Time-Differential–Perturbed-Angular-Correlation Measurement of the Hyperfine Field at Hg in Iron

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The hyperfine magnetic field at Hg nuclei in iron has been measured using the method of time-differential perturbed angular correlations on the 158-keV state of ^{199}Hg. The parent activity of ^{199}Tl was implanted into pure-iron foils employing an isotope separator. The results are consistent with about 80% of the nuclei experiencing a hyperfine field of $|H_{int}|=670\pm65$ kG and the remainder a field of $|H'_{int}|=455\pm85$ kG. These results, combined with previous lattice-location studies of Tl in Fe, indicate that the high field corresponds to the impurity atoms at substitutional sites. Using the value of H_{int} for the substitional sites derived here, the g factors of the first excited 2^+ states of the even-even nuclei $^{198-204}$Hg, obtained from previously reported experiments, have been recomputed.

I. INTRODUCTION

The hyperfine magnetic field at Hg in Fe has been measured by three groups,[1-3] all of whom employed the technique of integral perturbed angular correlations (IPAC) on the first excited 2^+ state of ^{198}Hg. Sources of ^{198}Au dissolved metallurgically in Fe were used in these experiments. The results are consistent with each other and typically yield a field of[3] $H_{int} = -440 \pm 105$ kG for Hg in Fe. The accuracy of these results has been limited by the imprecisely known g factor[4] of the ^{198}Hg level and the inherently limited precision of the IPAC method. The present work was undertaken to improve the accuracy of H_{int} by redressing both drawbacks. The time-differential PAC method was applied to the 158-keV level ($T_{1/2}=2.33$ nsec) of ^{199}Hg, the g factor of which has been measured with better accuracy than that of the ^{198}Hg level.[5] A more precise knowledge of H_{int} would be desirable in two respects: First, one can define more reliably the steep variation of H_{int} from a large negative value at Au to a moderately large positive value at Pb in Fe. Second, it may also increase the precision of the g factors of even-even Hg isotopes[6] measured by the recoil-implantation technique, using Fe as the ferromagnetic matrix.

II. EXPERIMENTAL DETAILS

A. Source

The parent activity of ^{199}Tl (7.4 h) was prepared by the ^{197}Au $(\alpha, 2n)^{199}$Tl reaction using the Princeton University cyclotron. Using an isotope separator, the ^{199}Tl activity was then simultaneously separated and implanted into thin, pure-Fe foils which were electropolished a few minutes before the implantation. This method of introducing the Tl activity

into Fe was necessary because of the widely different melting points of Tl and Fe, which makes metallurgical methods very difficult. In all, three sources were prepared for these measurements. The implantation energy was 60 keV for the first and was increased to 90 keV by post acceleration for subsequent sources. After implantation the foils were gently wiped to remove small amounts of surface activity and then used for the PAC measurements. The source strengths were typically 100 μCi.

B. PAC Apparatus

The experimental setup consisted of three NaI(Tl) detectors, two fixed at 90° to each other, each detecting the 158-keV γ ray (see, e.g., Ref. 5 for details of the decay scheme of ^{199}Tl), and the third movable detector alternating between 90° and 180° with respect to one of the fixed detectors. The movable detector was gated on the 334-keV γ ray. The 158–334-keV delayed coincidences for the two detector combinations were separately recorded for each of the two positions of the movable detector (90° and 180°) by means of suitable electronic routing. Standard slow-fast coincidence-counting electronics with a time-to-amplitude converter were employed. The system time resolution under experimental conditions was 2 nsec. No external polarizing field was applied to the source. The source foils were checked for random orientation of the magnetic domains by means of a Plastiform magnetic viewer,[7] a cell containing a colloidal suspension of magnetic particles.

III. DATA ANALYSIS AND RESULTS

The time-differential γ-γ angular correlation function perturbed by a randomly oriented magnetic interaction is given by

$$W(\theta, t) = 1 + A_{22}G_{22}(t)P_2(\cos\theta) \quad (A_{44} \approx 0), \qquad (1)$$

where the perturbation factor has been shown to be[8]

$$G_{22}(t) = \tfrac{1}{5}(1 + 2\cos\omega t + 2\cos 2\omega t). \qquad (2)$$

θ is the angle between emission directions of the γ rays, ω is the Larmor precession frequency, and A_{22} is the angular correlation coefficient, corrected for the solid angle subtended by the detectors at the source. In practice, from the experimental data, one computes the ratio

$$R^{\text{expt}}(t) = \frac{N(t, 180°) - N(t, 90°)}{N(t, 180°) + 2N(t, 90°) - 3C}, \qquad (3)$$

where $N(t, \theta)$ represents the coincidence-counting rate observed at time t and C is the chance coincidence background. Equation (3) can be shown to reduce to

$$R(t) = \tfrac{1}{2}A_{22}G_{22}(t), \qquad (4)$$

from which ω must be extracted. Experimentally, the observed PAC spectrum (3) does not resemble (4), due to the fact that the relative amplitudes of the two frequencies in (2) are changed by the finite resolving time of the apparatus. Thus the theoretical function (2) has to be folded with the prompt-time-resolution function before it can be fitted to the data. To accomplish this, we used the function

$$F(t) = \frac{\int P(t-t')e^{-t'/\tau}\left[\tfrac{1}{2}A_{22}G_{22}(t')\right]dt'}{\int P(t-t')e^{-t'/\tau}dt'}. \qquad (5)$$

The time-response function P was obtained experimentally by measuring the prompt-coincidence spectrum using a ^{60}Co source. τ is the mean life of the 158-keV level.

Fits such as described above were made to all of the individual runs from the three sources; no significant differences were detected between the cases of 60- and 90-keV implantation energy. The data were then added together and the final fit yielded a Larmor precession frequency of

$$\omega = (1300 \pm 60) \times 10^6 \text{ rad/sec.}$$

With a g factor of $g = 0.413 \pm 0.032$,[5] this leads to a field

$$|H_{\text{int}}| = 660 \pm 60 \text{ kG.} \qquad (6)$$

As an independent method of analyzing the data, the power spectrum of the Fourier transform of the function $R^{\text{expt}}(t)$ was computed. A single-valued field implied by result (6) would be reflected as two peaks in the power spectrum corresponding to the ω and 2ω components in $R(t)$ whose intensities are modified appropriately by the instrumental time resolution. Such an analysis for the present data, however (see Fig. 1), revealed four peaks, two corresponding approximately to a field of 600 kG and

two others to a field of about 450 kG. In order to obtain these values more accurately, the least-squares routine was altered by replacing the fitting function (5) by the function

$$A(t) = f[F_1(t)] + (1 - f)[F_2(t)] + K, \qquad (7)$$

where $F_1(t)$ corresponds to a Larmor frequency ω_1, $F_2(t)$ corresponds to ω_2, f is the relative fraction of the two field sites, and K is a constant. The results of this fit (see Fig. 2) were

$$\omega_1 = (1320 \pm 70) \times 10^6 \text{ rad/sec,}$$

$$\omega_2 = (895 \pm 150) \times 10^6 \text{ rad/sec,}$$

$$f = 0.77 \pm 0.08,$$

and

$$K = -0.012 \pm 0.010.$$

These frequencies correspond to the hyperfine field values

$$|H_{\text{int}}| = 670 \pm 65 \text{ kG,} \quad |H'_{\text{int}}| = 455 \pm 85 \text{ kG,} \qquad (8)$$

the ratio of atoms in these two sites being $4:1$. The quality of the fit with (7) was slightly better than that with (5).

In addition to yielding the frequencies, the fit gives information on the amplitudes, or the effective anisotropy observed, which have to be satisfactorily accounted for. The fit gives $A_{22}(\text{expt}) = -0.23 \pm 0.02$ and $K = -0.012 \pm 0.010$. The theoretical correlation coefficient is $A_{22}(\text{theor}) = -0.42 \pm 0.02$.[5] The correction for finite solid angles reduces this to $A'_{22}(\text{theor}) = -0.330 \pm 0.016$, which should then in principle be equal to $A_{22}(\text{expt})$. The discrepancy which is observed could arise from two sources.

(a) Obscure geometrical corrections, such as scattering in the foil, and the finite size of the source could reduce $A'_{22}(\text{theor})$ further. In our estimation however, this effect is not sufficient to account for the discrepancy.

(b) A certain fraction of the implanted atoms end up in a variety of irregular sites, experiencing a wide range of magnetic and/or quadrupolar precessions which contribute effectively only the hard-core anisotropy. The theoretical hard-core value is $\tfrac{1}{5}A'_{22}(\text{theor}) = -0.066$. Thus, as many as 20% of the implanted atoms may come to rest in these irregular sites (either in a surface oxide or at damage sites in the Fe lattice), contributing a hard-core anisotropy reflected by the value of K obtained in the fitting procedure.

The results of the Fourier and least-squares analysis of the data can thus be summarized as follows.

(i) Up to 20% of the implanted atoms are trapped in irregular sites, producing a small time-independent contribution to the observed anisotropy.
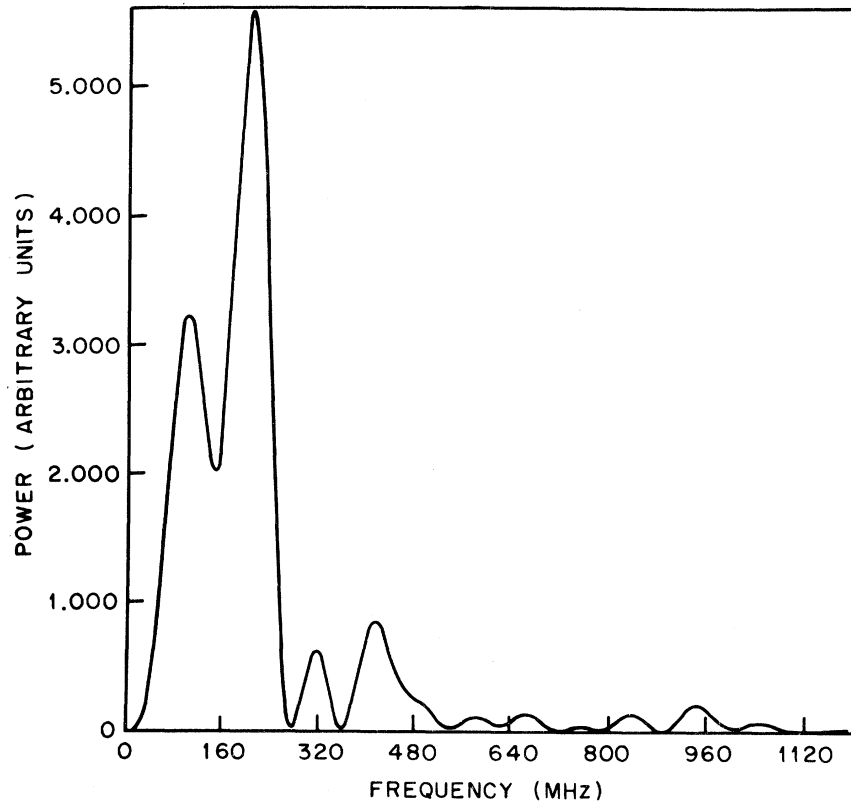
FIG. 1. Power spectrum of the Fourier transform of the function $R(t)$.

(ii) The remaining atoms are located in two distinct hyperfine field sites, 80% experiencing a field of $|H_{int}| = 670 \pm 65$ kG and 20% a field of $|H'_{int}|$ $= 455 \pm 85$ kG.

IV. DISCUSSION

A. Hyperfine Field of Hg in Fe

From the point of view of the systematics of hyperfine fields at impurity solute atoms in Fe, the first objective is to decide which of the two field sites observed in the present work is associated with Hg atoms at substitutional sites in Fe. Extremely relevant to this question is the recent work of Feldman *et al.*[9] who performed lattice-location studies[10] on Tl atoms implanted in Fe by means of an isotope separator. They concluded that as much as 85% of the implanted Tl atoms were located at substitutional sites. In our experiment, also performed by implanting Tl in Fe, 80% of the atoms displaying a distinct Larmor frequency are at the high-field site. Since recoil effects in the electron-capture decay of ^{199}Tl to ^{199}Hg are negligible, it seems fairly conclusive that the Hg atoms at the high-field site are substitutional. The hyperfine field of Hg in Fe is therefore $|H_{int}| = 670 \pm 65$ kG.

The occurrence of a small fraction of low-field sites in the implanted source with $|H'_{int}| = 455$ kG,

although puzzling, is not a cause for serious concern, since more than one field site has been observed in several cases where the probe atom was energetically propelled into Fe. It is possible that this field is associated with a Hg atom systematically located with a vacancy in its immediate neighborhood, and is a consequence only of the implantation process. Other reasons can also be proposed, but all these speculations are in general difficult to prove experimentally.

Several theoretical descriptions of the origin of hyperfine magnetic fields at impurities in ferromagnetic media have appeared in the literature.[11-15] Systematic trends have been well reproduced, but accurate predictions in specific cases have not been possible, because the fields result, in general, from detailed cancellation of the relatively large contributions from both conduction-electron and core polarization. It is instructive, however, to compare our result with the phenomenological theory of Balabanov and Delyagin,[14] which predicts a value of -735 kG for Hg and Fe. This is quite consistent with the present result, and much more so than the IPAC result of -440 ± 105 kG or the value at the low-field site observed in the present work. The IPAC experimental value is discussed in more detail in Sec. IV B.

Very recently, H_{int} for Hg in Ni has been mea-

sured by Mahnke[16] in a time-differential PAC experiment. The value is $H_{int}(NiHg) = -124 \pm 12$ kG. The ratio of H_{int} for Hg in Fe and in Ni is thus 5.4, which is very different from the host-moment ratio of 3.7. It is interesting to note that in the isoelectronic case of Cd, the ratio is 5.7,[17] which is very similar to that of Hg.

B.   IPAC Value for H_{int} and g Factor of 2^+ Level in ^{198}Hg

The IPAC experiments[1-3] to determine the field of Hg in Fe were performed on the first excited 2^+ level of ^{198}Hg (mean life[6] $\tau = 32$ psec) populated in the β decay of ^{198}Au. In these measurements, small amounts of ^{198}Au (typically 0.2 at.%) were either diffused into or melted with Fe. All three groups observed, within experimental errors, the same integral precession; the most accurate result was that of Zawislak et $al.$,[3] who reported $\omega\tau$
$= \mu_N H_{int} g_{198} \tau/\hbar = -36.5 \pm 3.2$ mrad. This implies a field of $H_{int} = -440 \pm 105$ kG if the measured g factor[4] of $g_{198} = 0.55 \pm 0.11$ is used. In view of the consistency of the IPAC precession measurements,

it is disturbing that the value of H_{int} derived from them is in clear disagreement with the present result. The dilute solution of Au in Fe has been used in the past in a variety of experiments with uniformly satisfactory results. In particular, the Oxford group showed[18] that sources of ^{198}Au carefully implanted in Fe produced the same hyperfine field as seen in sources of ^{198}Au diffused in Fe. Later, Alexander et $al.$[19] showed by channeling studies that over 85% of the implanted Au atoms are located at substitutional sites. It is thus clear that in the diffused sources of Refs. 1-3 also, the observed precession refers to the overwhelming majority of ^{198}Au-^{198}Hg atoms at substitutional sites. Therefore, while accepting the precession value reported in Refs. 1-3, we suspect that the g factor used by them was in error. We notice that our value of H_{int} and the precession value of Refs. 1-3 would become compatible if the g factor of the 2^+ level in ^{198}Hg is scaled down to 0.36 ± 0.06. This value is in good agreement with the Z/A value of 0.4 predicted by the hydrodynamical model. The life-
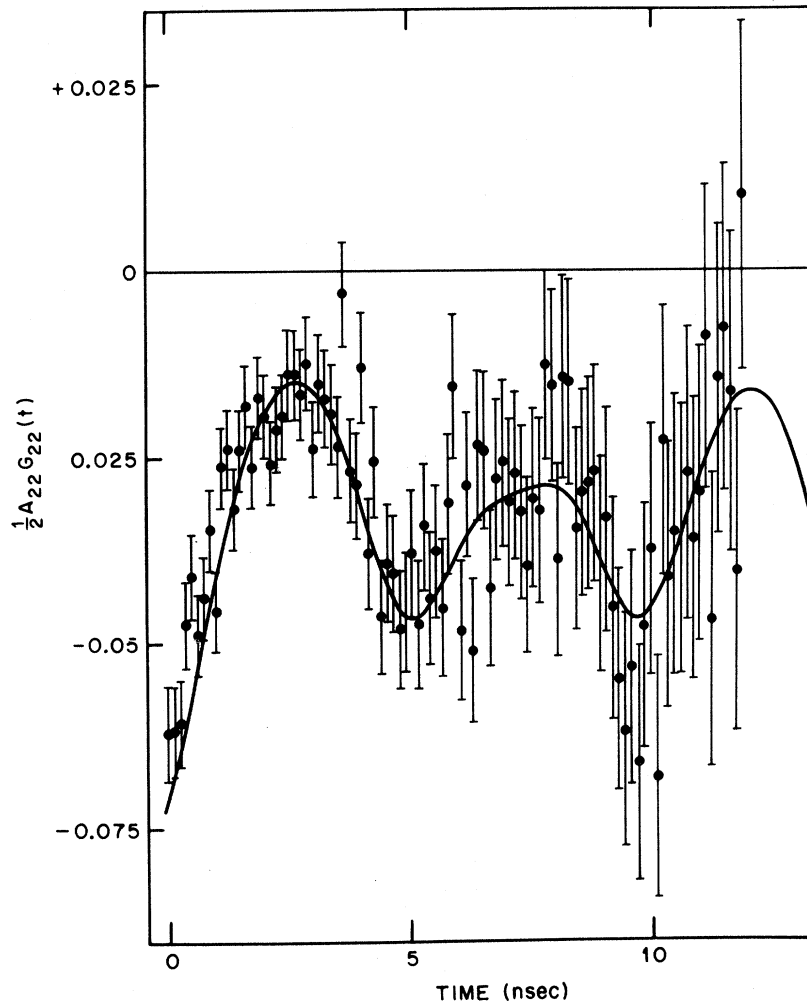

FIG. 2. Spin precession of the magnetic moment of the 158-keV levels in ^{199}Hg in the hyperfine field of Fe. The data points refer to the experimental ratio $R(t)$. The solid curve represents the least-squares fit to the function $A(t)$ [see Eqs. (5) and (7)].

time of the ^{198}Hg level is short and the application of an external field of 60 kG as was used in Ref. 4 produces only a small precession. It is therefore desirable to remeasure this number with an external field of ~100 kG or more, which is now possible in many laboratories through the availability of superconducting magnets.

Further support for the revision of g_{198} comes from Mahnke's recent result for H_{int} of Hg in Ni. The value of $H_{int}(Ni$Hg$)$ of -124 ± 12 kG from this work disagrees with the value derived by Zawislak *et al.*[3] using g_{198}, to the same extent as in the case of Hg in Fe. The discrepancy disappears when the revised value of g_{198} is used to evaluate the field from the precession observed by Zawislak *et al.*[3] Thus the value of $g_{198} = 0.36 \pm 0.06$ is independently confirmed in a satisfactory manner.

C. *g* Factors of Even-Even Hg Isotopes

Kalish *et al.*[6] have observed the Larmor precession of the first excited 2$^+$ states of the even-even nuclei ^{198}Hg-^{204}Hg by recoil implantation of these nuclei in Fe after Coulomb excitation (IMPAC). In these experiments, the major uncertainty in deriving the absolute *g* factors of these states is the presence of a large, transient magnetic field acting on the recoiling nuclei for very short times, the magnitude of which is difficult to evaluate. Therefore these workers obtained these *g* factors relative to that of the ^{198}Hg level, which eliminates the need to know the transient field. The crucial parameters they need are the *g* factor of the ^{198}Hg level and the integral precession of this level in Fe; both are determined from radioactivity measurements. For the former, they assumed $g_{198} = 0.55 \pm 0.11$ of Ref. 4, while for the latter they took the result of Refs. 1–3. We have pointed out in the preceding discussion the reliability of the

measured precession value and the need to revise the value of g_{198} to 0.36 ± 0.06. Using this new value of g_{198}, we have recomputed the *g* factors for the cases of ^{200,202,204}Hg from the experimental precession values of Kalish *et al.*[6] These, as given in Table I, are in better agreement with the trend predicted by the calculations of Kumar and Baranger.[20]

Note added in proof. The values of H_{int} for ^{197}Hg in Fe, Co, and Ni have been measured recently by a group at the University of Bonn (K. Krien, private communication) who obtain 692 ± 55, 483 ± 40, and 103 ± 8 kG, respectively. These values together with the precession results of Ref. 3 provide further support to the value $g_{198} = 0.36 \pm 0.06$. A. F. Dilmanian and R. Kalish (preprint) have performed a new IPAC measurement on the 2$^+$ level of ^{198}Hg in Fe. We thank Dr. Krien and Dr. Kalish for informing us of their work prior to publication.

ACKNOWLEDGMENTS

TABLE I. Revised values for the *g* factors of first excited 2$^+$ states of even-even nuclei ^{198}Hg-^{204}Hg.

A	Revised in present work	Kalish *et al.* (Ref. 6)
198	0.36 ± 0.06	0.55 ± 0.11[a]
200	0.28 ± 0.05	0.43 ± 0.11
202	0.385 ± 0.060	0.59 ± 0.15
204	0.26 ± 0.05	0.40 ± 0.10

[a]Value from Ref. 4.

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Electrical and Magnetic Properties of Some Cubic Intermetallic Compounds of Plutonium with Ru, Rh, Ir, Pd, and Pt[†]

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The electrical resistivities and magnetic susceptibilities of the cubic intermetallic compounds of the form PuX_2 (X =Ru, Rh, Ir, Pt) and PuX_3 (X =Rh, Pd, Pt) have been measured from 2 to 300 °K. The PuX_3 compounds order antiferromagnetically with Néel temperatures T_N of 6.6, 24, and 40 °K, respectively. The resistivities of the PuX_3 compounds decrease rapidly below T_N, and there is also an abrupt change in the slope of the resistivity-versus-temperature curve at T_N in $PuRh_3$. $PuPt_2$ orders ferromagnetically at a Curie temperature of $T_C = (6.0 \pm 0.3)$ °K, and the resistivity falls dramatically below T_C. $PuRh_2$ has a temperature-dependent susceptibility but there is no evidence for magnetic order, while the susceptibilites of $PuRu_2$ and $PuIr_2$ are temperature independent. However, the resistivities of $PuRh_2$, $PuRu_2$, and $PuIr_2$ have anomalous negative curvature at high temperatures, although the slopes remain positive throughout the temperature range investigated. The results are discussed in terms of the interatomic distance between plutonium atoms and the localized-spin-fluctuation model.

I. INTRODUCTION

A wealth of interesting magnetic and transport behavior has been observed recently in the actinide elements and their intermetallic compounds,[1] and the unusual nature of the phenomena in these systems is intimately related to how the actinide $5f$ electrons respond to a given environment. The results of Nellis and Brodsky[2] on $PuPd_3$ and $NpPd_3$ and of Hill and Elliott[3] on actinide-Pt compounds indicate that a study of actinide–transition-metal compounds should yield considerable information on the properties of actinide $5f$ electrons.

This paper presents the electrical resistivities and magnetic susceptibilities of the cubic intermetallic compounds of the form PuX_2 (X=Ru, Rh, Ir, Pt) and PuX_3 (X=Rh, Pd, Pt). These compounds were chosen because the crystal structures are relatively simple—the $MgCu_2$ cubic Laves phase and ordered fcc $AuCu_3$ structures, respectively[4]—and because the transition metals themselves are nonmagnetic. A preliminary account of the resistivity measurements has already been given.[5] A study of the magnetic properties has also been reported,[6] but new data on better samples of $PuRh_2$ and $PuPt_3$ are included in the present paper as a supplement to the previous investigation. The susceptibility measurements show that the PuX_3 compounds order antiferromagnetically, and this finding is supported by the resistivity results. The

PuX_2 compounds show a spectrum of magnetic behavior: $PuRu_2$ is strongly paramagnetic, $PuRh_2$ is nearly magnetic, and $PuPt_2$ is a weak ferromagnet. The results are discussed in terms of the interatomic distance between plutonium atoms and the localized-spin-fluctuation model.

II. EXPERIMENTAL PROCEDURE

The transition metals used in this study had purities which were typically ~99.995%. The high-purity plutonium was obtained from Los Alamos Scientific Laboratory and the impurities it contained were (in ppm by weight) Mg, 3; Al, 10; Si, 5; Ca, <3; Zn, <5; Am, 55 (on 2-18-71); Fe, 4; Ta, 11; Th, 9; W, 2.5; U, 25; N, 3; C, <10; H, 15; and all others, <2. From these starting materials, 1-g quantities of the intermetallic compounds were prepared by arc-melting appropriate amounts of the constituents in an argon-helium atmosphere. Resistivity samples with the approximate dimensions $1 \times 1 \times 7$ mm were machined out of the buttons with a spark-erosion apparatus, and magnetic-susceptibility samples were prepared from pieces of the remaining material. $PuPt_3$, $PuPd_3$, $PuRh_3$, and $PuRu_2$ formed congruently from the melt,[4] and consequently no further heat treatment was given. $PuPt_2$, $PuRh_2$, and $PuIr_2$ were given one-week anneals at 1100, 1250, and 1000 °C, respectively. Powder x-ray-diffraction techniques were used to confirm that the desired