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Americium under pressure

J.-C. Griveau [a,*], J. Rebizant [a], R.G. Haire [b], G. Kotliar [c], G.H. Lander [a]

[a] *European Commission Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, D-76125 Karlsruhe, Germany*
[b] *Oak Ridge National Laboratory (ORNL), Chemical Sciences Division, Office Box 2008, MS-6375, Oak Ridge, TN 37831, USA*
[c] *Center for Materials Theory, Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08854, USA*

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Abstract

Electrical resistivity measurements of Americium metal under pressure in the normal and in the superconducting state are reported up to 27 GPa and down to temperatures of 0.4 K under magnetic field (<0.6 T). Resistivity under pressure indicates that superconducting transition temperature T_c rapidly increased with pressures in the 5 GPa range. The pressure dependence of the upper critical field is surprising, with $\mu_0 H_c(0)$ increasing rapidly with pressure (estimated to 1 T at the maximum of T_c). Contrary to expectations, the 5f electrons play probably an important role in this material even at low pressures. We do not observe any sign of an ordered state in this material, as judged by an anomaly in the resistivity, in contrast to the predictions, but possible weak "localisation" features deduced from normalised electrical resistivity curves.

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1. Introduction

The discovery of superconductivity in Plutonium based compounds ($PuTGa_5$, T = Co, Rh) [1,2] has focused the interest of the scientific community on Transuranium elements and compounds, which were considered until recently only by the nuclear industry for their military and energetic applications. The particularity of these new materials is that they present surprisingly high superconducting properties: their critical temperature T_c – close to 18 and 9 K respectively – is one order of magnitude higher than all actinides based compounds [3–5] close to 1 K. But from a structural point of view, considering the occupancy of the 5f electrons, they would present a lot of similarities with the δ phase of plutonium metal [6,7]. This element is on the left side of the Mott transition observed in the Actinides considering the volume cell versus 5f electrons occupancy in the series [8]. On the right side of this Mott transition, Americium is also at a pivotal position. It is at the junction of heavy and light actinides. Although we should observe magnetic behaviour due to localised 5f electrons, americium metal present non-magnetic properties because of (or due to) its $5f^6$ ground state ($J = 0$). One interesting parameter used to tune this localisation/delocalisation aspect of the 5f electrons is pressure. Its effect should increase overlap between the 5f shells, and should lead to a transition from localised to delocalised state. This effect is accompanied by several structural phase transitions in the 10–20 GPa pressure range [9,10]. It has been also predicted that due to this particular non-magnetic ground state, it should display interesting superconducting behaviour [11]. Effectively, superconductivity appears around 0.8 K at 0 GPa [12] and changes quickly with pressure [13]. Americium is reported as type I superconductor with a relatively small critical field $H_c = 53$ mT [14]. But specific heat measurements [15] indicate that there is a huge discrepancy between direct determination of γ ($\sim$4 mJ mol^{-1} K^{-2}) and the estimated electronic specific heat coefficient estimated by BCS relations [16,17]: there is an overestimation by 2 orders of magnitude. Pressure induces a complex diagram with several maxima of $T_c(p)$ [13] and one part of the diagram remains unknown between Am III and Am IV because of technical limitation ($T_c < 1.3$ K). It is only recently that we observed a continuity of superconductivity all through the pressure domain up to 25 GPa [18], pressure at which

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[*] Corresponding author. Tel.: +49 7247 951 428.
E-mail address: jean-christophe.griveau@ec.europa.eu (J.-C. Griveau).

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T_c vanishes. This evolution is related to the different structural transitions and has been explained by the progressive delocalisation of the 5f electrons starting to participate to the bounding. Nevertheless, some theoretical calculation predicted the appearance of magnetism in some part of the diagram [19]. But the difficulty to conciliate the predicted structures [20,21] with the observed ones as the correct sequence of transitions lead us to re-examine the behaviour of americium metal under pressure. We determine its superconducting properties under pressure at low temperature under magnetic field using electrical resistivity.

2. Experimental

Measurements have been performed on two thin foils of americium metal (^{243}Am; $t_{1/2} = 7.38 \times 10^3$ years) with d.h.c.p. structure. Dimensions of the samples were ~550 μm × 80 μm × 30 μm. The low amount of material ($m < 100$ μg) reduces the self heating effect (6.3 mW/g). The resistance of sample is measured by a four probe-DC technique. Two different loadings have been achieved (samples A and B). For each sequence of measurements, the sample and a thin foil of lead (manometer) are held in a pyrophyllite gasket. The pressure-transmitting medium (steatite) is solid. The external pressure device is a piston-cylinder system made of non-magnetic CuBe and pressure is generated by two 1.5 mm diameter anvils made of non-magnetic tungsten carbide.

3. Normal state properties

Pressure effect reveals very interesting features (Fig. 1). We observe a strong increase of resistivity as a function of increasing pressure and temperature. This has been noted before [13,22,23] and appears as an intrinsic property of americium that is not observed in other (lighter) actinides. But the behaviour of the resistivity at low temperature differs from the one at room temperature (Fig. 2) and even if globally resistivity values are drastically increased by pressure, we can observe a T-dependent "crossover" domain in the Am II–Am III pressure range. A more detailed analysis of the resistivity in the Am I–Am II region is carried out by deriving $\Delta\rho_{\text{norm}}(T) = [\rho_p(T)/\rho_p(300\,\text{K})] - [\rho_0(T)/\rho_0(300\,\text{K})]$ ($\rho_0 \sim \rho$ at low pressure) and is shown in Fig. 3. The normalization by the room-temperature values roughly removes the influence of the form factor with pressure and the subtraction of the low-pressure contribution is an attempt to eliminate the less-sensitive pressure effects (phonons, defects) on the resistivity. $\Delta\rho_{\text{norm}}$ shows only the proportional change (with T) in resistivity as a function of pressure. We attribute the extra scattering $\Delta\rho_{\text{norm}}$ as a signature of the Mott transition when the 5f states mix with the conduction states. The two insets of Fig. 3 show (left) the minimum of $T_{\max}$ and (right) the maximum of $\Delta\rho_{\text{norm}}(T_{\max})$ as a function of pressure. The value of $\Delta\rho_{\text{norm}}$ is large near the maximum in T_c (~8 GPa). This analysis of the data of Fig. 3 gives credence to the belief that major changes are occurring in the electronic structure between 7 and 10 GPa, essentially within the Am II phase.

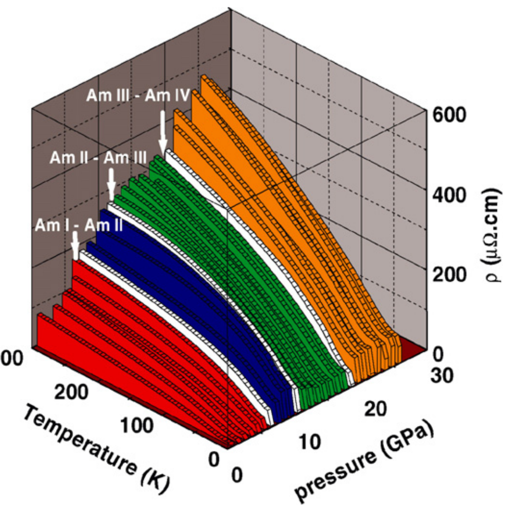

Fig. 1. Evolution of the resistivity of americium metal with pressure. We can notice the huge increase of value while pressurizing.

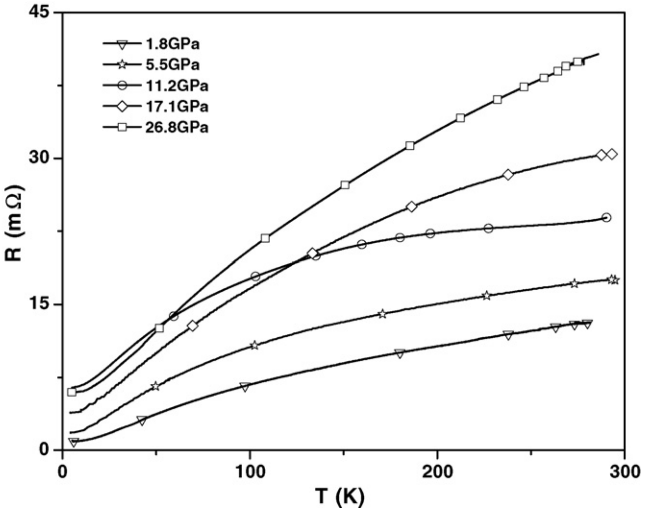

Fig. 2. Selected curves of americium metal for characteristic pressure values. We observe a difference of behaviour between low temperature and room temperature. This suggests a possible cross over with temperature vs. pressure.

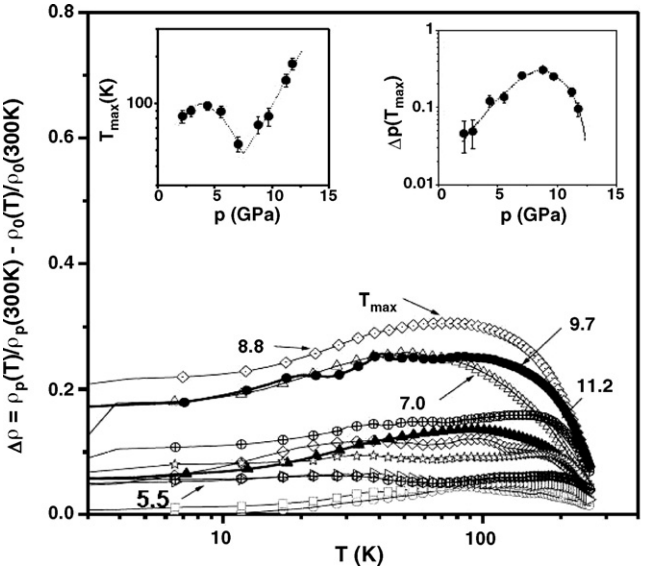

Fig. 3. Variation with pressure of the normalized resistivity curves as explained in the text. Insets show the variation of the value and temperature value obtained by this treatment.

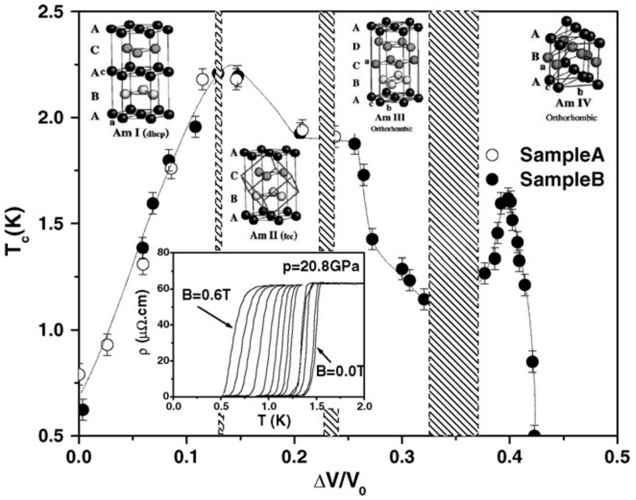

Fig. 4. T_c vs. volume cell under pressure. There is a clear difference between Am IV and the lower pressure structures. The inset shows the quality of the superconducting transition at 20.8 GPa.

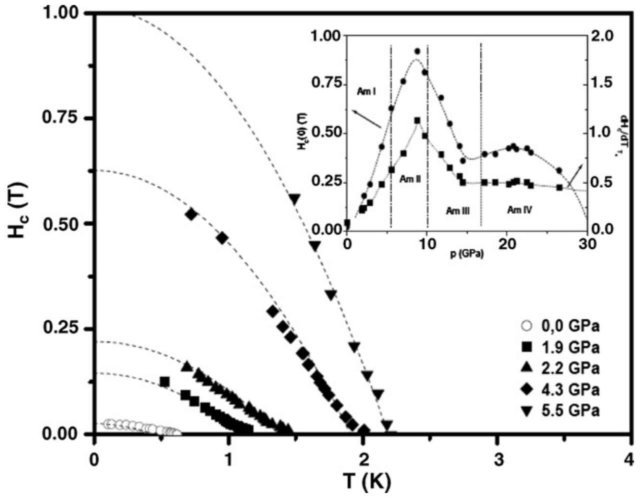

Fig. 5. Evolution of the critical field under pressure for Am I structure (d.h.c.p.). Inset shows respectively the variation of H_c (top curve, left axis) and the slope at T_c (down curve, right axis).

4. $T_c(p)$ diagram

The position of the maximum in $T_c(p)$ is related to phase transition Am I–Am II. The $T_c(p)$ diagram for ^{243}Am has been determined till 25 GPa and down to 400 mK (see Fig. 4). We observe the same global behaviour as previously reported [13]. In the 14–18 GPa pressure range, superconductivity is still present so that Am III remains a superconductor. At the highest pressure of our experiment, $p \sim 27$ GPa (in Am IV phase), T_c first increases and then linearly decreases with pressure with a slope of −0.15 K/GPa. Superconductivity is extrapolated to collapse around 30 GPa. The behaviour of the superconductivity in the high pressure domain is very interesting. A clear maximum develops without structure change. The graph representing T_c versus the volume (cell Fig. 4), make appear a clear peak just at the border of the Am III–Am IV transition.

5. Critical field

The variation of the critical field $H_c(T)$ at different pressures is illustrated in Fig. 5. With applying pressure, we observe a large enhancement of $H_c(T)$ and $H_{c0} \sim 1$ T at the maximum of T_c ($p \sim 6.3$ GPa). At higher pressure, critical field is reduced following the behaviour of T_c. Considering $H_c(T)$ shape, we are close to the orbital limit, which seems to fit well in all the pressure ranges. The slope at T_c evolves globally with the values of T_c up to Am IV. Then, it remains more or less constant. Considering critical field values, there could be a possible change of type of superconductivity under pressure from type I to type II.

6. Discussion

Several interesting features concerning the superconducting states of americium have been revealed.

The huge value of dH_c/dT at T_c induced by pressure indicates that the effective mass of Cooper pairs is very important (estimated close to 20 m_0 in clean limit). It is contradictory to the direct measurement of specific heat at ambient pressure in the normal state [15]. We have reported the same features for a single crystal of $AmCoGa_5$ measured at relatively low pressure [24,25]. In this latter case, the possibility of an excess of gallium metal has been evoked. But, it is appealing to relate these common features to an underlying physics associated to the mechanism of americium related superconductivity. Electrical resistivity treatment makes appear a maximum varying in temperature and in values. Fig. 3 and insets present the characteristics of the "Kondo like" shape observed on the normalized curves and of its pressure dependence. We can notice that the maximum of the critical field and of electrical resistivity corresponds to a change of regime of T_{max} and not associated directly to a structure change. Recent theoretical work [26] supports this approach of a possible "Kondo-like" phenomenon. When pressure increases, the $5f^6$ ground state of the atom starts admixing an $5f^7$ configuration with a very large total momentum and due to hybridization with the spd bands, this large spin gets screened, lowering the energy of the system. Simultaneously, hybridization increases by applying pressure, reducing the energy of the $5f^7$: this confirms our observation of resistivity increase.

7. Conclusion

We suggest that the 5f electrons play an important role in the transport properties, strongly scattering the spd conduction electrons when they are localized, and contributing to the transport when they are itinerant. The $T_c(\Delta V/V_0)$ diagram shows a progressive delocalisation instead of an abrupt loss of 5f character. In the entire pressure domain studied, neither clear metallic nor insulator state has been observed, but a weak "Kondo-like" aspect could take place in Am II–Am III from data treatment. Americium features under pressure stress the fact that despite the recent progresses achieved in our understanding of the 5f electrons characteristics by modelisation, we are still lacking maybe the essential of their nature. The discovery of the superconductivity in the Plutonium compound [1,2] and the surprising novel structure phase in Curium [27] at high pressure are only two striking examples among a lot in the Actinides series. Only a better knowledge acquired by accumulating experiments from simple or more sophisticated techniques (specific heat below 1 K, thermo-electrical transport, neutrons, μSR, etc.) will maybe reveal it.

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