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Isochronal annealing studies in Pu and Pu alloys using magnetic susceptibility

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Abstract

The isochronal annealing of the low temperature accumulated damage from the radioactive decay of plutonium in α-Pu, δ-$Pu_{1-x}Ga_x$ ($x = 0.043$) and δ-$Pu_{1-x}Am_x$ ($x = 0.224$) was characterized using magnetic susceptibility. In each specimen, thermal annealing, as tracked by magnetic susceptibility, only commenced when $T > 33$ K and the magnetic susceptibility changes due to defects were fully annealed at $T \sim 300$ K. The α-Pu magnetic susceptibility isochronal annealing data is similar to earlier measurements of resistivity characterized isochronal annealing. However, the δ-$Pu_{1-x}Ga_x$ ($x = 0.043$) magnetic susceptibility isochronal annealing data, when compared with similar resistivity data, indicates that for this alloy magnetic susceptibility studies are more sensitive to vacancies than to the interstitials accumulated at low temperatures. The $Pu_{1-x}Am_x$ ($x = 0.224$) alloy shows a remarkable change in properties, over a limited temperature range beginning where interstitial defects are first mobile, and characterized by an induced effective moment of order 1.1 μ_B/Pu. This transient behavior may be evidence for a disorder driven low temperature phase transition, perhaps indicative of a compositional and structural proximity to a state possessing significant magnetic moments.

Keywords: Magnetic measurements; Metals and alloys; Vacancy information

1. Introduction

The 5f-electrons of both α-Pu and δ-Pu occupy a narrow f-band as indicated by the enhanced values of both the electronic specific heat [1] and Pauli contribution (temperature independent) to the magnetic susceptibility [2]. While some theoretical models of Pu accurately predict the energy-volume relationship of the various Pu phases, they require non-vanishing magnetic moments. Yet, there are no experimental measurements indicating any sort of localized moments in the "pure" metal [3–5]. However the narrow f-band [3,6] suggests that Pu is nearly magnetic, and should display evidence of magnetic moments given an appropriate perturbation.

This idea is supported by the Hill conjecture which postulates, that within the actinides, magnetism, or its absence, is determined primarily by the actinide separation [7]. For plutonium compounds, Hill predicts that a separation greater than 3.4 Å should create local magnetic moments. Indeed, this is supported by compounds such as PuH_2, an isostructure of fluorite, where the Pu atoms retain an FCC structure akin to the δ-phase but the hydrogen atoms effectively act as spacers to stretch the Pu–Pu distance from 3.28 Å in pure δ-Pu to 3.79 Å in PuH_2. This leads to local moments, (∼1 μ_B/Pu), that antiferromagnetically order at 30 K [8]. Similarly, a Pu atom in isolation is also expected to have a local moment arising from the incomplete 5f shell, and one way to experimentally achieve this is to dissolve dilute amounts of Pu in a non-magnetic lattice such as Pd. Instead of forming the simple local moments of a Kondo impurity as was observed for dilute Np in Pd, Pu acts like a local spin fluctuation system with a characteristic temperature, $T_{sf} \sim 1$ K and an effective moment of 1 μ_B/Pu [2]. Similar evidence for magnetic behavior of isolated Pu is seen by its influence on the superconducting critical temperature, T_c, of a host material. Dilute doping of Pu into La sharply decreases T_c in contrast to doping Th, U, or Am into La which have a minimal impact on T_c [9]. This again implies a magnetic state for the Pu

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atoms when individual atoms are isolated within a non-magnetic lattice.

It is reasonable, in light of the foregoing discussion, to question what, if anything might be revealed by the introduction of disorder into a regular Pu or Pu-alloy lattice. A consequence of stabilizing the δ phase of Pu by alloying with other elements such as Am or Ga, is an increase in the disorder of the system. An elegant alternative method is to explore the influence of disorder on the magnetic properties of the metal or alloy by exploiting the radioactive properties of Pu that create self-damage in the lattice. The radioactive decay of ^{239}Pu (half life: 24,110 years) yields a 5 MeV alpha particle that results in several hundred isolated vacancy interstitial pairs. The corresponding 86 keV U recoil initiates a cascade of approximately 2500 vacancies and interstitials over a relatively small volume. Most of these cascade defects immediately recombine, but a few hundreds remain, and at sufficiently low temperatures ($T < 33$ K), they become frozen within the lattice. In other words, at low temperatures Pu continuously, but non-uniformly, dopes itself with vacancies and interstitials. As reported in earlier work [10], this is observable as a continuous increase in the magnetic susceptibility with time (t) that can be annealed at 350 K returning the specimen to its original $t = 0$ value. However, it does provide an avenue to explore how disorder perturbs the system, perhaps providing insight into how the organizational principles that determine the regular atomic lattice also act to eliminate magnetism in the 5f electrons of Pu.

This paper examines the isochronal annealing of the excess magnetic susceptibility (EMS) for α-Pu, and δ-$Pu_{1-x}Ga_x$ ($x = 0.043$) and compares this to corresponding isochronal annealing data characterized by resistivity. Magnetic susceptibility annealing data for $Pu_{1-x}Am_x$ ($x = 0.224$), an FCC δ-stabilized phase where the average Pu–Pu separation distance has been expanded approximately halfway to the Hill criterion, is compared to δ-$Pu_{1-x}Ga_x$ ($x = 0.043$).

2. Isochronal annealing measurement techniques

Radiation damage creates defects within a crystal lattice that can be observed indirectly through the measurement of any physical property that is changed by the damage. Isochronal annealing studies map the change in a physical property as the radiation damage is annealed away by soaking for a fixed time at progressively higher temperatures. The basic protocol is to accumulate damage at a low temperature and measure the property of interest at that temperature, T_M. Then rapidly warm the specimen to a higher temperature, soak for a fixed period of time, return to T_M, and repeat the measurement. This process is repeated for successively higher temperatures until the measurement returns to the initial undamaged value and it is assumed to be fully annealed. In radioactive specimens, where damage continues to accumulate while the isochronal annealing curve is measured, first order corrections to this additional damage can be included by repeating the identical protocol for the annealing curve on a fully annealed specimen and subtracting it from the resulting curve (see Ref. [11]).

Resistivity is the most commonly used physical property for this type of experiment because the measurement can be made rapidly with extremely high precision through careful design of the instrumentation and sample geometry. Resistivity is very sensitive to the total number of scattering centers including point defects (vacancies and interstitials) and their aggregates. Magnetic susceptibility is a thermodynamic and therefore bulk measurement, which may have different sensitivity to the defects that occur during the isochronal annealing process, and hence there is no *a priori* reason to expect that magnetic susceptibility would mirror the characterization of resistivity with respect to the population of defects in a particular specimen. However, defect created local moments that have been observed in the high temperature superconducting cuprates (HTSC) when Cu sites are replaced by a spin zero Zn impurity (a spin vacancy) [12] or by vacancies from radiation damage [13], so it is reasonable to speculate that radiation damage may influence the magnetic properties of some materials. The high sensitivity magnetic susceptibility measurements necessary to perform isochronal annealing experiments tend to be slower then corresponding resistivity studies. Additionally, at low temperature, magnetic impurities have strong temperature dependencies (Curie-like: $1/T$), so uncertainties in temperature are reflected much more strongly than for resistivity. Careful design of the experiment can overcome these impediments as are shown in this paper. One such consideration is the magnetic field applied to make the measurement, which should be sufficiently large to guarantee a repeatable value with a good signal-to-noise ratio. For these samples, an applied magnetic field of 3 T was used in all measurements providing a signal-to-noise ratio greater than one thousand.

In the present work, after annealing the specimen at 350 K for an hour, damage is accumulated for several weeks at 5 K, after which the specimen is annealed for a fixed time (isochronal) of 30 min at successively higher temperatures (T_A), with each anneal followed by a measurement of $\chi(T = 5\,\mathrm{K})$. The resulting $\chi(T = 5\,\mathrm{K})|_{T_A}$ values are normalized according to:

$$f_{\mathrm{damage}}(T_A) = \frac{\chi(T = 5\,\mathrm{K})|_{T_A} - \chi(T = 5\,\mathrm{K})|_{350\,\mathrm{K}}}{\chi(T = 5\,\mathrm{K})|_{5\,\mathrm{K}} - \chi(T = 5\,\mathrm{K})|_{350\,\mathrm{K}}} \quad (1)$$

and plotted as a function of annealing temperature, T_A, in Fig. 1 (red circles) for both α-Pu and δ-Pu (4.3 at% Ga), showing the fraction of damage-induced EMS retained after annealing at each temperature. For comparison, similar isochronal annealing studies from previously reported experiments [11,14] using resistance to characterize the annealing are also included (blue diamonds). The resistive annealing data for the α-Pu study were soaked at a low temperature for several months, by contrast the resistive data for the δ-Pu(Ga) study was damaged at low temperatures for only three days prior to the annealing experiment, and also contains a lower concentration of Ga (3.3 at% versus 4.3 at% for the corresponding magnetic susceptibility data).

3. Results

3.1. α-Plutonium

The qualitative features of the resistive and magnetic susceptibility α-Pu isochronal annealing curves in Fig. 2a are quite

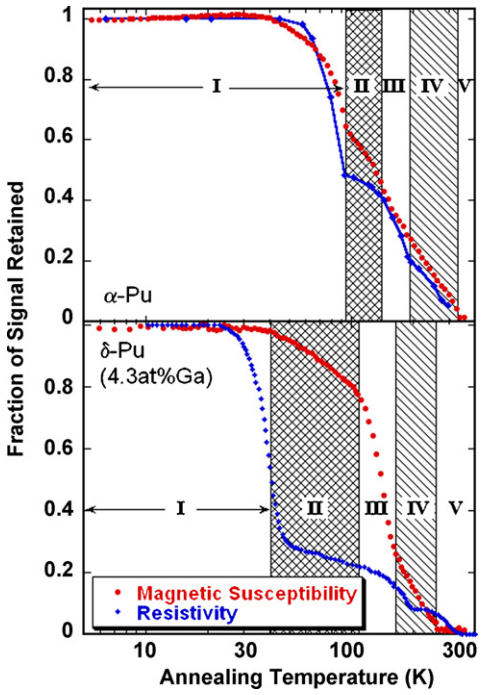

Fig. 1. Isochronal annealing curves for α-Pu and δ-Pu (4.3 at% Ga) determined by magnetic susceptibility (red). The resistivity curves (blue) are from references [14] and [11], respectively, on similar samples. The annealing stages identified on the plots are primarily based on the magnetic susceptibility data with the exception of Stage I in the δ-Pu specimen, which appears to be absent in the magnetic data (see text for details). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

similar, indicating that magnetic susceptibility is a useful technique to make isochronal annealing measurements. The low temperature transitions are commonly associated with interstitial motion and interstitial impurity interactions, while the higher temperature transitions are the result of vacancy motion, vacancy impurity interactions and vacancy cluster dissolution and annihilation [15].

For both the resistivity and magnetic susceptibility measurements, while $T < 40$ K, the signal is independent of temperature

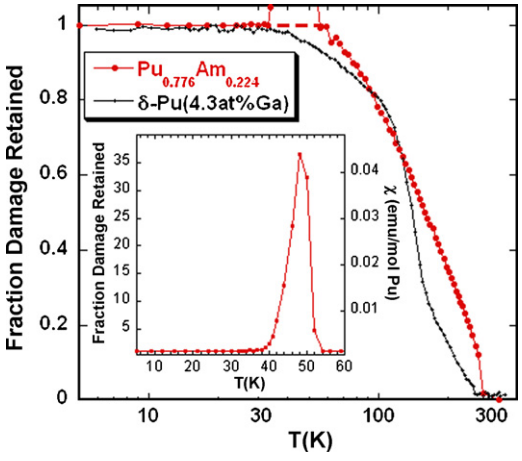

Fig. 2. Isochronal annealing curve for $Pu_{1-x}Am_x$ ($x = 0.224$) specimen aged 40 days. The black line shows the stabilized δ-Pu (4.3 at% Ga) specimen as a comparison. The inset shows the anomalous increase in signal relative to $\chi(T = 5\,K)$ before the onset of annealing.

thereby indicating little, if any change, in the structure or number of defects and thus the accumulated defect population is frozen in place. Above this temperature, the damage begins to anneal away with a nearly 40% decrease for the magnetic susceptibility data, and greater than 50% decrease for the resistivity. This is a Stage-I-like annealing, where the interstitials become mobile, and migrate through the lattice until they annihilate by interacting with a vacancy, grain-boundary, or surface; or they become trapped at a dislocation or impurity. The next region, between 90 K and 130 K is Stage-II-like annealing, where interstitials escape from trapping centers and small interstitial clusters merge into larger clusters. Here, the resistivity is relatively flat, indicating there is little change in the total number of scattering centers, while the magnetic susceptibility continues to decrease suggesting that some de-trapped interstitials are annihilating with vacancies. Moving to temperatures above ~130 K, the slope increases again as Stage III where the vacancies become mobile, and in Stage IV form clusters, loops and other aggregates before eventually dissolving away in Stage V. There is a change of slope in the resistivity curve just below 200 K possibly indicating Stage IV, but there is no obvious equivalent in the magnetic susceptibility curve, only a slight change in the slope. Finally, near 300 K, the distinct change in slope of the magnetic susceptibility data shows Stage V annealing where the remaining damage signal disappears as the vacancy aggregates dissolves away, diffuses, and annihilates, returning the sample to an annealed or undamaged state, apart from the small numbers of U, and He atoms created from the decay process, which are apparently not detectable in this measurement.

The fact that the magnetic susceptibility and the resistivity curves appear to approximate each other indicates, that in the case of the α-Pu, the magnetic susceptibility response to interstitials relative to vacancies is comparable to the response in the resistivity curve. This is a surprising result as one might have expected that the appearance of an EMS would track more strongly with vacancies, because any concomitant localization of f-electrons should favor an open lattice. A possible explanation is that as the interstitials begin to move, a significant fraction annihilate with vacancies as compared to moving to grain boundaries. Alternatively, disorder may be more important than simply free volume in determining the magnetic properties of α-Pu.

3.2. δ-Plutonium

The magnetic susceptibility annealing data was obtained on a $\delta\text{-}Pu_{1-x}Ga_x$ ($x = 0.043$) electro-refined specimen alloyed less than two years prior to the reported measurements, and then annealed at 350 K for one hour just before the measurements began. The specimen was held at low temperatures for six weeks to accumulate sufficient damage so as to make the magnetic susceptibility annealing statistically robust. In contrast, the resistive annealing data comes from a 540 °C annealed $\delta\text{-}Pu_{1-x}Ga_x$ ($x = 0.033$) specimen approximately two years old since last melted (removed helium) but 20 years old since removal of americium, and uranium and aged five days at low temperatures [11]. Slight differences in the overall annealing curves might be expected from the differing alloy concentrations, but the overall

behaviors are expected to behave in a similar manner. Ideally, a comparison of multiple isochronal annealing curves will make use of identical soak times (isochronal) at each annealing temperature. In reality, this may not be practical due to experimental constraints, and the overall effect is that the experiment performed with a longer soak time will shift the curve towards lower temperatures as the anneal at each temperature is more complete in these cases.

The resistivity and magnetic susceptibility isochronal annealing curves for the two Ga stabilized δ-Pu specimens are shown in Fig. 1b The isochronal anneal period for the magnetic susceptibility study (red circles) was 30 min while for the resistivity study (blue diamonds) it was 5 min. The longer and more complete annealing time for the magnetic susceptibility explains why the corresponding resolvable features in the data are shifted to lower temperatures. This is particularly evident for the Stage V annealing. Further differences in the results can be explained by the relative sensitivities of each technique to the types and numbers of defects present. Whether or not the different alloy compositions contribute to this remains an open question requiring further investigation.

The most obvious difference is associated with the Stage-I-like annealing, which is clearly observable in the resistivity curve with a reduction of nearly 70% between 30 K and 40 K but is apparently absent in the magnetic susceptibility data. Nevertheless, the magnetic susceptibility data does appear to reveal the remaining four Stages of annealing. One way of understanding this is to hypothesize that the magnetic susceptibility measurements are rather insensitive to interstitials in δ-Pu but are sensitive to vacancies. Such an idea is reasonable given the much lower density of δ-Pu (15.92 g/cm^3) compared to α-Pu (19.84 g/cm^3), and its ability to more easily accommodate interstitial atoms. Furthermore, the roughly 4 at% Ga that stabilizes the δ-phase means that one of every 25 atoms is an impurity site, so the density of impurities available to trap interstitials in this alloy is considerably higher than for the α-phase. Working with this hypothesis, as the interstitials become mobile based on the evidence from the resistivity data they tend to become trapped at impurities or interstitial clusters instead of annihilating with vacancies, so the vacancy population remains relatively unchanged. Then, as Stage II annealing occurs between 50 K and 110 K, interstitials escape from impurity sites, and some begin to annihilate with vacancies leading to the approximately 20% decrease in the magnetic susceptibility annealing with a smaller change in the resistivity annealing curve, as observed. Next at the onset of Stage III annealing, vacancies begin to move with a concomitant large drop in the magnetic susceptibility annealing curve. The formation of vacancy loops, clusters, and other aggregates is indicated by the change of slope just below 200 K, a temperature consistent with the more subtle change of slope suggested for the onset of Stage IV annealing in α-Pu, and then finally Stage V occurs just below 300 K in the magnetic susceptibility data, and around 310 K in the resistivity data. The important observation from this data is that the vacancies appear to be the primary contributor to the magnetic susceptibility annealing curve in δ-Pu, while it is less clear that this is the case for α-Pu.

3.3. $Pu_{1-x}Am_x$ ($x = 0.224$)

Fig. 2 shows the EMS annealing results for δ-$Pu_{1-x}Am_x$ ($x = 0.224$) where the damage was accumulated at sufficiently low temperatures that once formed, the damage cascades are frozen in place. Previous work on radiation damage in PuAm alloys has also investigated the consequence of damage on magnetic susceptibility, but such damage was accumulated near room temperature [16,17], which as shown in Fig. 2, is very close to the annealing stage where small vacancy clusters are unstable. Therefore, measurements of damage accumulated for extended periods of time at room temperature cannot be easily related to the data from this study. There is no resistivity annealing measurements for this or a similar alloy specimen with which to compare, so this data is shown along with the δ-$Pu_{1-x}Ga_x$ ($x = 0.043$) curve for comparison. There are several general similarities, including no observable decrease in the EMS for annealing temperatures below 33 K, and the return to an effectively fully annealed state just above 300 K. However, δ-$Pu_{1-x}Am_x$ ($x = 0.224$) shows no obvious intermediate states between 60 K and 300 K, instead just a broad monotonic decrease in the fraction of EMS signal retained. An examination of the derivative suggests there are changes of slope around 100 K and around 260 K which, from analogy with the δ specimen would be the onset of Stage III, and Stage V annealing respectively. These are slightly lower in temperature than observed for delta, but that is not surprising considering the interatomic spacing for the δ-$Pu_{1-x}Am_x$ ($x = 0.224$) is greater, which should facilitate the mobility of defects. The truly spectacular feature observed in this system is shown in the inset of Fig. 2 where between 35 K and 56 K the signal from the EMS unexpectedly grows by nearly a factor of 40 before returning to "normal" behavior. This clearly does not indicate that the amount of damage suddenly increases by a factor of 40, but indicates that something very dramatic happens to the magnetic susceptibility in the initial or early stages of annealing, most likely a result of a structural feature which develops during the annealing protocol. Hence, the inset shows this phenomena, plotted in units of emu/mol Pu, as well as relative signal strength where unity is the value of $\chi(T = 5\,\mathrm{K})$ when the annealing protocol begins.

More detailed time and temperature studies to investigate this transient feature of the annealing protocol were undertaken. After annealing at 350 K for 1 h, the specimen was again held at low temperatures for 40 days (~6.26 α-decays per million atoms), and then the magnetic susceptibility was systematically measured as it was warmed to, and cooled from progressively higher temperatures between 35 K and 60 K in 5 K steps. Measurements were made during both the warming, and cooling cycles in 1 K steps with 10 sets of measurements taken over the course of 5 min at each temperature. A subset of these measurements is shown in Fig. 3. Fig. 3a shows $\chi(T)$ on the initial warming to 35 K (green circles) after having been maintained at $T \leq 30$ K for 40 days, during which $\chi(T = 2\,\mathrm{K})$ grew from 850 μemu/mol to 1000 μemu/mol. At temperatures above 33 K, the magnetic susceptibility begins to increase with temperature. The data showing warming to 45 K (red squares) after previously warming to 40 K reveal an increase in $\chi(T = 2\,\mathrm{K})$ of

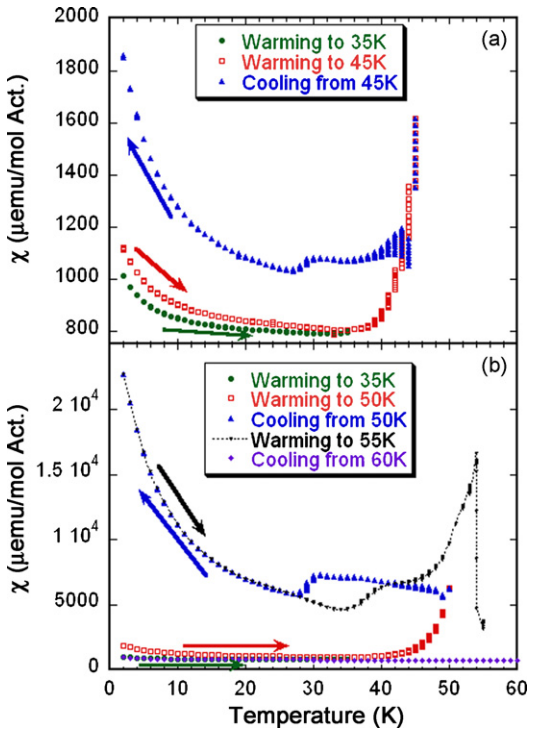

Fig. 3. The magnetic susceptibility increases dramatically, and dynamically when warmed after soaking at $T<30$ K for a month. (a) Green curve shows $\chi(T)$ warming to 35 K, the red curve is systematically warming to 45 K after annealing briefly at 40 K, and the blue curve shows $\chi(T)$ as the sample is cooled back from 45 K. (b) Green curve again shows 35 K data, while the red (blue) curve shows systematic warming to (cooling from) 50 K, and the black curve warming again to 55 K. After warming to 50 K, $\chi(T=2\,\text{K})$ exceeds its initial value by more than 20× (see text for detailed discussion). Finally, the cooling from 60 K data shows a return to the behavior expected for a typical annealing process. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

approximately 100 μemu/mol, nearly the equivalent to a month of damage accumulation in only a few hours. This shows that warming to temperatures where the first signs of annealing are expected leads to an increase in magnetic susceptibility that remains when cooling to low temperatures again More dramatically, as the temperature increases through 40 K, there is a rapid increase in the magnetic susceptibility, which is changing dramatically over the course of minutes as may be observed from the spread of data points at each temperature. Upon cooling from 45 K (blue triangles), the magnetic susceptibility is irreversible, following a different path, resulting in $\chi(T=2\,\text{K})$ increased by 850 μemu/mol from the data prior to heating. Several other features are noteworthy in the cooling data, including a shoulder at about 30 K not observed in the warming data. As the temperature decreases to below 30 K, the spread of the isothermal data points shrinks to a negligible value, demonstrating that the defects again become frozen into place and active annealing ceases. This strong time dependence at $T>33$ K explains why the low temperature $\chi(T)$ values are smaller than the factor of 20 observed for the 45 K isochronal anneal (see inset of Fig. 2).

A tremendous increase in magnetic susceptibility is spectacularly demonstrated in Fig. 3b, where further cycles in temperature extending to 50 K and 55 K are shown with $\chi(T=2\,\text{K})=$ 22,500 μemu/mol after the 50 K anneal—an increase of 22× over the value obtained after aging at low temperature. A comparison between Fig. 3a and Fig. 3b shows a shoulder in the cooling data at both 45 K (Fig. 3a, blue triangles), and 50 K (Fig. 3b, blue triangles) which is not observed in the warming data. Presently, there is no simple explanation for this, but it may be related to relaxation of structures formed at higher temperatures which are not accessible on subsequent heating cycles. Below this shoulder, the damage retained appears to be frozen in place once more as the time dependence of the isothermal magnetic susceptibility becomes constant (within experimental uncertainty). However, in this frozen state there is a remarkable change in the thermodynamic state of the alloy when compared to the state prior to warming. Annealing to 58 K rapidly dissipates the observed enhancement, and by 60 K the anomalously large magnetic susceptibility has vanished both at the anneal temperature and all temperatures below, as shown by the data cooled from 60 K in Fig. 3b (purple diamonds) where the extraordinary increase in the magnetic susceptibility has disappeared. On subsequent annealing at higher temperatures the isochronal annealing behavior is now similar to Ga-stabilized δ-Pu.

The low temperature behavior of this large transient EMS after annealing to 50 K (Fig. 3b: blue triangles, black triangles) can be fit to a modified Curie Weiss law: $\chi(T)=\chi_0+C/(T-\theta)$ for $T<25$ K which results in an enhanced temperature independent contribution of $\chi_0=1400$ μemu/mol as compared to an annealed value of approximately 700 μemu/mol, a paramagnetic Weiss temperature (θ) of −3.9 K, and most astoundingly, an effective moment (μ_{eff}) in excess of 1.1 μ_B/Pu atom. Such a dramatic change in a thermodynamic property, from essentially non-magnetic to possessing an effective moment comparable to that of magnetically ordered Pu compounds, such as PuH_2, implies a phase change in this alloy driven by the accumulated damage. This is further supported by the near doubling of χ_0, which implies a dramatic change in the electronic band structure, with this transient phase possessing a much narrower density of states.

The idea that radiation damage could drive a magnetic phase transition in a Pu–Am alloy is worthy of consideration. Pure Pu possesses multiple solid-state phases below the melting temperature, with the α, β, γ, δ, and δ' phases all separated by less than 2 mRyd in energy [3], as compared to the 10–30 mRyd differences in energy between the solid-state phases of the neighboring actinides: Np and Am. Thus, multiple phases of Pu are nearly degenerate, and can be stabilized by perturbations, such as doping with Ga or Am to stabilize the δ-phase. The δ-$Pu_{1-x}Am_x$ ($x=0.224$) alloy may also have other nearly degenerate phases, some of which have very different magnetic properties and become accessible by perturbing the system with disorder.

There is insufficient data to form a complete picture of the phenomena observed in this alloy, but one possible interpretation is outlined as follows. As in the other Pu samples discussed in this work, self-damage is frozen in place at low temperature, while simultaneously increasing the entropy of the system. This increase must change the overall free energy of the system, and may lower the free energy of a competing phase below that of the undamaged phase; yet a phase transition is kineti-

cally inhibited at the low temperatures. By warming the system to a point where annealing begins, the barrier is lowered sufficiently for the phase transition to occur, which is reflected as a change in the magnetic (thermodynamic) properties. As the system is further annealed, the new phase grows, as indicated by the further increase in magnetic properties. However, the annealing also reduces the entropy of the system making the new phase less stable, so after sufficient annealing transpires, the original non-magnetic phase again becomes favourable, and the alloy reverts to its initial state. Such a phase transition need not consume the entire sample. For example, early stage annealing could nucleate a quantum Griffiths phase in the alloy, where the disorder creates magnetic droplets distributed throughout a paramagnetic background [18,19]. These droplets might arise at the damage cascades generated by the recoiling nucleus from the α-particle decay where the defect population, and therefore disorder is considerably greater. Additional measurements, particularly specific heat, and isothermal magnetization, need to be performed on this alloy to understand the remarkable changes observed in the magnetic susceptibility measurements. Nonetheless, this remarkable, if transient, increase in the magnetic susceptibility may be a sign of new magnetic phases in Pu–Am alloys, and perhaps signal proximity to a stable magnetic phase for the proper combination of Am-doping, disorder, and magnetic field.

4. Conclusions

A comparison of the magnetic susceptibility, and resistivity based isochronal annealing curves of α-Pu and Ga stabilized δ-Pu provides insight into the influence of self-damage in these systems. There are similar features observed at comparable temperatures in each type of measurement, presenting a consistent prediction of the different annealing stages. However, the relative magnitudes of the annealing stages suggest that magnetic susceptibility measurements are far more sensitive to vacancies than to interstitials in the δ-Pu specimens. The high sensitivity of magnetic measurements to vacancies is consistent with measurements of the influence of vacancies in other materials such as the high temperature superconductors [13]. Alloying Pu with Am produces an FCC phase similar to δ-Pu(Ga), but results in a remarkable 2200% increase in the magnetic susceptibility after accumulating damage, and then annealing at temperatures between 35 K and 55 K. Such a remarkable change in a thermodynamic property suggests that the annealing defects induce a phase transition in this alloy, and may indicate that disorder can precipitate a magnetic phase in $Pu_{1-x}Am_x$ (x near 25%).

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