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Magnetic properties of Pu–Ga alloys

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ABSTRACT

The magnetization and magnetic susceptibility of polycrystalline Pu_3Ga compound and $Pu_{0.92}Ga_{0.08}$ alloy have been measured as functions of temperature (between 2 and 300 K) and external magnetic dc field (from 1.0 to 5.0 T) and ac field. The detailed analysis indicates the antiferromagnetic ordering in both compounds. The dynamic magnetic susceptibility shows sharp peaks at 40.3 K in Pu_3Ga and at 30.0 K in $Pu_{0.92}Ga_{0.08}$ that correspond to Néel temperatures. The high-temperature behavior of static susceptibility gives evidence that it has both Curie–Weiss and enhanced Pauli contributions and the positive magnitudes of paramagnetic Curie temperature is indicative of the presence of competing the nearest-neighbor and next-nearest-neighbor exchange interactions in both compounds.

1. Introduction

It has been known for many years that plutonium lies in the periodic table at a position where it is intermediate between itinerant- and localized-electron behavior. Due to the absence of magnetic order in early actinide elements (from Th to Np) the 5*f* electrons of these early actinide elements are assumed to be itinerant. In the heavier actinide elements, due to an abrupt jump (at δ-Pu and Am) in the volume and the presence of ordered magnetism in Cm and the elements beyond, the 5*f* electrons of these actinide elements are assumed to be localized. Thus due to such intermediate behavior of 5*f* electrons and to six crystallographic structures at different temperatures the magnetic properties of Pu put a great challenge to researchers and attract their attention for many years. The main interest is focused on the α-Pu, δ-Pu, and δ-Pu, stabilized by adding a small amount (a few percent) of Ga and thus existing at room and low temperatures. It is assumed that in α-Pu the 5*f* electrons are itinerant, but despite a vast body of experimental and theoretical works, there is no consensus in understanding the magnetic properties of δ-Pu and Pu–Ga alloys. The struggle between opposite points of view (between sticklers and opponents of magnetism in these objects) is well described in the review by Lashley et al. [1]. On the other hand, there exist plutonium monopnictides, in which Pu atoms occupy the fcc lattice (just as in δ-Pu). The plutonium monopnictides show various types of magnetic ordering, for example, PuSb with localized Pu magnetic moment, equal to $0.745\mu_B$, orders at 85 K in an incommensurate magnetic phase, and then with gradually varying the wave–vector of magnetic structure undergoes the transition to ferromagnetic phase at 67 K (see [2,3]).

2. Experimental and statement of the problem

The samples were prepared using Pu metal in α-phase with a 19.37 density. The total amount of impurities did not exceed 0.03 wt%. The gallium was of 99.9997% purity. The samples were prepared by melting in ceramic magnesium oxide crucible sodden with barium chloride melt in electric furnace in a 5×10^{-3} Pa vacuum. Every alloy was thrice remelted to achieve better alloy homogeneity. The Pu_3Ga compound has a tetragonal Cu_3Au type structure ($a = 4.5$ Å, $c = 4.54$ Å) and $Pu_{0.92}Ga_{0.08}$ alloy is in a δ-phase (fcc structure, $a = 4.57$ Å). For measurements, the samples (mass of Pu_3Ga is 0.27 g and $Pu_{0.92}Ga_{0.08}$ 0.39 g) were tightly packed in Pyrex ampoules. The helium pressure in ampoules did not exceed 0.5 atm. The measurements were carried out (on the samples six months old) in the 2–300 K temperature range in dc magnetic fields up to 5.0*T* and in ac fields of 298, 80, and 10 Hz frequencies using a SQUID magnetometer. The susceptibility was determined as the *M*/*H* ratio.

Fig. 1(a) shows very peculiar behavior of Pu_3Ga magnetization as a function of temperature in dc fields of 5.0, 3.0, and 2.0*T*, and Fig. 1(b) shows magnetization of $Pu_{0.92}Ga_{0.08}$ at 5.0, 3.0, and 1.0*T*. Magnetizations are measured in the units μ_B per Pu atom.

Let us analyze the data obtained by a modified Curie–Weiss law, as it is done, for example, in [4,5].

$$\chi = \chi_{CW} + \chi_{\text{Pauli}}, \quad \chi_{CW} = \frac{C}{T-\theta}, \quad \chi_{\text{Pauli}} = \chi_0\left[1 - \frac{\pi^2}{12}\left(\frac{k_B T}{E_F}\right)^2\right]. \tag{1}$$

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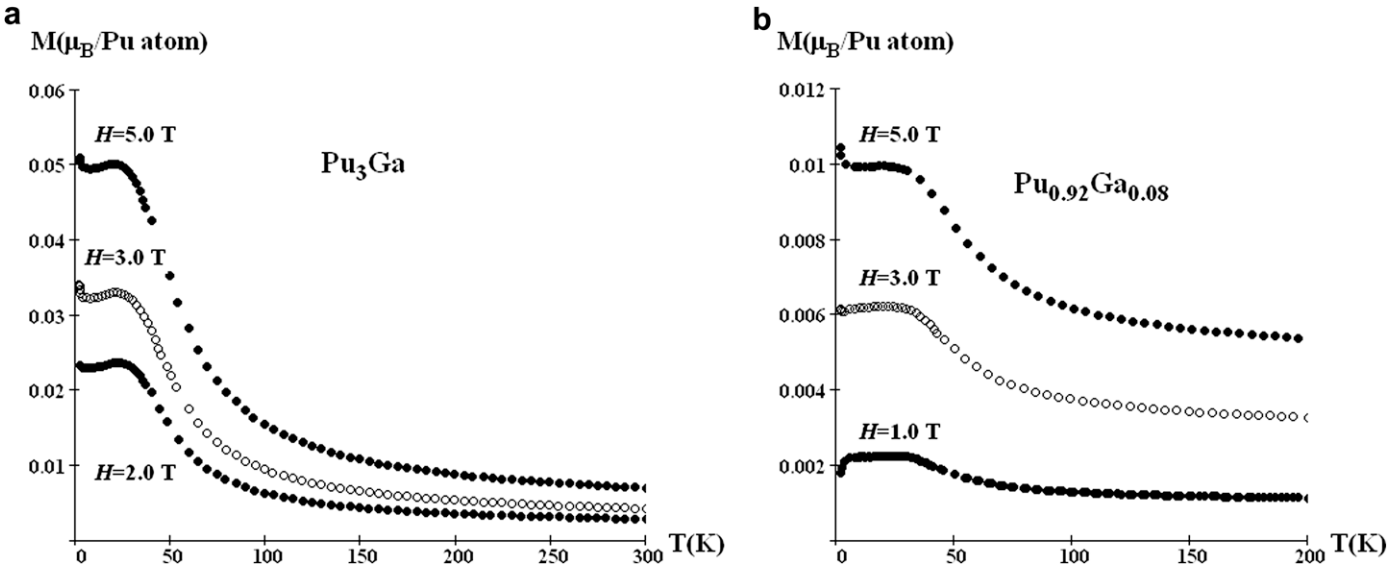

Fig. 1. The magnetizations of Pu_3Ga versus temperature at H = 5.0, 3.0, and 2.0T (a) and $Pu_{0.92}Ga_{0.08}$ at H = 5.0, 3.0, and 1.0T (b).

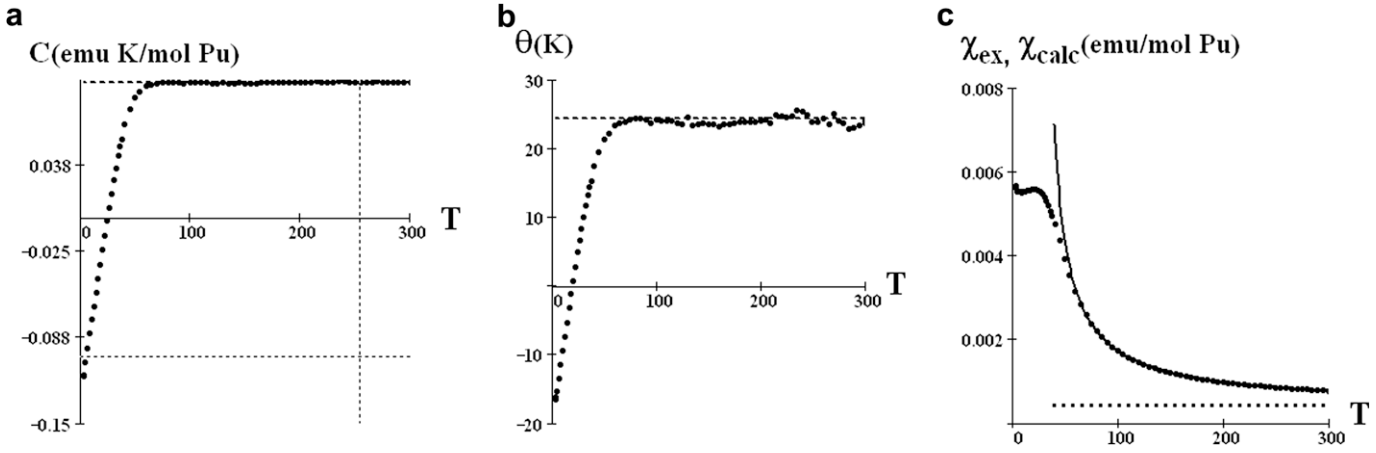

Fig. 2. Calculated C (a), θ (b) and experimental and calculated high-temperature susceptibility (c) for Pu_3Ga.

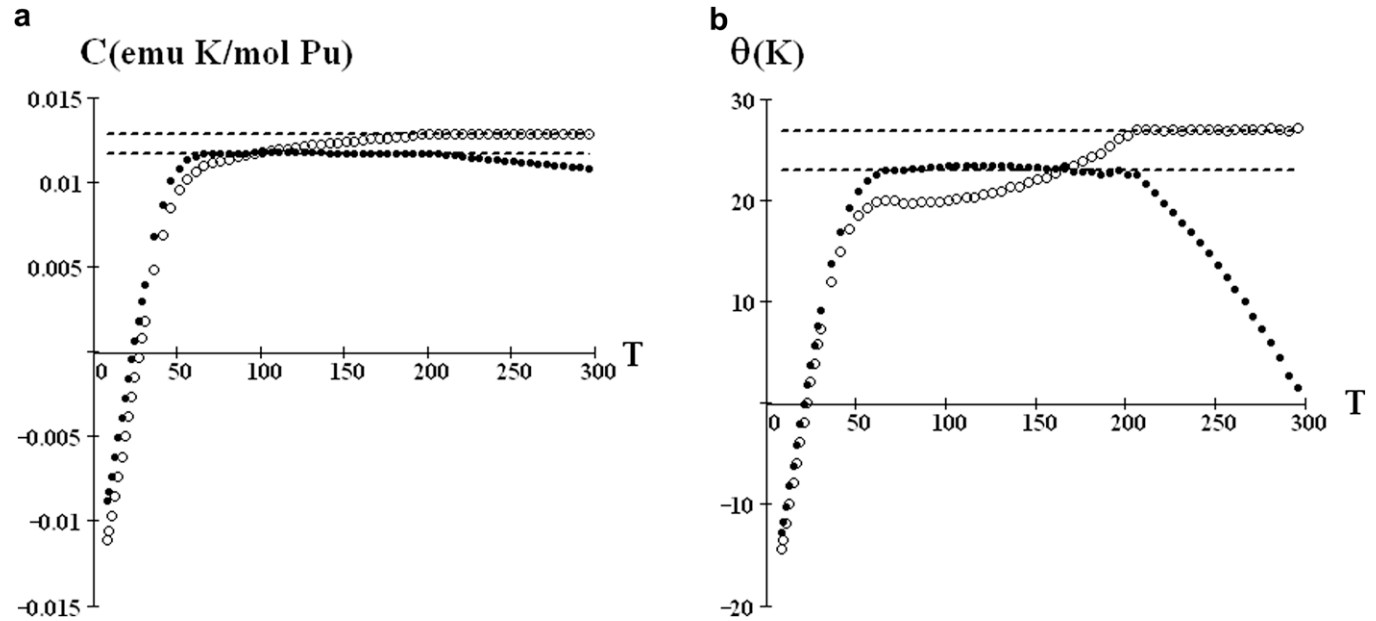

Fig. 3. Calculated C (a) and θ (b) for $Pu_{0.92}Ga_{0.08}$. Results of fitting for temperatures below 210 K – solid circles; for temperatures above 210 K – open circles.

Eq. (1) consists of two contributions to the magnetic susceptibility. The Curie–Weiss susceptibility, which is due to localized moments and Pauli susceptibility characteristic of itinerant electrons (delocalized moments). The necessity of taking into account a T^2 correction in Pauli susceptibility, that depends on the Fermi energy and represents departures from free-electron behavior, was substantiated in [5]. We slightly modified the fitting scheme expressing the Curie constant C and paramagnetic Curie point θ as functions of temperature through the experimentally obtained data for susceptibility, which is $\chi = M/H$. In this way, both magnitudes C and θ should have horizontal high-temperature asymptotes corresponding to the sought-for magnitudes, and from the calculated magnitudes of C one may extract the values of μ_{eff}

$$C = (\chi - \chi_{\text{Pauli}})(T - \theta), \quad \theta = T - \frac{C}{\chi - \chi_{\text{Pauli}}}. \tag{2}$$

Fig. 2 shows the calculated magnitudes of C and θ and the result of such fitting for the high-temperature susceptibility in the case of Pu_3Ga.

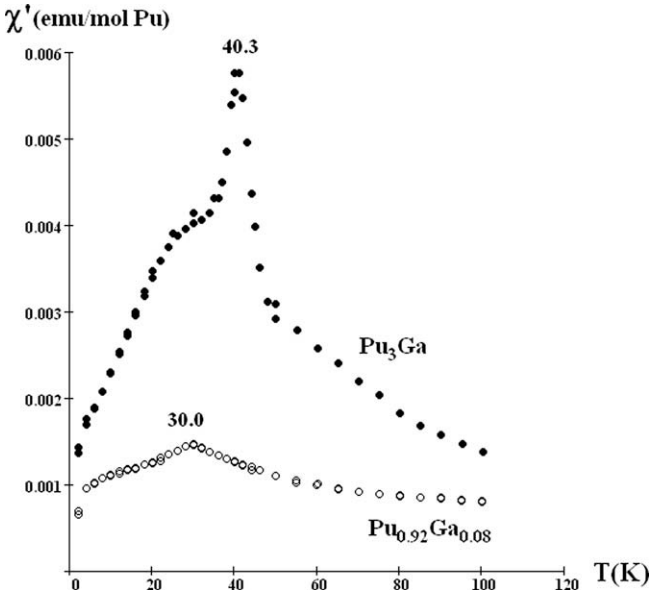

Fig. 4. The real part of dynamic susceptibility versus temperature.

Fig. 3 shows the results of such calculation scheme in the case of $Pu_{0.92}Ga_{0.08}$. Here we observe a new effect. The C and θ curves are broken near $T = 210$ K and the fitting can now be performed not for the whole high-temperature region, but for two separate regions: above and below the inflection point (210 K).

We also performed the measurements of dynamic magnetic susceptibility in ac fields of 299, 80, and 10 Hz frequencies and obtained the results that are almost independent of frequency both in Pu_3Ga and $Pu_{0.92}Ga_{0.08}$. Fig. 4 shows the temperature dependence of the real part of dynamic susceptibility (measured in ac field of 80 Hz frequency) with an asymmetric λ-shaped sharp peak at 40.3 K indicating the temperature of magnetic ordering T_N in Pu_3Ga and a similar peak, corresponding to $T_N = 30.0$ K in $Pu_{0.92}Ga_{0.08}$.

For the sake of comparison, we took the dc susceptibility experimental data for $Pu_{0.957}Ga_{0.043}$ alloy from [6], applied our fitting scheme to these data, and discovered the same effect (inflection point near 220 K) as in $Pu_{0.92}Ga_{0.08}$ (Fig. 5).

3. Results and discussion

The Pu_3Ga magnetization curves Fig. 1 show the temperature behavior more complicated than in antiferromagnets with one transition (in which it has only a single hump). The paramagnetic Curie temperature of Pu_3Ga is positive (not negative as in antiferromagnets with only negative nearest-neighbor exchange interaction) (see Fig. 2) and the real part of Pu_3Ga dynamic susceptibility is asymmetric. All these facts indicate that competing interactions (positive next-neighbor and negative next-nearest-neighbor) play important role in Pu_3Ga that leads to the paramagnetic–antiferromagnetic phase transition at 40.3 K and most probably one more transition at lower temperature. Analyzing the experimental data and the calculation results in the proposed fitting scheme we came to the conclusion that in $Pu_{0.92}Ga_{0.08}$ the phase transition also occurs (at 30.0 K). Comparing the whole set of χ_0 and μ_{eff} magnitudes: 4.21×10^{-4} emu/mol Pu, $0.88\mu_B$ for Pu_3Ga; 5.28×10^{-4} emu/mol Pu, $0.32\mu_B$ at high-temperatures, 5.36×10^{-4} emu/mol Pu, $0.30\mu_B$ at intermediate temperatures for $Pu_{0.92}Ga_{0.08}$; 5.05×10^{-4} emu/mol Pu, $0.24\mu_B$ at high-temperatures, 5.18×10^{-4} emu/mol Pu, $0.20\mu_B$ at intermediate temperatures for $Pu_{0.957}Ga_{0.043}$ we conclude that the degree of 5*f*-electrons localization is the highest in Pu_3Ga ($\mu_{\text{eff}} = 0.88\mu_B$) and decreases with decreasing Ga concentration: $\mu_{\text{eff}} \sim 0.3\mu_B$ in $Pu_{0.92}Ga_{0.08}$, and $\mu_{\text{eff}} \sim 0.2\mu_B$ in $Pu_{0.957}Ga_{0.043}$.

A new effect is discovered in $Pu_{0.92}Ga_{0.08}$ and $Pu_{0.957}Ga_{0.043}$ alloys that is absent in Pu_3Ga compound. The magnetic susceptibility in

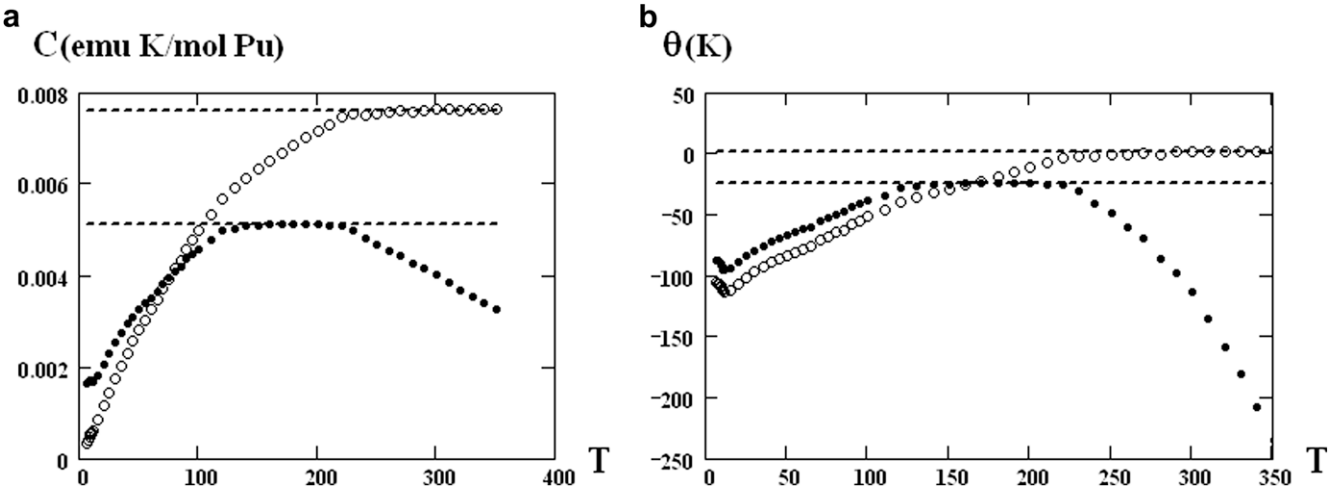

Fig. 5. Calculated C (a) and θ (b) for $Pu_{0.957}Ga_{0.043}$. Results of fitting for temperatures below 220 K – solid circles; for temperatures above 220 K – open circles.

these alloys has different asymptotes at high and intermediate temperatures with an inflection point, indicating that the magnetic system in them undergoes a rearrangement at this point. The degree of localization increases with temperature increasing: μ_{eff} raises from $0.30\mu_B$ to $0.32\mu_B$ in $Pu_{0.92}Ga_{0.08}$ and from $0.30\mu_B$ to $0.32\mu_B$ in $Pu_{0.957}Ga_{0.043}$.

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