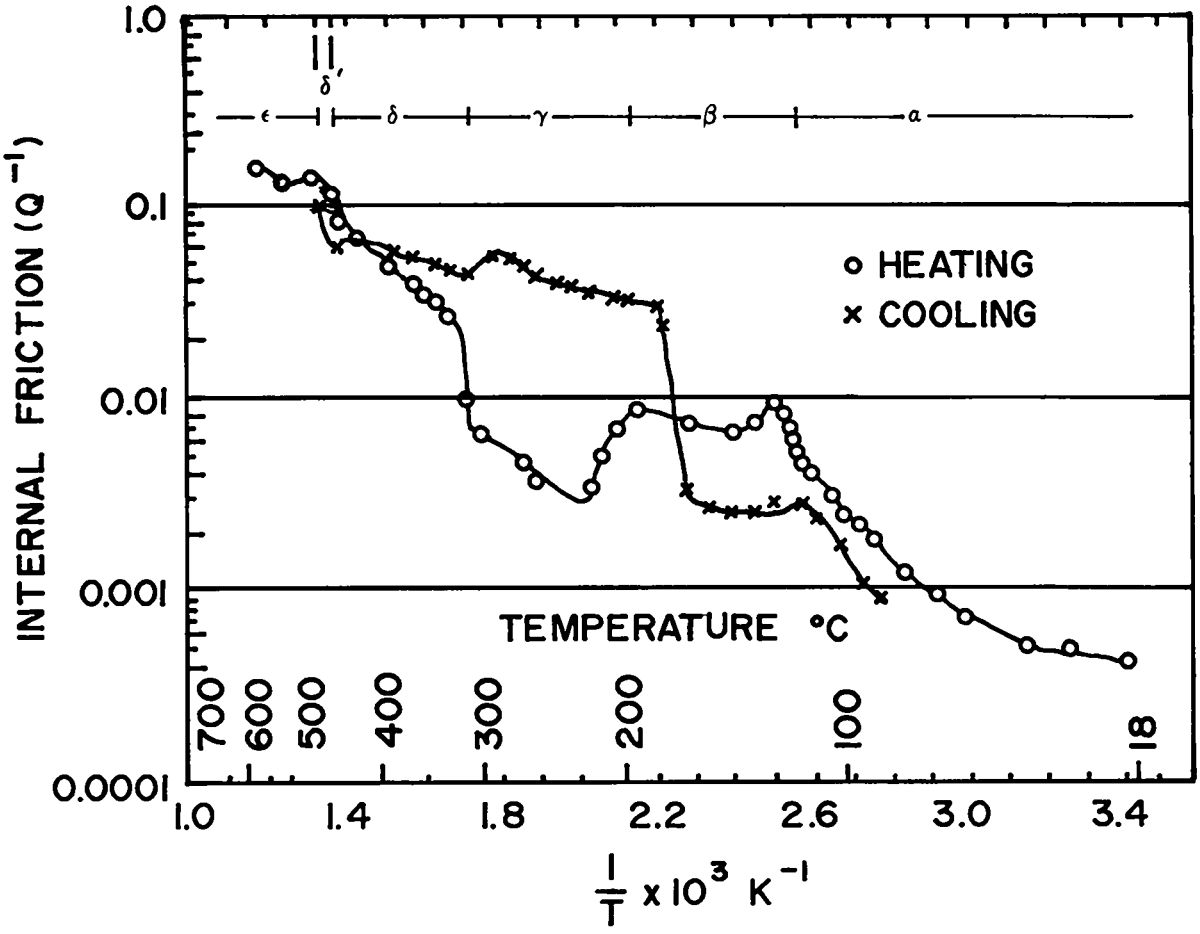

FIG. 3. STEADY-STATE INTERNAL FRICTION OF Pu AS A FUNCTION OF RECIPROCAL TEMPERATURE, FREQUENCY OF 1.343 cps. [42]

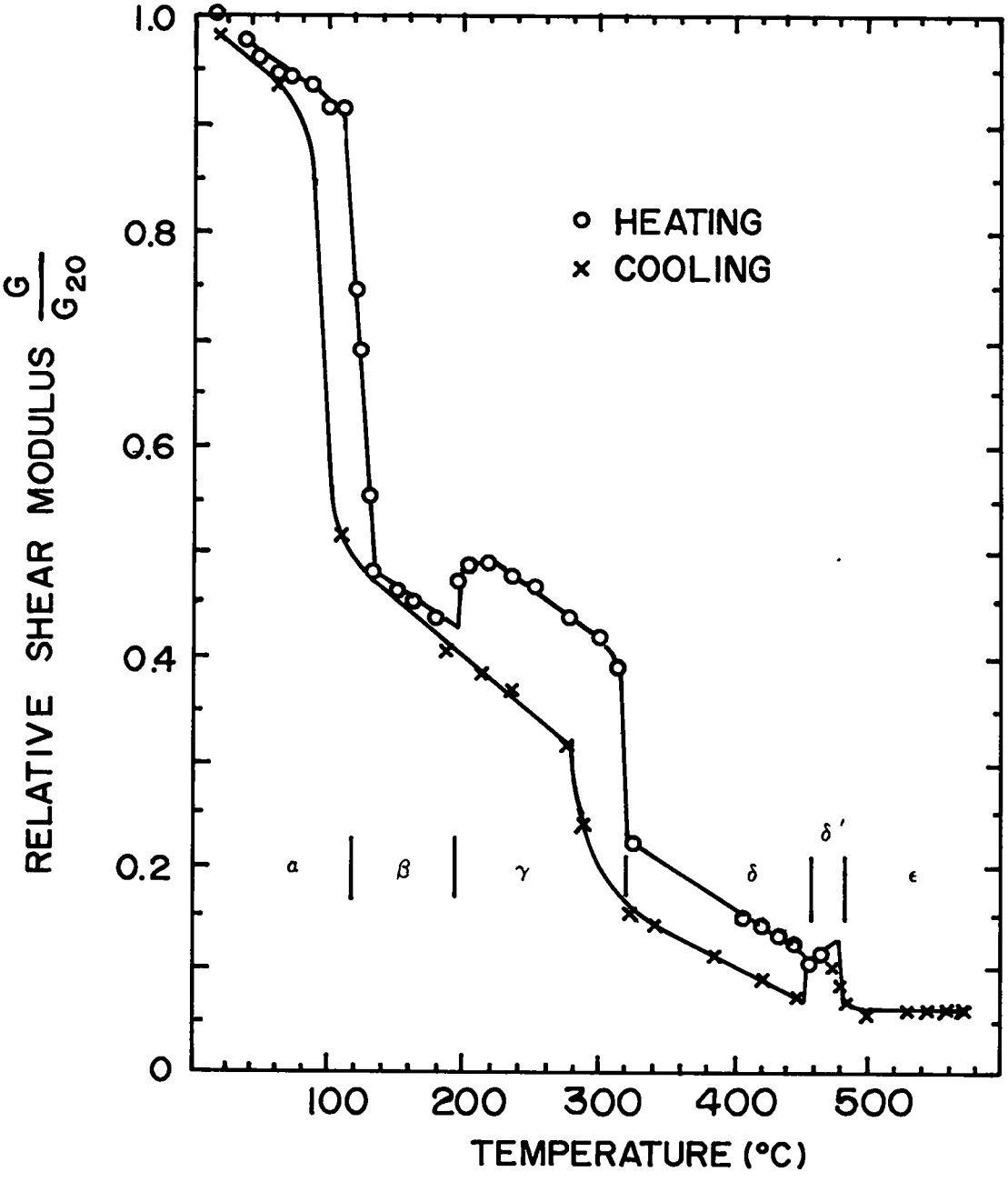

FIG. 4. RELATIVE SHEAR MODULUS OF Pu AS A FUNCTION OF TEMPERATURE.[42]

the $\beta \rightarrow \alpha$ transformation. This finding suggests that there may be a two-phase structure of α and β just below the $\alpha \rightarrow \beta$ transformations. (See Fig. 2).

Goldberg and Massalski[11] note that there seems to be a memory effect in the $\gamma \rightarrow \delta$ phase transformation. A remnant of the structure of the γ-phase and, in particular, residues of how it was formed are carried into the δ region and, if not eliminated, influence the reverse transition. Fung et al.[16] note that 2H-$TaSe_2$ exhibits a remarkable memory effect. If the material is repeatedly cycled through the CCDW$\leftrightarrow$ICDW transition and down to $\sim$30 K, the twin boundaries return to almost identical positions in spite of there being no visible reason for this to occur. The minimum in the Hall coefficient curve of 2H-$TaSe_2$ occurs at $\sim$30 K. This appears to be the lower temperature limit of the CCDW phase region.

The γ-phase range of Pu metal has a three-dimensional, twofold screw axes space group structure without a center of symmetry in any dimension (Fddd), as reported by Zachariasen and Ellinger.[44] There must be a type of symmetry breaking at about 483 K on warming because the lattice constants vs temperature of γ-Pu show a contraction in one dimension.[44] The structure appears to develop chiral symmetry in one dimension and could be a type of striped phase at the $\gamma \rightarrow \delta$ phase transformation with CCDWs in two dimensions and an ICDW in the third.[17a] The γ-phase should be predominantly commensurate on a warming cycle from the β-phase and thus show a temperature-independent resistivity.

The $\gamma \rightarrow \delta$ phase transformation involves a change from what appears to be a one-dimensional chiral symmetry to another form with three-dimensional chiral symmetry. The δ-phase should be incommensurate as there is a three-dimensional contraction (on warming), which is not obvious in Fig. 2.

The major specimen contraction occurs in the δ'-phase. The mean contraction of the c-axis is given as $-1063.5 \pm 18.2 \times 10^{-6}/^{o}C$.[38] The twofold and fourfold screw axes in this phase (δ') are one-dimensional and chiral. The Pu specimen should be fully incommensurate by the $\delta' \rightarrow \varepsilon$ transition as there appears to be a normal expansion in the ε-phase. The ε-phase (No. 229) may be the only normal CDW phase of the Pu metal system. It may be a case where the three-dimensional, twofold and fourfold screw axes with their center of symmetry (chiral) are balanced by the nonchiral twofold,

threefold, and fourfold screw axes. It should be noted that the δ' space group structure (No. 139, I4/mmm) is a maximal non-isomorphic subgroup for both the δ-phase structure (No. 225) and the ε-phase structure (No. 229). (See pages 680 and 704 of Ref. 26.)

The large contraction shown in the δ'-phase range by Hocheid et al.[41] and reproduced on our Fig. 2 may be due to chiral symmetry of the three phases, δ, δ', and ε. The actual contraction may be combined action of these three listed phases. This suggestion comes from observations of Ellinger[45] made as a result of the crystal structure determination of δ'-Pu. The comments of Ellinger[45] are essentially as follows: A small portion of ε forms at 450°C (723 K), which coexists with δ up to 465°C (738 K). X-rays did not reveal δ' until about 465°C (738 K). At this temperature the metal transforms largely to δ'. A small amount of δ and ε continue to coexist with δ' up to 485°C (758 K). At 485°C (758 K), the proportion of δ is decreasing and the proportion of ε is increasing. At 490°C (763 K), the metal transforms completely to ε.

VI. INTERNAL FRICTION AND RELATIVE SHEAR-MODULUS RESULTS AS CONFIRMATION OF ICDW AND CCDW STATES IN Pu PHASES

An internal-friction study of the allotropic transformations of Pu metal was reported in 1969 by Selle and Focke.[42] They showed figures for the steady-state internal friction and relative shear modulus of Pu-phases as a function of temperature. Their figures showed not only hysteresis in transformation temperatures, but also large swings in internal-friction or shear-modulus values dependent upon whether data were taken on heating or cooling cycles.

Barmatz et al.[46] have reported results for Young's modulus and internal-friction measurements made on the layered dichalcogenides $TaSe_2$ and $NbSe_2$ through normal to ICDW and ICDW to CCDW transitions. They noted that Young's modulus and internal friction exhibited extremes at the incommensurate $\rightleftarrows$ commensurate transitions and that there were large hysteresis effects. A maximum in internal-friction data occurred at the ICDW→CCDW transition temperature, and a minimum in Young's modulus occurred near the same temperature. Barmatz et al.[46] also noted a small hump in

internal-friction values at the normal to incommensurate transition at 121 K in 2H-$TaSe_2$ (their Fig. 3) and a small hump in modulus values through the ICDW temperature range (their Fig. 2).

The results of Selle and Focke[42] give needed information for deciding if one or more of the allotropic transformations in Pu metal are ICDW $\overset{\rightarrow}{\leftarrow}$ CCDW transitions. The criteria should be whether there is an increase in internal friction at approximately the same temperatures where a rapid decrease in shear modulus occurs.

ICDW $\overset{\rightarrow}{\leftarrow}$ CCDW transitions have been listed as displacive (shear). The transformation mechanisms for phase transformations in Pu metal have been tabulated by Selle and Focke.[42] They suggested that the $\alpha \rightarrow \beta$ and the $\beta \rightarrow \alpha$ transformations are always by shear and that the $\beta \rightarrow \gamma$ and $\gamma \rightarrow \beta$ transformations are always by diffusion. The other transformations are different on heating than they are on cooling. The $\delta \rightarrow \gamma$, $\delta \rightarrow \delta'$, and $\delta' \rightarrow \varepsilon$ are shear transformations, whereas $\gamma \rightarrow \delta$, $\varepsilon \rightarrow \delta'$, and $\delta' \rightarrow \delta$ appear to be diffusional. Transformations that are different on heating and cooling cycles should show hysteresis. Also, if a transformation is martensitic, one should expect the M_s and M_f temperatures observed on cooling to be quite different than the A_f and A_s temperatures observed on warming.

Because the $\alpha \overset{\rightarrow}{\leftarrow} \beta$ transformation of Pu metal appears to be martensitic (shear) in nature, any hysteresis in physical properties at this phase transformation does not require a CDW explanation, but one appears necessary. The behavior on warming from the α-phase to the β-phase, as shown by internal-friction and relative shear-modulus results,[42] suggests the transition is from a CCDW to an ICDW state in β-phase. There is a small hump in internal friction at the $\alpha \rightarrow \beta$ transformation and a large decrease in relative shear modulus (Figs. 3 and 4).

Evidence suggests that the γ-phase is in a CCDW state on warming. The shear modulus rises at the $\beta \rightarrow \gamma$ transformation while the internal friction decreases appreciably. The shear modulus remains high in the γ-phase.

The relative shear modulus drops sharply at the $\gamma \rightarrow \delta$ transformation (on warming) while the internal friction values rise sharply.[42] This hints that the δ-phase of Pu is in an ICDW state on warming. The hump in relative shear modulus seen through the short δ'-phase range also suggests

that this phase is ICDW on warming. This hump is like that seen in the ICDW phase range of 2H-$TaSe_2$.[46]

A slight maximum in internal-friction values occurs at the $\delta' \rightarrow \varepsilon$ transformation in Pu.[42] The state of the ε-phase could be CCDW, but it is more likely to be normal CDW because of the temperature-independent nature of the relative shear modulus on the cooling cycle.

The repeat of the small hump in relative shear modulus on cooling through the δ'-phase[42] (as also seen with 2H-$TaSe_2$[46]) suggests that it is also an ICDW state, as it was on warming.

There was very little change in internal friction at the $\delta' \rightarrow \delta$ transformation, but there was a moderate decrease in relative shear modulus.[42] The internal-friction values decreased slowly on cooling through the δ-phase range, whereas the shear-modulus values increased. The δ-phase must be in an ICDW state, as it was on warming. The differing nature of phase transformations could account for hysteresis at $\delta \overleftrightarrow{} \delta'$ and $\delta' \overleftrightarrow{} \varepsilon$ transitions.

The internal-friction and relative shear-modulus behavior on cooling through the γ- and β-phase ranges[42] is anomalous. CDW states seem to be completely reversed from what they were on the warming cycle. The internal-friction behavior of Pu metal on cooling through the γ-phase range appears to be a continuation of the δ-phase behavior. There may be a gradual change as shown by Region I on Fig. 2. The shear-modulus behavior on cooling also suggests that the γ-phase remains in an ICDW state.[42] This may be only one-dimensional, as suggested by x-ray data of Zachariasen and Ellinger[44] where there was a contraction in one dimension of the γ-phase structure. This may be represented by Region II of Fig. 2. What is being seen on cooling through Region III (Fig. 2) may be a step-wise transformation of the ICDW states into commensurate states with completion at the $\gamma \rightarrow \beta$ transition.

The results of Selle and Focke[42] showed a large decrease in internal friction at the $\gamma \rightarrow \beta$ phase transformation, whereas there was little change in relative shear modulus. The relative shear-modulus level for the β-phase (cooling) appeared to be approximately the same as it was for the phase on warming. This possibility of different CDW states on warming and cooling could account for the great difference in ductility reported by Nelson et al.[47] for β-phase formed from α-phase and the β-phase formed from

γ-phase. The inference from the low level of internal-friction values noted through the β-phase on cooling is that the β-phase may initially be CCDW. There is increasing hardness below ∿160 K in the α-phase range of Pu metal where the Hall coefficient behavior suggests a CCDW state.

VII. CDW CONTRIBUTIONS TO f-BONDING IN Pu

Reasons given for the distortion and modulation of CDWs suggest that CDWs may be playing a role in the f-bonding of Pu metal and other light actinide metals.

McMillan[48] considered that the physics of the distortion was simple; the CDW gains bonding energy by placing the charge-density peaks between transition-metal atoms to form bonding charge. An f-bond (light actinides) is explained as meaning that f-electrons are conducting and bonding.[49] If the charge-density peaks were between Pu atoms, then the f-electrons could be part of the CDW bonding.

Bak[50] considered that an incommensurate structure arises from interactions between conduction electrons and the atomic lattice, the so-called Peierl's mechanism. In his explanation, the conduction electron density in the distorted phase is spatially modulated to form the CDW that accompanies the periodic lattice distortion. It has been suggested above that breaking of chiral symmetry, or a Peierls transition, may play a considerable role in the ICDW or CCDW nature of Pu metal phases.

VIII. SUMMARY

The space group structure of α-phase Pu ($P2_1/m$) is also seen in TiNi martensite,[3a,b] TaS_3,[5] and $NbSe_3$.[6] Experimental evidence suggests that these materials have CDW phases and in some cases solitary excitations (solitons). Walker and Jacobs's theory[13] also implies a CCDW phase for this space group. It is reasonable to conclude that α-Pu should also have CDW states.

A similarity in hysteresis loops in resistivity vs temperature curves for O-TaS_3,[19] $Ti_{50}Ni_{47}Fe_3$,[20] and α-Pu[7] hints that there should be an ICDW phase in α-Pu between ∿160 K and ∿290 K. A similarity in Hall coefficient

behavior of α-Pu[8] and 2H-$TaSe_2$[21] also suggests that there are both CCDW and ICDW phase ranges below 300 K in α-Pu. The fact that the Hall voltage[8] became nonlinear above a critical current density also appears to be proof of CDW states.

The similarity in the Hall coefficient behavior of 2H-$TaSe_2$ and α-Pu metal along with the lattice parameter behavior of Pu metal below 60 K intimates that there may be a new, ICDW, orthorhombic phase αI below this temperature, possibly Pmnm. A comparison of the attenuation of longitudinal waves in α-Pu and $NaNO_2$ (an incommensurate crystal) suggests the possibility of another CCDW state below ∿30 K. Proof for this possibility comes from the comment of Loree and Pinnick[8] that Hall voltage magnitude was only sensitive to current direction below ∿25 K.

It is suggested that physical property behavior of all Pu metal phases may be influenced by CDW states and that most of the noted phase transformations may be between CDW states. The energy barrier between phases may be solitons (discommensurations or domain walls).

The hysteresis effects between phases are attributed to the chiral nature of screw axes, diads, and tetrads and their possible ability to contract or expand like springs and be the source of contraction in one lattice direction when there may be an expansion, sometimes different, in the other two lattice directions. Attention is called to a comment by Su and Sakita[9] that chiral symmetry is inherent to the incommensurability of a quasi-one-dimensional CDW system.

Space group structures, which have been assigned to four Pu phases, are listed in a tabulation of superspace groups describing incommensurate structures with a one-dimensional modulation.[40] The diffraction spots of these phases, α, β, γ, and δ', may have to be considered in four-dimensional space. The space group assignments of Pu metal phases may be added proof of the incommensurate nature of that metal.

It is proposed that CDWs may be contributing to f-bonding in Pu metal. It is noted that there is a memory effect in both the $\alpha \leftrightarrow \beta$ and the $\gamma \leftrightarrow \delta$ transformation. A memory effect has also been reported in the CDW material 2H-$TaSe_2$ and in many incommensurate crystals.

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