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Temperature dependence of elastic moduli of polycrystalline β plutonium

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The elastic moduli of pure polycrystalline beta plutonium were measured over its full range of existence (417–491 K) using resonant ultrasound spectroscopy. The Debye temperature (138 K), Poisson's ratio (0.28), Gruneisen parameter (2.3), and the zero-temperature atomic volume (21.2 Å^3) were computed from the measurements. Both bulk and shear moduli decrease smoothly on warming with expected discontinuities at the phase boundaries. The shear modulus is surprisingly nearly the same for beta and gamma Pu. The temperature dependence of bulk moduli for beta Pu is, like gamma Pu, unusually small. Poisson's ratio shows very strong differences among alpha, beta, and gamma Pu indicating they are entirely different metals. The zero-temperature elastic moduli were computed for the three phases as well as for gallium-stabilized delta Pu (also measured by us) and compared to calculations.

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I. INTRODUCTION

Plutonium is one of the two most interesting elements because it has six crystallographic allotropes, unusual thermal expansion, and apparently completely different bonding behavior among its phases.[1] Some of its isotopes can sustain a fission chain reaction.[2] Surprisingly, neither accurate elastic moduli measurements exist, nor do satisfactory first-principles electronic structure models; the first because of the difficulties in working with Pu,[3–6] the second because Pu electrons straddle the itinerant/localized boundary, frustrating understanding.[7–11] Lacking essential measurements for a very challenging element further distances understanding.

The lowest-temperature (α) phase (Fig. 1) of Pu crystallizes in a low-symmetry monoclinic structure, unusual for any metal, with a 16-atom unit cell.[13] At 398 K, the volume expands by 10% and forms a body-centered monoclinic structure, perhaps, with a 34-atom unit cell (β) phase.[14] At 488 K, the volume expands by 3% and forms a face-centered orthorhombic structure with an eight-atom unit-cell (γ) phase.[15] At 593 K, the volume expands by 7% (25% larger than that of the α-phase) and forms the (close-packed) face-centered cubic (δ) phase with the largest solid volume and negative thermal expansion.[16] The volume contracts at 736 K for the δ'-phase with dubious properties,[17] followed by the (ε) body-centered cubic structure at 756 K (Ref. 16) until it liquefies at 913 K.[18] This complex phase diagram occurs within a factor of 2 in absolute temperature,[19] indicating how delicate the energy and entropy balances must be.

One set of properties that is incompletely measured[20–24] is the set of adiabatic elastic moduli, the second derivatives of energy with respect to strains, and often the first properties computed from the many electronic structure models.[25–39] Elastic moduli are also one of a few thermodynamic susceptibilities that are essential for an understanding of the phases of Pu[40–50] and, with their temperature dependence, are important for applications.

For these reasons, Young's modulus and shear modulus were measured by several investigators. In 1960, by a resonance method for all six phases,[51,53] Pu was thermally cycled many times. After each cycle, the moduli changed.[53] The most reliable bulk modulus measurements were performed by Kay and Linford[52] (Fig. 2). Young's modulus was also measured by an extensometer for four phases[54] and by an ultrasonic resonance method for six phases.[55] There are many elastic modulus measurements at room temperature for α-Pu.[56–59] Temperature dependence of α-Pu moduli over a limited temperature range was measured by an ultrasonic resonance method,[60–62] by an ultrasonic pulse technique,[63] and by noncontacting laser ultrasonics.[64] The β-α transformation was studied with an ultrasonic pulse technique.[65] The relative shear modulus as well as ultrasonic attenuation were measured using an inverted torsion pendulum for the six phases.[66] All pure Pu measurements were performed with polycrystalline samples because monocrystalline pure Pu was not available, however, monocrystalline elastic moduli were measured in gallium-stabilized δ-Pu (δ-Pu 3.3 at. % Ga) at ambient temperature using a pulse-echo method by Ledbetter and Moment.[67] Inelastic x-ray scattering measurement produced values close to the Ledbetter and Moment result.[68] The temperature dependence of elastic moduli for polycrystalline δ-PuGa was measured by noncontacting laser ultrasonics,[64] by a high-pressure x-ray measurement,[69] and by resonant ultrasound spectroscopy (RUS) for different gallium concentrations.[70] δ-PuAl was also measured by an ultrasonic resonance method[62] and RUS.[44] The change of elastic moduli with time was used to study Pu aging by self-irradiation in α-Pu and δ-PuGa.[71]

II. MEASUREMENTS

RUS (Refs. 72–74) measurements of the adiabatic bulk and shear moduli of pure polycrystalline β-Pu are reported here. The Pu measured had the highest available purity, and measurements were completed without thermal cycling. One specimen was used for the work described here on the β-phase. It was also used for measurement of α-Pu above 300 K, and γ-Pu. The specimen was made from electrorefined ^{239}Pu with

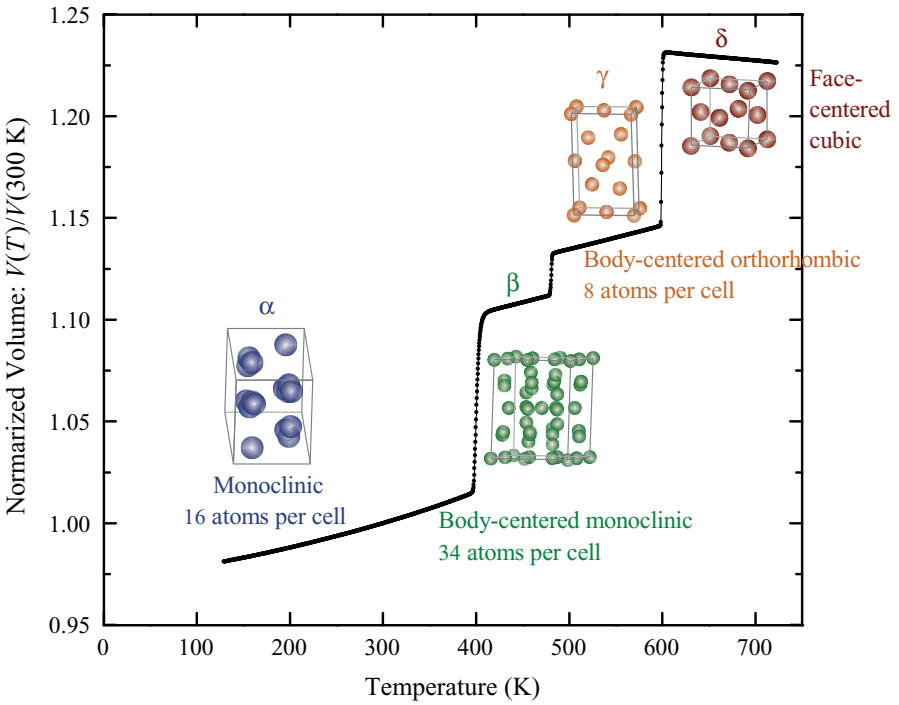

FIG. 1. (Color online) Normalized volume of pure Pu relative to 300 K was derived from dilatometer measurements and is plotted from 120 to 720 K for α-Pu, β-Pu, γ-Pu, and δ-Pu.[12]

99.96 wt % Pu, 115 ppm W, 49 ppm Np, 50 ppm O, 53 ppm Si, 32 ppm Am, and the sum of remaining impurities less than 25 ppm. The specimen was cut from a larger button that was arc melted and was quenched to room temperature on a copper hearth, cut and polished, examined optically for voids and metallurgical defects, and the process was repeated (about ten times) until metallurgical imperfections were judged negligible.[21]

By the usual metallographic cut-grind-polish method, the specimen was prepared in a parallelepiped of 0.265 × 0.268 × 0.270 cm^3 all ±0.002 cm. The immersion density at 300 K was determined to be 19.55 g/cm^3 ±0.02%. The

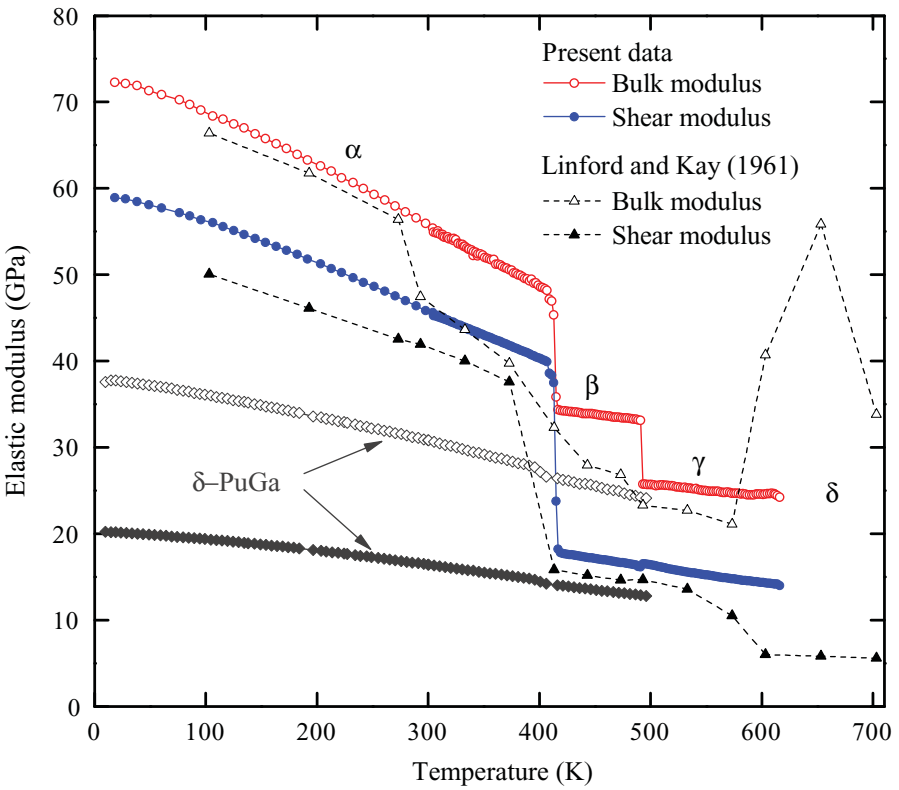

FIG. 2. (Color online) Bulk and shear moduli of pure Pu from 18 to 616 K for α-Pu, β-Pu, and γ-Pu compared to Kay and Linford.[52] The bulk moduli (empty diamonds) and shear moduli (solid diamonds) for δ-Pu 2.36 at. % Ga (previous work) are also plotted and are consistent with the values of pure Pu because the extrapolated values of δ-Pu 2.36 at. % Ga into pure γ-Pu's temperature range are smaller than that of γ-Pu as expected.

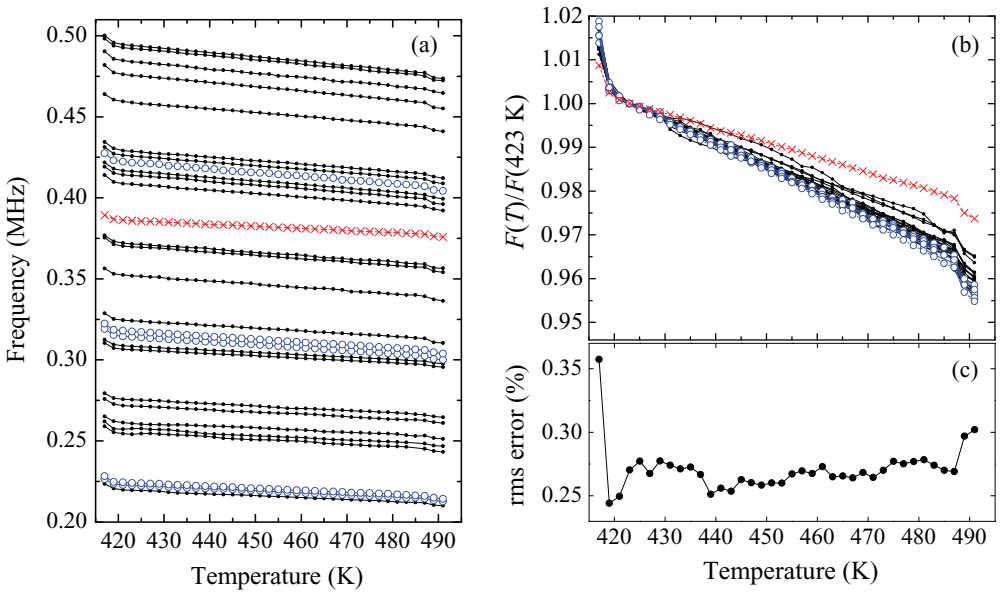

FIG. 3. (Color online) The temperature dependences of 28 resonances, which were used for the RUS fit for β-Pu, are plotted in (a). They are smooth and almost linear except at the phase boundaries. The RUS fit determined that the resonances in circles (○) in (a) and (b) are associated almost solely with the shear modulus, G, while crosses (×) are partially dependent on the longitudinal modulus C_L. The ones in black depend on linear combinations of G and C_L. The frequencies in (a) were divided by the values at 423 K and are plotted in (c). This clearly shows the difference in the temperature dependences for the two modes. In (c), the rms percent of errors for 28 resonances are plotted. They are very small, less than 0.3% and a little larger at the edges of the β-phase.

density determined from mass and dimensions at 300 K was 19.70 g/cm^3, and the x-ray diffraction density was 19.86 g/cm^3 at 294 K. While the differences are small, thermal activation of defects makes it impossible to achieve (and unreasonable to expect) x-ray density in Pu above cryogenic temperatures.

Temperature was controlled by a helium gas-flow cryostat. Measurements were made in (constantly pumped) vacuum.

TABLE I. Elastic moduli (B, bulk; G, shear; C_L, longitudinal; and E, Young's modulus) and ν (Poisson's ratio) of α-Pu,[20,21] β-Pu,[39] γ-Pu,[22] and δ-Pu 2.36 at. % Ga (Ref. 24) at selected temperatures.

Temperature (K)	Phase	B (GPa)	G (GPa)	C_L (GPa)	E (GPa)	ν
18.36	α	72.28	58.91	150.82	138.97	0.1795
95.82		69.03	56.36	144.18	132.90	0.1791
203.3		62.56	51.24	130.88	120.75	0.1783
297.9		55.93	45.85	117.05	108.02	0.1781
407.0		48.17	39.91	101.39	93.82	0.1754
415.0	α-β[a]	35.81	23.74	67.46	58.33	0.2285
417.0	β	34.36	18.22	58.65	46.44	0.2748
451.0		33.80	17.09	56.59	43.89	0.2836
491.0		33.11	16.12	54.60	41.61	0.2905
493.0	γ	25.72	16.51	47.74	40.81	0.2356
551.0		24.96	15.19	45.22	37.89	0.2470
616.0		24.23	14.00	42.90	35.21	0.2578
14.63	δ[b]	37.77	20.18	64.68	51.39	0.2732
94.48		36.21	19.43	62.12	49.45	0.2724
203.8		33.46	18.00	57.46	45.79	0.2719
299.9		30.80	16.39	52.65	41.76	0.2740
400.9		27.13	14.43	46.37	36.77	0.2741
451.2		25.48	13.49	43.47	34.40	0.2750
496.0		24.11	12.78	41.15	32.58	0.2748

[a]See Ref. 75.
[b]δ-Pu 2.36 at. % Ga.

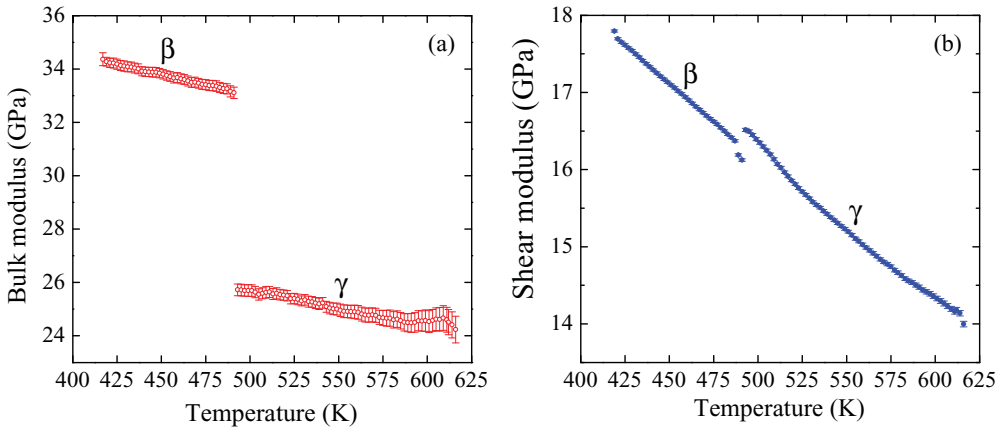

FIG. 4. (Color online) The bulk and shear moduli for β-Pu and γ-Pu are plotted in (a) and (b). The error bars represent the uncertainty that was produced by the inverse RUS calculation. The bulk modulus has larger error bars because few of the measured modes depend strongly on it. The nonmonotonic behavior of the bulk modulus for γ-Pu at high temperatures is probably an artifact of the RUS fit. A monotonic dependence is within the error bars. The bulk modulus has a clear jump at the β-γ transition. The shear modulus is similar between β-Pu and γ-Pu. This is surprising for a temperature-driven phase transition because shear phonon softening is often the primarily entropic driver of high-temperature phase transitions.[79] The bulk modulus, which contributes less to lattice entropy, has a large decrease at the β-γ transition as expected, overwhelming the anomalous behavior of the shear modulus. The lattice entropy also depends on the atomic density that is reduced by 3% at the β-γ transition. Nevertheless, the calculated Debye temperature of γ-Pu is slightly larger than β-Pu. While this calculation is just a crude estimate, β-Pu and γ-Pu are closely competing phases in entropy and energy.[29,41]

The specimen was heated only once (after sample preparation where it was hearth quenched) from room temperature to 650 K by ~2 K/h. A crude dilatometer recorded length jumps at the same temperatures at which the elastic moduli displayed steplike changes at the phase transitions.

The technique used for the measurements, RUS, acquired resonance frequencies that were very different for different phases, while the variation for frequencies within phases was smooth.[75] The typical resonances and the detailed descriptions for the RUS fit process for α-Pu,[20,21] γ-Pu,[22] and δ-Pu 2.36 at. % Ga (Refs. 23 and 24) are described elsewhere.

For the specimen of β-Pu (see Fig. 3) used here, the frequency typically was swept from 0.2 to 1 MHz.[76] The inverse calculation (RUS fit)[77] was used to obtain two polycrystalline elastic moduli, C_L (the longitudinal sound speed is controlled by this modulus) and the shear modulus, G where the bulk modulus can be calculated by $B = C_L - (4G/3)$ Errors arise principally from errors in sample geometry and the measurement of dimensions. The recorded resonances have relatively low Q (typically 400–700) with many resonances overlapping with each other. The resonances that appeared as shoulders of larger resonances or had large background noise were omitted from the fit. The fitting procedure obtained small ~0.3% rms errors[77] (the rms error is not the error for the moduli) using 28 frequencies to determine two moduli.[78] The estimated errors for the moduli are 0.09% for G and 0.33% for C_L (0.61% for B).

For the RUS fit, the x-ray density was used in the calculation. It is easy to scale to other densities. Measured thermal expansion (or contraction) was used to correct density in Fig. 1 for all the phases. The relation in Eq. (1) also can be used for density correction,

$$C(\rho_1) = C(\rho_0)\left(\frac{\rho_1}{\rho_0}\right)^{1/3}, \quad (1)$$

where $C(\rho_1)$is the RUS-computed elastic modulus when the sample was assumed to have the density ρ_1.

TABLE II. Parameters obtained for linear fits for the measurements of B, G, and C_L of the form $a + bT$ for β-Pu from 421 to 487 K.

Elastic modulus	a (GPa)	b (GPa/K)
B	40.58	−0.01509
G	26.07	−0.01990
C_L	75.34	−0.04162

III. RESULTS

Table I shows elastic moduli of pure Pu and gallium-stabilized δ-Pu (δ-Pu 2.36 at. % Ga) measured by us at selected temperatures. The elastic moduli of β-Pu and γ-Pu are plotted in Fig. 4. The shear modulus is surprisingly similar between β-Pu and γ-Pu (even more surprising, the shear modulus is slightly larger for γ-Pu) while the bulk modulus decreased at the β-γ transition as expected. γ-Pu resonances were analyzed slightly beyond the temperature range reported previously[22] for its full range of existence (493–616 K).[80] The specimen was measured above 616 K. At 618 K, the dilatometer and resonances exhibited a jump, indicating that it was in the δ-phase. However, at the same time, the sample seemed to have lost good contact with the transducers. We were unable to detect a sufficient number of resonances to perform the RUS fit to obtain accurate elastic moduli for δ-Pu. For β-Pu, the temperature dependences of both bulk and shear moduli are quite linear (except at the phase boundaries). Table II contains

TABLE III. Comparison of the fractional changes of the bulk and shear moduli with temperature at relatively high temperatures. The temperature dependence of bulk moduli for β-Pu and γ-Pu is unusually small. The others values are quite similar to each other.

	α (380 K) [a]	β (450 K)	γ (550 K) [a]	δ (480 K) [a]
$-dB/B\,dT$ (K^{-1})	1.4×10^{-3}	$\mathbf{0.4 \times 10^{-3}}$	$\mathbf{0.4 \times 10^{-3}}$	1.3×10^{-3}
$-dG/G\,dT$ (K^{-1})	1.4×10^{-3}	1.2×10^{-3}	1.4×10^{-3}	1.2×10^{-3}

[a]Reference 22.

parameters obtained from linear fits to the measurements of B, G, and C_L. We found the transition temperatures for α-β, β-γ, and γ-δ to be 415, 492, and 617 K, respectively, which are higher than what others found. Thermometry and its errors were described previously.[21]

In Fig. 2, the elastic moduli of α-Pu, β-Pu, and γ-Pu, measured on the same high-quality sample without thermal cycling are plotted along with the measurements by Kay and Linford.[52] Even accounting for the scatter in the previous study, the larger moduli observed here are expected for higher-purity single-phase material. The bulk moduli (empty diamonds) and shear moduli (solid diamonds) for δ-Pu 2.36 at. % Ga, measured by our group, are also plotted and are consistent with the values of pure Pu because extrapolated values of δ-Pu 2.36 at. % Ga into pure δ-Pu's temperature range are smaller than that of γ-Pu as expected. We note that the thermal expansion of δ-Pu 2.36 at. % Ga approaches that of pure δ-Pu as gallium content is reduced.[81] δ-Pu 2.36 at. % Ga reported here has a stable δ-phase at low temperatures.[70]

Table III shows the temperature dependence of the fractional change of the bulk and shear moduli at relatively high temperatures. The temperature dependence of bulk moduli for β-Pu and γ-Pu is unusually small, while the temperature dependences of the shear moduli for β-Pu and γ-Pu are similar to both the shear and bulk moduli of α-Pu and δ-Pu 2.36 at. % Ga.

The Varshni function[82,83]

$$C(T) = C^0 - \frac{s}{\exp(\theta/T) - 1} \tag{2}$$

is commonly used to fit the temperature dependence of elastic moduli where C^0, s, and θ are adjustable parameters. Here, C^0 denotes a zero-temperature elastic modulus, θ is closely related to the Einstein characteristic temperature, and $s/2$ is the difference between C^0 and the zero-temperature harmonic elastic stiffness coefficient obtained by extrapolating dC/dT linearly from high temperatures.[84,85] For $T \gg \theta$, the derivative of Eq.(2) is

$$\frac{\partial C(T)}{\partial T} = -\frac{s}{\theta}. \tag{3}$$

The linear extrapolating function from high temperatures is expressed as

$$\tilde{C}(T) = C^0 + \frac{s}{2} - \frac{s}{\theta}T, \tag{4}$$

and $\tilde{C}(\theta/2) = C^0$. Therefore, the zero-temperature elastic moduli can most accurately be obtained from high-temperature measurements by linear extrapolation to $T = 3\Theta_D/8$ where $\theta \approx \Theta_E \approx 3\Theta_D/4$,[86] Θ_E is the Einstein temperature, and Θ_D is the Debye temperature.

The Debye temperature, was computed using[79,87]

$$\Theta_D = \frac{h}{k}\left(\frac{3}{4\pi V_a}\right)^{1/3} \upsilon_m, \tag{5}$$

where h, V_a, k, and υ_m denote Planck's constant, the atomic volume, Boltzmann's constant, and the mean-sound velocity, respectively. We used the x-ray diffraction value[14] for V_a ($\rho = M/N_A V_A$ where N_A is Avogadro's number, and $M = 239$ is the atomic weight). The mean-sound velocity can be obtained by

$$\upsilon_m = \left[\frac{1}{3}\left(\frac{1}{\upsilon_l^3} + \frac{2}{\upsilon_t^3}\right)\right]^{-1/3}, \tag{6}$$

where the longitudinal sound velocity, $\upsilon_l = \sqrt{C_L/\rho}$ and the transverse sound velocity, $\upsilon_t = \sqrt{G/\rho}$.

The zero-temperature elastic moduli and V_a^0 (estimated value for V_a at 0 K) were used for the computation of Θ_D. V_a^0 was estimated by extrapolating the high-temperature x-ray and thermal-expansion measurements to 0 K using Grüneisen's relationship,

$$\beta = \frac{\gamma C_p}{V B_S}, \tag{7}$$

TABLE IV. The zero-temperature elastic moduli, the Debye temperatures, and the Grüneisen parameters for β-Pu compared to literature values.

Method	B^0 (GPa)	G^0 (GPa)	C_L^0 (GPa)	Θ_D (K)	γ
Kay (elastic moduli)[88]	46.2	24	78	133	
Wallace ($\gamma = V\beta B_S/C_P$)[42]					1.4
Our calculation ($\gamma = V\beta B_S/C_P$)					2.1
Present (elastic moduli)	39.8	25.0	73.2	138	2.3

TABLE V. The Debye temperatures (computed from the zero-temperature elastic moduli[a] and V_a^0), Θ_D^T (computed from the measured elastic moduli and V_a^T at high temperatures), and the Grüneisen parameters are compared for the different phases.[20,22,24,89]

Phase	V_a^0 (Å^3)	Θ_D (K)	V_a^T (Å^3)	Θ_D^T (K)	γ
α	19.4	207	20.3 (380 K)	175 (380 K)	5.2 (380 K)
β	21.2 [b]	138	22.3 (450 K)	116 (450 K)	2.3 (450 K)
γ	22.3 [b]	140	23.3 (550 K)	109 (550 K)	1.9 (550 K)
δ[c]	24.5	115	24.7 (650 K)	91 (650 K)	3.8 (480 K)

[a]The values are shown in Table VI.
[b]Estimated value for V_a at 0 K.
[c]δ-Pu 2.36 at. % Ga was used. Θ_D^T was derived by extrapolating the elastic modulus values at 650 K at which the δ-phase of pure Pu exists. The values for δ-Pu 2 at. % Ga in Ref. 81 were used for V_a^0 and V_a at 650 K.

where β is the isobaric volumetric thermal expansion coefficient, γ is the Grüneisen parameter (we hope that context will sort out the double meaning of β and γ in this paper), C_P is the heat capacity at constant pressure, V is the volume, and B_S is the adiabatic elastic modulus. We assumed γ is constant in temperature and $C_P \approx C_V$ where C_V is the heat capacity at constant volume. We used the Debye model for C_V with the Debye temperature computed from Eq.(5). For B_S, we used Eq.(2) with our measured bulk modulus.

B^0, G^0, V_a^0 and Θ_D for β-Pu (and also γ-Pu) were determined numerically using Eqs. (3)–(7) (the Appendix). We obtained $\Theta_D = 138$ K and $V_a^0 = 21.2$ Å^3 for β-Pu ($\Theta_D = 140$ K and $V_a^0 = 22.3$ Å^3 for γ-Pu). The zero-temperature elastic moduli are compared to the values derived by Kay *et al.* using linear extrapolation to 60 K of data reported by Kay and Linford (1960)[53,88] (see Table IV). Table V shows comparison to the other phases. We also obtain $\Theta_D^T = 109$ K for β-Pu using the elastic moduli and V_a at 450 K. We consider Θ_D^T for comparison especially because α-Pu and δ-Pu 2.36 at. % Ga have strong temperature dependences, i.e., large anharmonic effects.

At higher temperatures, a Grüneisen-Einstein model yields the following expression:[84]

$$\frac{\partial B}{\partial T} = -\frac{3k\gamma(\gamma+1)}{V_a^0}. \tag{8}$$

The Grüneisen parameter, the quintessential measure of anharmonicity, for β-Pu was determined to be $\gamma = 2.3$. The anharmonicities of β-Pu and γ-Pu are more like metals, such as copper ($\gamma = 2.0$), while α-Pu and δ-Pu 2.36 at. % Ga have relatively larger values. Wallace estimated γ for β-Pu to be much smaller using Eq. (7) (see Table IV).[42] With $\Theta_D^T = 116$ K, we calculated the molar C_V of the phonon contribution in the Debye model as 24.9 J K^{-1} mol^{-1} at 450 K. Using $C_P - C_V = N_A V_a T\beta^2 B_T$ and $C_P/C_V = B_T/B_S$,[90] we have $C_P = 29.4$ J K^{-1} mol^{-1} from $C_P = C_V^2/(C_V - N_A V_a T\beta^2 B_S)$ with $\beta = 1.37 \times 10^{-4}$ K^{-1} and $V_a = 22.3$ Å^3, while Oetting and Adams[41] measured $C_P = 33.3$ J K^{-1} mol^{-1}. Their large measured C_P value was attributed to the conduction electrons, anharmonic lattice vibrations, nuclear characteristics, and magnetic properties.[41] We calculated $\gamma = 2.1$ using our measured B_S and calculated C_P in (7).

When considering interatomic bonding, Köster and Franz[91] emphasized that Poisson's ratio, ν is the most relevant elastic parameter. For polycrystalline samples,

$$\nu = \frac{3B - 2G}{2(3B + G)}. \tag{9}$$

In Fig. 5, Poisson's ratios for the three phases of pure Pu as well as δ-Pu 2.36 at. % Ga are plotted as a function of temperature. Poisson's ratio for most materials falls in the range of 0.25–0.35.[99] A low Poisson's ratio reflects a high ratio of bond-bending stiffness to bond-stretching stiffness and a high shear-stiffness to bulk-modulus ratio. Note that Poisson's ratio on warming (Köster and Franz[91]) should increase toward 0.5, characteristic of the liquid state. The Poisson ratios of aluminum,[92,93] austenitic steel,[94] α-Fe,[95] and diamond[96,97] are plotted for comparison.[98] The values and temperature dependence of Poisson's ratios for β-Pu and

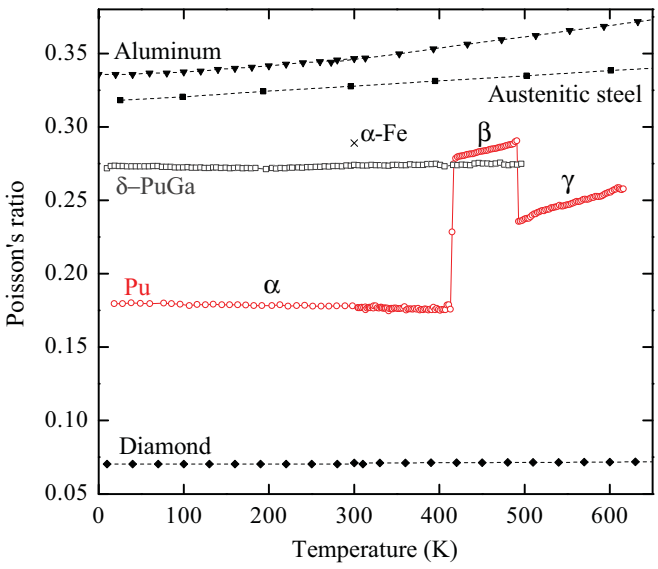

FIG. 5. (Color online) Poisson's ratio calculated from measured bulk and shear moduli is plotted. Poisson's ratio for δ-Pu 2.36 at. % Ga is also plotted. The Poisson ratios of aluminum,[92,93] austenitic steel,[94] α-Fe,[95] and diamond[96,97] are plotted for comparison.[98] α-Pu, β-Pu, and γ-Pu (and δ-Pu 2.36 at. % Ga) have large differences in their Poisson's ratios and temperature dependences as if they are completely different metals.

TABLE VI. The zero-temperature elastic moduli and the zero-temperature atomic volume for four phases of Pu are compared with other results.

	Phase	Method	B^0 (GPa)	G^0 (GPa)	C_L^0 (GPa)	V_a^0 (Å^3)
Theory	α	Robert (nonspin polarized)[100]	169.2			18.10
		Robert (spin-polarized AFM)[100]	101.1			18.47
		Kutepov (NM)[101]	175.4			18
		Kutepov (AFM)[101]	119			18
		Söderlind (SO)[39]	30.6	49.9	97.1	19.0
		Söderlind (SO + OP)[39]	34.4	51.3	102.8	20.3
Experiment		Kay (ultrasonic resonance)[88]	70	50	136	
		Our result (RUS)[21]	72.0	58.6	150.2	
		(X-ray,[13] dilatometry[102–105])				19.4
Theory	β	Söderlind (SO)[39]	36.0	26.2	70.9	22.0
		Söderlind (SO + OP)[39]	38.5	25.3	72.2	23.1
Experiment		Kay (ultrasonic resonance)[88]	46.2	24	78	
		Our result (RUS)	39.8	25.0	73.2	
		Our calculation[a]				21.2
Theory	γ	Robert (nonspin polarized)[100]	129.2			18.20
		Robert (spin-polarized AFM I) [100]	35.2			22.14
		Robert (spin-polarized AFM II)[100]	44.4			21.90
		Söderlind (SO)[39]	36.5	26.4	71.7	22.7
		Söderlind (SO + OP)[39]	34.6	22.2	64.2	23.8
Experiment		Kay (ultrasonic resonance)[88]	40.6	24	73	
		Our result (RUS)	30.3	25.6	64.4	
		Our calculation[a]				22.3
Theory	δ	Robert (nonspin polarized)[100]	99.9			19.57
		Robert (spin-polarized AFM)[100]	54.8			23.43
		Kutepov (NM)[101]	90.7			20
		Kutepov (AFM)[101]	51.0			24
		Shick (LSDA)[106]	76.1			20
		Shick (FLL LSDA + U)[106]	67.5			28
		Shick (AMF LSDA + U)[106]	31.4			27
		Söderlind (SO)[39]	39.0	27.8	76.0	24.2
		Söderlind (SO + OP)[39]	41.0	30.6	81.8	24.9
		Söderlind (EMTO)[39]	39.6	42.3	96.0	25.5
Experiment		Kay (ultrasonic resonance)[88]	51	19	77	
		Our result (RUS)[b],[24]	37.8	20.2	64.7	
		Lawson[c],[81]				24.8

[a][x-ray,[14,15] dilatometry, RUS].
[b]δ-Pu 2.36 at. % Ga was used.
[c](Neutron diffraction[107]) calculated value for nonalloyed δ-Pu based on the Invar model.

γ-Pu are similar to that of many metals, while α-Pu has small values and almost no (slightly negative) temperature dependence, which is more characteristic of covalent bonding. δ-Pu 2.36 at. % Ga has a value near Fe but a flat temperature dependence. Overall, α-Pu, β-Pu, and γ-Pu (and δ-Pu 2.36 at. % Ga) have large differences in their values and temperature dependences as if they are completely different metals.

In Table VI, zero-temperature elastic moduli and the zero-temperature atomic volume for the four phases are compared

TABLE VII. Calculated bulk moduli with thermodynamic effects are compared with our measurements.[20,22,24]

Method	α (294 K)	β (388 K)	γ (508 K)	δ (593 K)	δ (650 K)
Baskes (EAM)[40]	41	31	23	25	
Wang (CMF)[111]					43.0
Our result (RUS)	56.2	34.7 [a]	25.6	21.5 [a,b]	19.9[a,b]

[a]The extrapolated values.
[b]δ-Pu 2.36 at. % Ga was used.

TABLE VIII. $T_1, C_{L1}, C'_{L1}, G_1, G'_1, B_1, B'_1$, and β for β-Pu and γ-Pu (and α-Pu for testing), which we used for Eqs. (A2), (A3), and (A7). $C_{L1}, C'_{L1}, G_1, G'_1, B_1$, and B'_1 were extracted from our measurements for $T_1 \pm 20$ (± 10 for α-Pu) K. $\beta(T_1)$ were extracted from Ref. 112, which summarizes the best previous thermal expansion and x-ray measurements of pure Pu.

Phase	T_1 (K)	C_{L1}(GPa)	C'_{L1}(GPa/K)	G_1 (GPa)	G'_1 (GPa/K)	B_1 (GPa)	B'_1(GPa/K)	$\beta \times 10^{-4}$ (K^{-1})
β-Pu	450	56.6	−0.0416	17.1	−0.0199	33.8	−0.0151	1.37
γ-Pu	550	45.2	−0.0386	15.2	−0.0210	25.0	−0.0106	1.04
α-Pu	380	105.3	−0.1416	41.4	−0.0545	50.1	−0.0689	1.92

with theory. Magnetism[108–110] seems to play an important role in theoretical treatments of Pu, but magnetic effects have never been observed. Söderlind *et al.* capture most aspects of the measured behavior while underestimating the bulk modulus of α-Pu.[39] The *ab initio* electronic structure treatments of Pu are, at their core, zero-temperature ones. The unusually low elastic moduli observed here require that entropic considerations are essential in understanding the phases of Pu, especially in light of the very low elastic moduli. Such effects are absent in most *ab initio* models. In Table VII, theoretical predictions for the bulk modulus are compared with our data for the four phases. The calculated values by the atomistic model[40] are in good agreement with our data.

IV. CONCLUSIONS

We have reported accurate elastic moduli of pure polycrystalline β-plutonium for its entire existence temperature range at ambient pressure. The elastic response of pure β-Pu, when compared to other phases of Pu and to other metals, displays many bizarre features including an increase (albeit small) in the shear modulus on warming from β-to γ-phase and Poisson's ratios that differ strongly among the three phases explored here. These measured properties support the conjecture that Pu lies on a knife edge of stability caused by the 5*f* electrons.

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APPENDIX

Using measured high-temperature elastic moduli, we estimated the zero-temperature elastic moduli, the zero-temperature atomic volume, and the Debye temperature with the most accurate method we are aware of. From the high-temperature measurements where the elastic moduli are linear in temperature [Eq. (2)], using three measured values: T_1, C_1, and C'_1 [where $C_1 = C(T_1)$ is the elastic modulus and $C'_1 = dC(T)/dT|_{T=T_1}$], from Eq. (4), the zero-temperature elastic modulus can be estimated:

$$C^0 = C_1 + C'_1\left(\frac{3}{8}\Theta_D - T_1\right). \tag{A1}$$

From Eqs.(5) and (6),

$$\Theta_D = \left(\frac{9}{4\pi}\right)^{1/3}\frac{h}{k}\sqrt{\frac{N_A}{M}}V_a^{1/6}\left\{\left[C_{L_1} + C'_{L_1}\left(\frac{3}{8}\Theta_D - T_1\right)\right]^{-3/2} + 2\left[G_1 + G'_1\left(\frac{3}{8}\Theta_D - T_1\right)\right]^{-3/2}\right\}^{-1/3}. \tag{A2}$$

Table VIII lists the values of $C_{L_1}, C'_{L_1}, G_1, G'_1$, and T_1 for β-Pu and γ-Pu that we used. The right-hand side of Eq. (A2) was calculated numerically starting with $\Theta_D = 100$ K and iterated.

Equation (A2) is a very slowly varying function of V_a. A 5% error in V_a yields a less than 1% error in Θ_D. The temperature dependence of V_a for β-Pu and γ-Pu in Fig. 1 suggests that V_a at 0 K should be within 5% of that at higher temperatures so that V_a at high temperatures can be used safely to estimate Θ_D. This estimation of the zero-temperature V_a is most useful for comparison with first-principles calculations.

To estimate V_a at 0 K, again, we took three values from the thermal expansion measurement: T_1, V_1, and V'_1 [where $V_1 = V(T_1)$ is the volume and $V'_1 = dV(T)/dT|_{T=T_1}$]. Similar to what we did for elastic moduli, we need to find the ratio of

TABLE IX. The solutions for ε for different δ and n values using Eqs. (A3)–(A7).

δ	$n=1$	2	3	4	5	6	7	8	9	10
0.01	0.295	0.348	0.391	0.441	0.502	0.576	0.664	0.765	0.881	1.012
0.02	0.298	0.367	0.440	0.534	0.655	0.806	0.986	1.20	1.44	1.72
0.05	0.308	0.425	0.593	0.835	1.16	1.58	2.11	2.76	3.54	4.48
0.075	0.316	0.476	0.731	1.11	1.65	2.38	3.33	4.59	6.28	8.78
0.10	0.324	0.529	0.879	1.43	2.24	3.40	5.13			
0.15	0.342	0.642	1.22	2.22	4.04					
0.20	0.360	0.767	1.64	3.60						

TABLE X. The test for our method of calculation for extracting zero-temperature parameters using measured data[a] at 380 ± 10 K for α-Pu. The extracted values are compared to the measured values[20] at low temperatures.

Method	B^0 (GPa)	G^0 (GPa)	C_L^0 (GPa)	Θ_D (K)	V_a^0 (Å^3)
Measurement	72.0	58.6	150.2	207	19.4
Our calculation	71.0	57.9	148.2	205	19.4

[a]The values are shown in Table VIII.

temperature to Debye temperature ε and then to extrapolate to find V^0,

$$V^0 = V_1 + V_1'(\varepsilon\Theta_D - T_1). \tag{A3}$$

From Eq. (7),

$$\beta(T) = \frac{1}{V(T)}\frac{dV(T)}{dT} = \frac{\gamma C_P(T)}{V(T)B_S(T)}. \tag{A4}$$

We approximated $C_P \approx C_V$ (or $B_T \approx B_S$) because we measured B_S, but with measurements of B_T, the approximation is not necessary because $\beta = \gamma C_P/VB_S = \gamma C_V/VB_T$. The volume at T_1 can be calculated as

$$V(T_1) = \int_0^{T_1} \frac{\gamma C_V(T)}{B(T)}dT + V(0). \tag{A5}$$

If we assume that the temperature dependence of $B(T)$ is negligible and use the Debye model for $C_V(T)$, we have $\varepsilon = 0.375$ for $T_1 \gg \Theta_D$. However, the contribution from softening of $B(T)$ to $V(T)$ is rather large and increases the values of ε away from 0.375 at high temperatures.

From Eqs.(2) to (4),

$$B(T) = B^0\left(1 - \frac{\delta}{\exp(3\Theta_D/4T) - 1}\right), \tag{A6}$$

where

$$\delta = \frac{s}{B^0} = -\frac{3\Theta_D B_1'}{4B^0} = -\frac{3\Theta_D B_1'}{4\left[B_1 + B_1'\left(\frac{3\Theta_D}{8} - T_1\right)\right]}. \tag{A7}$$

δ is ~0.05–0.1 for typical metals as well as for β-Pu (0.039) and γ-Pu (0.037). $V'(T)$ also never converges at $T \gg \Theta_D$. Therefore, ε is a function of δ and n where $n = T_1/\Theta_D$. It can be solved numerically for ε for known values of δ and n. The solutions of ε for different δ and n are displayed in Table IX. For convenience, the values for ε can be approximated within a few percent error for $n > 1.5$ by the function

$$\begin{aligned}\varepsilon \approx\ & 0.311 + 0.0128n - 5.68\times10^{-3}n^2 + 5.28\times10^{-4}n^3 \\ & - (2.01 + 0.119n - 1.18n^2 + 0.0530n^3)\delta \\ & - (35.4 - 63.6n + 24.9n^2 - 1.85n^3)\delta^2 \\ & + (131 - 163n + 25.5n^2 + 7.82n^3)\delta^3.\end{aligned} \tag{A8}$$

The zero-temperature atomic volume is

$$\begin{aligned}V_a^0 &= V_a^T\frac{V^0}{V_1} = V_a^T\left(1 + \frac{V_1'(\varepsilon\Theta_D - T_1)}{V_1}\right) \\ &= V_a^T[1 + \beta(T_1)(\varepsilon\Theta_D - T_1)],\end{aligned} \tag{A9}$$

where V_a^T is the atomic volume at T_1 obtained from the measurement (see Table V) and $\beta(T_1) = V_1'/V_1$ V_a in Eq. (A2) can be replaced by V_a^0 in Eq. (A9) and more accurate values for C_L^0, G^0, V_a^0, and Θ_D can be obtained by iterating Eq. (A2).

This method was tested on α-Pu. Our elastic modulus measurement and the thermal-expansion measurement[112] for 380±10 K were used (see Table VIII) to extract the zero-temperature parameters. $\delta = 0.150$ for α-Pu, and it is much larger than for β-Pu and γ-Pu. Table X contains the comparison to measured values. At $T_1 = 380$ K, $n = 1.85$, and it is at the lower limit for Eq. (3) to work. Still, this method produced excellent predictions for zero-temperature parameters using the measured values at 380 K in the 20-K window.

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was performed using the average density of the α- and β-phases. Please note that this uncertainty in density gives only $\pm 2\%$ errors in elastic moduli, and Poisson's ratio is not affected by the density change.

[76]The two lowest resonances were below this frequency range. They were recorded for a few temperatures for which we used a larger frequency range. For them, the value difference in the elastic moduli, which were derived with the two lowest resonances (30 resonances total) or without them (28 resonances total) were negligible. Therefore, the elastic moduli for all temperatures were obtained without the lowest two resonances.

[77]The codes used for the calculation are available at [http://www.magnet.fsu.edu/inhouseresearch/rus/index.html (date last viewed 01/28/11)].

[78]The RUS fit program in Ref 77 also has the capacity to fit the sample dimensions. For a consistency check, four parameters (two elastic moduli and two relative dimensions) were fit. The result showed the negligible change in elastic moduli and dimensions, and the rms errors were slightly smaller.

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