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Superconducting critical field and low temperature heat capacity of americium (*)

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Résumé. — Des mesures à basse température sur le supraconducteur américium donnent un coefficient de chaleur spécifique électronique $\gamma = 2 \pm 2$ mJ/mole $\cdot$ K² et un champ critique $H_c(0) \sim 500$ Oe. Le premier est plutôt petit tandis que le champ critique élevé est inattendu.

Abstract. — Low temperature measurements on the superconductor americium yield an electronic heat capacity coefficient of $\gamma = 2 \pm 2$ mJ/mole $\cdot$ K² and a critical field of $H_c(0) \sim 500$ Oe. The former is rather small while the critical field is unexpectedly large.

1. Introduction. — The element americium has recently been shown to be a superconductor [1]. Since general thermodynamic properties of superconducting elements are an aid to understanding superconductivity, we decided to measure the critical field (H_c) and electronic heat capacity coefficient (γ) of americium. These two measurements are thermodynamically related and we hoped that they would constitute a check of the experimental self-consistency. The only previous measurement of this nature on Am was the value of $\gamma = 3 \pm 3$ mJ/mole $\cdot$ K² from the recent resistivity and heat capacity work on americium by Müller *et al.* [2].

Our measurements proved more difficult than we had anticipated due to the heating (6.8 mW/g) and radiation damage of the ²⁴³Am metal. Furthermore, the results were more interesting than expected and thus, work is still in progress.

2. Experimental results and discussion. — The samples were a new reduction of americium using the techniques of reference [1]. The heat capacity measurements were performed with a 5 mg bare sample greased to a sapphire platform that was thermally weakly-tied to a 1.38 K liquid helium bath. The heat capacity was determined by the increase in thermal relaxation time that the sample added to the platform when it was heated with pulses above its equilibrium temperature [3]. The measurement was thus independent of the sample heating rate which we determined to be ~ 6.8 mW/g. However, the self-heating limited the lowest temperature attained to ~ 7 K. Our results from 7 to 19 K are shown in figure 1. The electronic heat capacity coefficient

Fig. 1. — Heat capacity of ²⁴³Am at low temperatures.

was determined from the linear extrapolation of the C/T versus T^2 plot to be 2 ± 2 mJ/mole $\cdot$ K². The large uncertainty is due to the fact that even at 7 K the electronic contribution is still only 4 % of the total specific heat. The determination of γ then allows the Debye temperatures (θ_D) to be calculated and they are shown in the figure as a function of T . These values of θ_D show a dramatic drop from higher temperature values [2]. This softening could reflect a highly elastically anisotropic structure or may be due to a martensitic transformation that could be associated with the 69 K heat capacity anomaly [2].

The critical field determinations were made in the dilution refrigerator described in reference [1] with the addition of a superconducting solenoid centred on the samples. Most of the measurements were made on a sample 0.3 mm thick, 0.76 mm wide, and 1.02 mm long that was cut from the heat capacity sample. The magnetic field was applied perpendicular to the large faces of the sample. A demagnetizing coefficient of 0.56 was assumed yielding an

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$H_{\text{internal}} \sim 2.3 H_{\text{applied}}$. (One attempt to run a 2.3 mg sphere was limited in temperature to 0.38 K by the poor geometry for cooling.)

Some typical traces of ac susceptibility *versus* applied field at constant temperature are shown in the inset of figure 2. These are very unusual. A non-radioactive, pure, strain-free superconductor should yield a trace resembling a step function with the step either occurring at the critical field (H_c) or ending at the upper critical field (H_{c2}). The observed curves for Am are a result of two competing mechanisms, both of which are extremely complicated to deal with in detail for a particular geometry. First of all the heat generated within the sample must maintain the centre at a relatively high temperature, probably it is in the normal state [1]. Secondly, the application of a magnetic field at first drives part of the surface normal (raising its thermal conductivity non-linearly), and in this case (since no sign of magnetic hysteresis is seen) finally breaks the sample into regions of normal and superconducting metal before finally driving it completely normal at H_{c2} . Now a determination of the normal and superconducting regions of the sample taking into account the thermal and magnetic restrictions for a practical geometry is almost impossible.

We believe that something useful can be extracted. It must be recalled that superconducting regions

Fig. 2. — Critical field points for Am (likely H_{c1}) as a function of temperature. The inset shows recorder traces. The curve is

$$H_c(T)/H_c(0) = 1 - T^2/T_c^2$$

where $H_c(0) = 530$ Oe and $T_c = 0.625$ K. Also

$$dH_c(T)/dT|_{T=T_c} = 1700 \text{ Oe/K.}$$

can shield normal regions making the ac susceptibility, possibly, unrealistically sensitive to subtle changes in the superconducting state. For this reason we disregard the ends of the traces. (We also could not extract any sensible information from the ends.) All of the traces exhibit a change of slope in the central portion (marked by the arrows in the figure) which we believe is the onset of the mixed state (H_{c1}) caused by the applied field.

Taking this viewpoint results in the critical field curve shown in figure 2 which has a very believable shape. For purposes of further discussion in this limited space we shall call this $H_c(T)$ rather than $H_{c1}(T)$ since it represents a lower limit of $H_c(T)$ which as we shall see is rather large. Finally our values of H_c increase by 2-4 %/day presumably due to stored radiation damage. In support of reference [2], annealing at 75 K restores most of the changes.

To barely touch on theoretical considerations we first go to McMillan [4]. Starting with

$$\gamma = 2.0 \text{ mJ/mole} \cdot \text{K}^2, \quad T_c = 0.625 \text{ K (from Fig. 2),}$$

$$\theta_D = 121.4 \text{ K (from Fig. 1),}$$

and assuming $\mu^* = 0.13$ (probably high) we arrive at a density of states ($N(0)$) of 0.57 states/eV-atom which is very small for an elemental superconductor and we obtain an electron-phonon interaction pairing potential (V) of 0.32 eV-atom which is rather large. As the most unusual result, consider the expression

$$\gamma = (1/2 \pi) (H_c^2(T=0)/T_c^2)$$

which is a result of the simple two-fluid model [5]. Using $H_c(T=0) = 530$ Oe, we derive $\gamma = 200 \text{ mJ/mole} \cdot \text{K}^2$. Clearly americium is some type of a high critical field material. Calculations of the coherence length, penetration depth, and the mean free path are rather dependent on the choice of starting parameters and are no more satisfying than the two-fluid model result.

Although there is a great deal of uncertainty in our determinations of γ and H_c they are not large enough to make the superconducting properties of americium straightforward. As mentioned in reference [1] the superconductivity of americium is *surprising*.

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