# Resistivity studies of cubic americium hydrides from 20 to 300 K

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## Abstract

We report low temperature resistivity measurements of cubic americium hydrides. For all compositions $AmH_{2+x}$, $0 \le x \le 0.7$, there is a change in temperature dependence of the resistivity at 120 K that is yet unexplained. For $AmH_{2.6}$, there is an additional change observed near 210 K typical of long-range ordering of octahedral hydrogen that is typical of fluorite-type neptunium, plutonium, and rare-earth hydrides.

Keywords: Resistivity; Cubic americium hydrides

## 1. Introduction

Little is known of the americium–hydrogen system; experimental studies have been limited to the pressure-composition-temperature (P-C-T) and X-ray diffraction work of Refs. [1] and [2]. Americium forms fluorite-type hydrides $AmH_{2+x}$ with $0 \le x \le 0.7$, and hexagonal trihydride. Both phases are isostructural with the corresponding phases in the $NpH_{2+x}$, $PuH_{2+x}$, and most of the rare-earth hydride systems. The lattice contraction typical of the cubic rare-earth and plutonium hydrides is also characteristic of the $AmH_{2+x}$ system; $NpH_{2+x}$ expands anomalously with hydrogen content [1–3].

Neutron diffraction studies of isostructural rare-earth and actinide fluorite-type hydrides show that hydrogen fills interstitial tetrahedral sites (T-sites) preferentially for low compositions but at higher compositions enters octahedral sites (O-sites) before the tetrahedral sites are saturated (Fig. 1) [4–6]. No magnetic or transport data have been published for americium hydrides to date, but because of its $f^6$ configuration, americium should form nonmagnetic hydrides. We report here the first electrical resistivity studies of cubic americium hydrides.

## 2. Experimental details

A high-purity sample of $^{243}Am$ with dimensions $3 \times 3 \times 0.635$ mm was prepared by chromatographic reduction of the oxide and multiple distillation onto
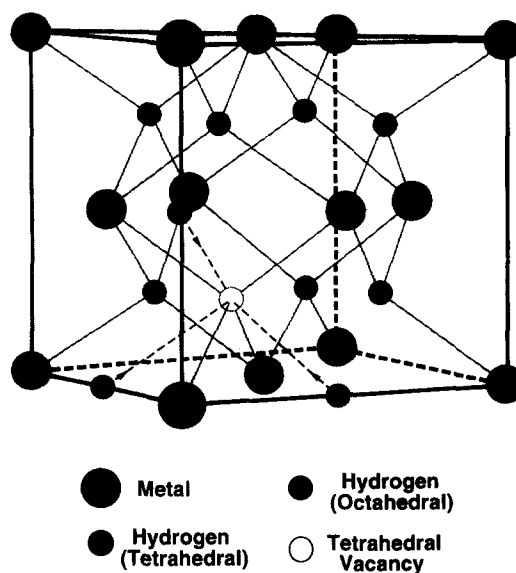


Fig. 1. Structure of fcc actinide hydrides, showing octahedral and tetrahedral hydrogen positions.

Metal ●  Hydrogen (Octahedral) ●
Hydrogen (Tetrahedral) ●  Tetrahedral Vacancy ○

a cooled receiver. The resulting metal was hydrided very slowly, resulting in monolithic polycrystalline metal hydride samples for all compositions but $x=0.6$, for which a sample was pressed from the resulting finely divided metal hydride. The pressed pellet was 3.2 mm in diameter and 1.5 mm thick.

Resistivity measurements are made in an Air Products Displex refrigerator. Samples are mounted on a sapphire disk with Wakefield Thermal Compound. The sample thermometer is mounted on the opposite side of the
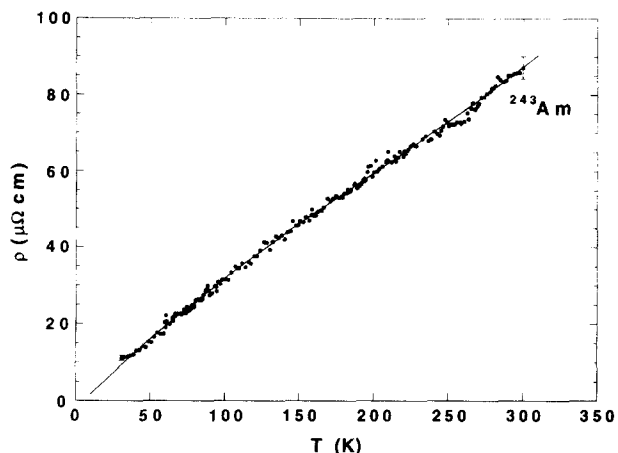
Fig. 2. Resistivity data for americium metal compared with those of Ref. [7] (solid line). The error bars represent the maximum deviations of the data to a least-squares fit (not shown) with multiple correlation coefficient $R = 0.999$; the error is 3.4% at 300 K increasing to 6% at 30 K.

sapphire disk inside an OFHC copper sleeve bonded to the sapphire with Thermalbond epoxy. Electrical contact is made to the sample with four spring-loaded contacts. The resistivity measurement is made with low frequency (approximately 100 Hz) AC current, and the small signal detected with a lock-in amplifier. The cooling rate is typically 5 K $min^{-1}$ and the warming rate, 1 K $min^{-1}$.

Resistivity data for our polycrystalline $^{243}$Am metal sample (dhcp structure) are shown in Fig. 2. The solid curve shows for comparison early data published by Schenkel et al. [7] for $^{241}$Am deposited thin films. The room temperature resistivity for our bulk metal sample is 85 $\mu\Omega$ cm, somewhat higher than the 72 $\mu\Omega$ cm published in Ref. [7] for the film sample. The estimated residual resistivity for our sample is 10 $\mu\Omega$ cm, compared with 11.53 $\mu\Omega$ cm for the film sample of Ref. [7]. These small discrepancies in the two measurements may be accounted for by preferential orientation (columnar growth) in the film — important in a nonisotropic crystal structure — and differences in purity.

## 3. Resistivity results

Fig. 3(a) shows the resistivity of $AmH_{2.0}$, the composition at the lower phase boundary, as a function of temperature from 30 to 300 K. There is a change in temperature dependence at 120 K. Above 120 K, $\rho$ is linear with $T$ with a slight negative curvature, while the data below 120 K fit a second-order polynomial well (Fig. 3(b) and (c)). Similar behavior in rare-earth hydrides has been shown to be associated with short-range ordering of tetrahedral hydrogens [8]. Fig. 4 shows data for an intermediate composition, $AmH_{2.2}$, with temperature dependence similar to that for $AmH_{2.0}$ but with a slight positive curvature in the higher tem-
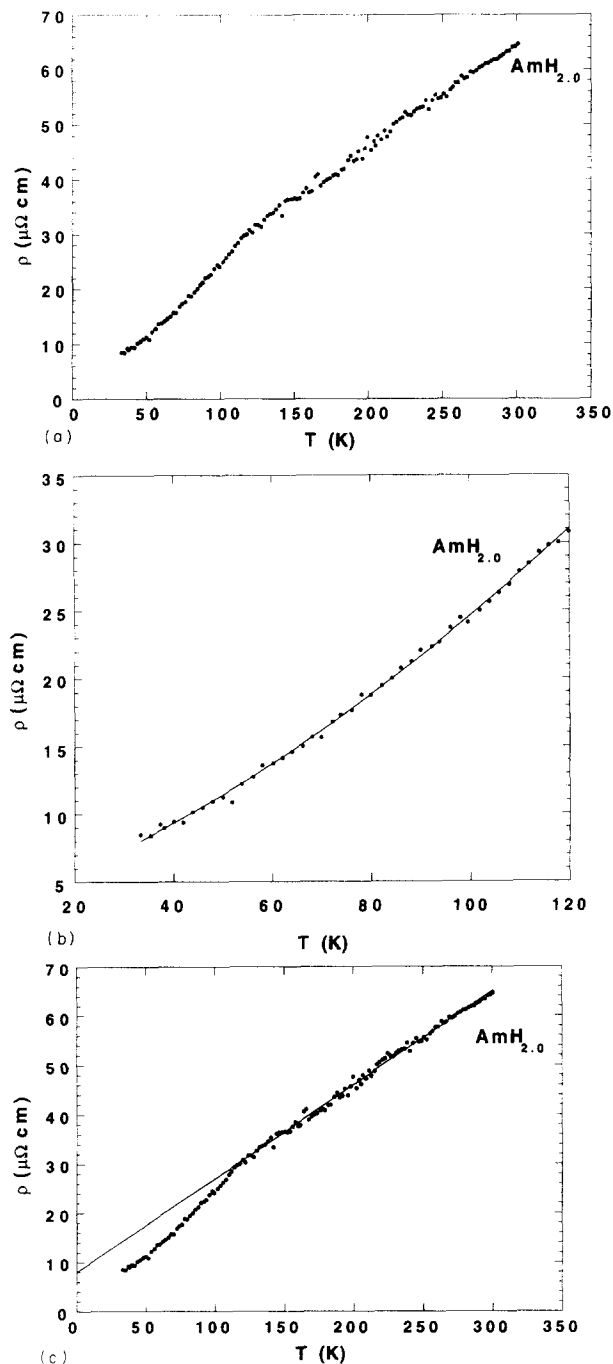


Fig. 3. (a) Resistivity data for cubic americium hydride at the lower phase boundary, showing the inflection near 120 K. (b) Low temperature resistivity data for $AmH_{2.0}$, showing the polynomial fit to the data, $\rho = 2.6 + 0.13\ T + 0.00087\ T^2$; $R = 0.999$. (c) Resistivity data for $AmH_{2.0}$, showing the linear fit, $\rho = 7.9 + 0.19\ T$, to the data above 120 K; $R = 0.998$.

perature range and the inflection occurring nearer to 150 K.

The americium hydride resistivity is considerably lower than that of the light-actinide dihydrides, as exemplified in Fig. 5 where our data for $AmH_{2.0}$ are plotted with those for $NpH_{2.13}$ [5], and for $PuH_{1.93}$ [9], i.e., at the lower phase boundaries for these systems.
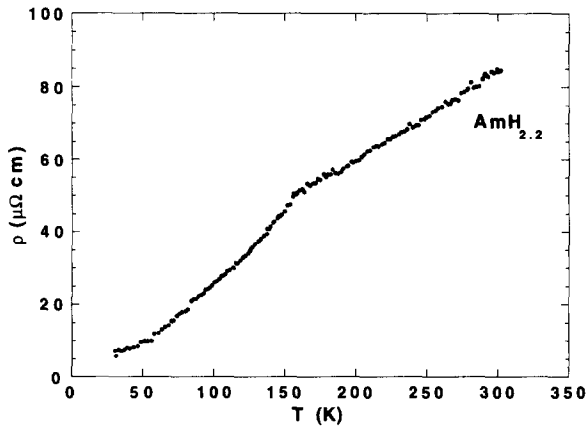
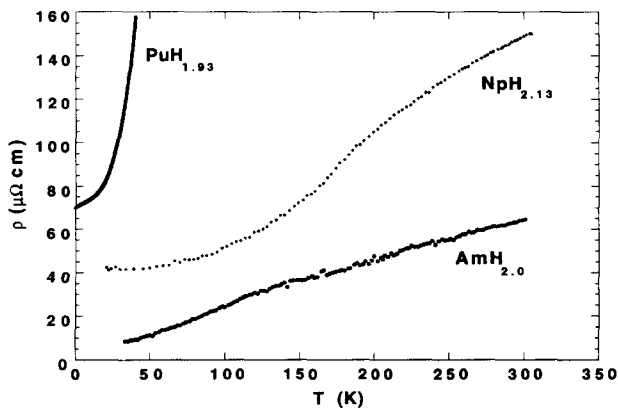Fig. 4. Resistivity data for $AmH_{2.2}$, showing behavior similar to that of $AmH_{2.0}$.



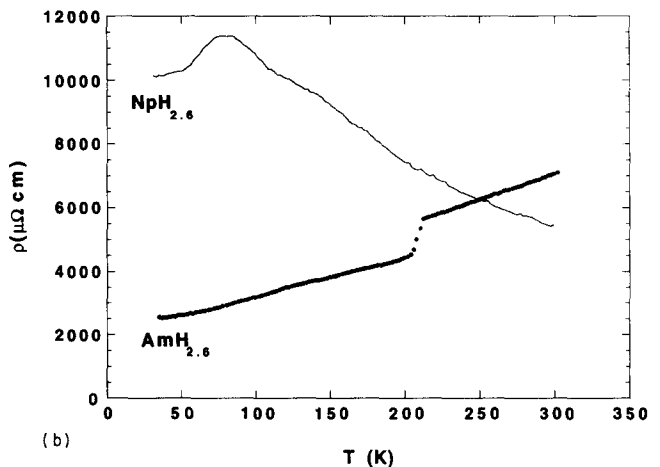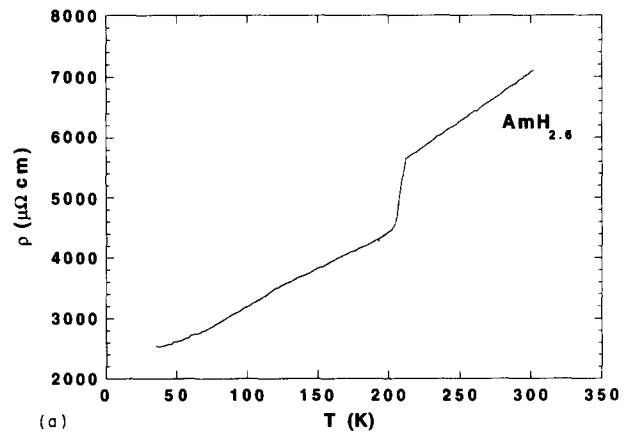Fig. 5. Resistivity of $AmH_{2.0}$ compared with $NpH_{2.13}$ and $PuH_{1.93}$.



Fig. 6. (a) Resistivity data for cubic americium hydride near the upper phase boundary. (b) Resistivity data for $AmH_{2.6}$ plotted with those for $NpH_{2.6}$ for comparison.

For the plutonium hydride system, there is a sharp ferromagnetic transition at about 65 K (transition temperature depending on composition); the resistivity for $PuH_{1.93}$ rises thereafter to a value of 527 $\mu\Omega$ cm at room temperature. The data for $AmH_{2.0}$ show a resistivity actually lower than that for the metal (cf. Fig. 2), which is typical of a true rare-earth system, i.e., with fully localized, core-like f electrons.

Resistivity data for a higher-composition hydride, $AmH_{2.6}$, show two prominent features (Fig. 6(a)). Still present is a change from linear to $T^2$ temperature dependence of the resistivity data at 118 K upon cooling, similar to that observed in the lower-composition hydrides. A second, large transition observed near 210 K occurs only in the 2.6-composition hydride. This feature is typical of long-range ordering of octahedral hydrogens that is common in isostructural rare-earth hydrides; we have observed this ordering in the isostructural neptunium hydrides using resistivity and neutron diffraction techniques [5]. In these hydrides there is a tetragonal distortion upon cooling whereby the unit cell doubles, and octahedral hydrogens fill those O-sites along the $c$ axis preferentially. Fig. 6(b) shows the resistivity of $AmH_{2.6}$ compared with our data for

$NpH_{2.6}$. In $NpH_{2.6}$, the resistivity rises upon cooling to a maximum centered near 75 K; the hydrogen ordering is manifested by a broad shoulder that begins at 200 K and extends to just above 100 K.

The lower-temperature transition could also be structural or electronic, but, as noted previously, it is unlikely that it is magnetic because of the americium $f^6$ configuration. Neutron diffraction studies would be useful to determine the exact nature of the low temperature transitions in this hydride system.

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## References

[1] W.M. Olson and R.N.R. Mulford, J. Phys. Chem., 70 (1966) 2934.

[2] J.W. Roddy, *Inorg. Nucl. Chem.*, *35* (1973) 4141.

[3] J.W. Ward, in A.J. Freeman and C. Keller (eds.), *Handbook on the Physics and Chemistry of the Actinides*, 3, Elsevier Science, 1985, pp. 1-74.

[4] C.E. Holley, Jr., R.N.R. Mulford, F.H. Ellinger, W.C. Koehler and W.H. Zachariasen, *J. Phys. Chem.*, *59* (1955) 1226.

[5] J.W. Ward, B. Cort, J.A. Goldstone, A.C. Lawson, L.E. Cox and R. G. Haire, in L.R. Morss and J. Fuger (eds.), *Transuranium Elements, A Half Century*, ACS Books, 1992, pp. 404-415.

[6] J.W. Ward, J.M. Haschke, J. Rebizant and W. Bartscher, *J. Less-Common Met.*, *100* (1984) 195.

[7] R. Schenkel, H.E. Schmidt and J.C. Spirlet, The electrical resistivity of $^{241}$americium metal, in W. Müller and R. Lindner (eds.), *Transplutonium 1975*, North-Holland, 1976, pp. 149-159.

[8] P. Vajda, J.N. Daou and J.-P. Burger, *Z. Phys. Chem. Neue Folge*, *163* (1989) 637.

[9] J.O. Willis, J.W. Ward, J.L. Smith, S.T. Kosiewicz, J.M. Haschke and A.E. Hodges III, *Physica*, *130B* (1985) 527.