Low-temperature specific heat of $PdPb_2$

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We have measured the low-temperature specific heat in a single crystal of $PdPb_2$ in the normal and superconducting states. The normal-state data allow us to determine the coefficient of the electronic specific term $\gamma = 1.2$ mJ/g-at. K^2, and the Debye temperature $\Theta_D = 147$ K. The electronic specific heat in the superconducting state follows exponential behavior $C_{es}/\gamma T_c = 11.53\exp(-1.42T_c/T)$ in the temperature range investigated. The data suggest that $PdPb_2$ is a weak-coupling superconductor with a high value of the pairing potential.

$PdPb_2$ is a body-centered-tetragonal compound with a $CuAl_2$ structure.[1] A number of compounds with this structure are superconducting[2,3] with $PdPb_2$ being one of them. Havinga, Damsma, and Kanis[3] studied a number of alloys involving $PdPb_2$ by measuring the superconducting transition temperature T_c, room-temperature magnetic susceptibility, and thermoelectric power. The authors found that all of these quantities show oscillatory behavior when plotted against the electron to atom ratio. This result was attributed to the electron density-of-states variation at the Fermi surface. In order to confirm this supposition it is necessary to perform low-temperature heat-capacity measurements in these alloys. To start with we have measured the low-temperature heat capacity of $PdPb_2$ partially to check the assertion of Havinga, Damsma, and Kanis[3] and partially to determine the nature of superconductivity in this material. For various reasons[4,5] the possibility of superconductivity in palladium metal and its alloys has interested researchers. Low-temperature heat capacity in this structure has been measured in two borides,[6] W_2B and Ta_2B, though not much attention was given to the results in the superconducting state.

The sample in the present measurement was a single crystal of $PdPb_2$ weighing 123 mg. The measurement was performed with a standard heat-pulse method. The sample holder consisted of a sapphire substrate (weighing 40 mg) supported by 50-μm thin Constantan leads, with a Nichrome film deposited on one side serving as the heater. The thermometer was a bare Ge resistor supported from the substrate by thin gold wires. The sample was attached to the substrate with a very thin layer (a few micrograms) of Apiezon-N grease. The heat capacity of the sample holder was measured separately and constituted roughly 10% of the total. The resistance of the thermometer was measured by an ac bridge utilizing a lock-in technique. Below the superconducting transition temperature T_c, which is found to be 3.02 K, the normal-state specific heat was measured in a magnetic field of 200 G. This field was found sufficient to turn the sample normal even at the lowest temperature. The accuracy of these measurements were better than 5%.

Figure 1 shows the measured specific heat C as a function of temperature up to 4 K in the normal and superconducting states. The width of the transition at 3.02 K is about 50 mK. The jump in specific heat C at the transition temperature is 6.5 mJ/g-at. K.

Figure 2 shows a plot of C/T vs T^2 where T is the temperature. This plot shows two distinct slopes, one at temperatures below 4.5 K and the other at higher temperatures. This kind of behavior is not uncommon in the heat capacity of metals and alloys.[7]

The normal-state specific heat at low temperatures in metals is given by

$$C = \gamma T + \beta T^3 . \quad (1)$$

The first term γT gives the electronic contribution and the second term βT^3 gives the lattice contribution to specific heat. The values of γ and β have been evaluated from Fig. 3, which is plotted as C/T against T^2 below 4.5 K. From the straight-line fit we find

$$\gamma = 1.2 \text{ mJ/g-at. K}^2 ,$$

$$\beta = 0.615 \text{ mJ/g-at. K}^4 .$$

From the value of β we have evaluated the Debye temper-

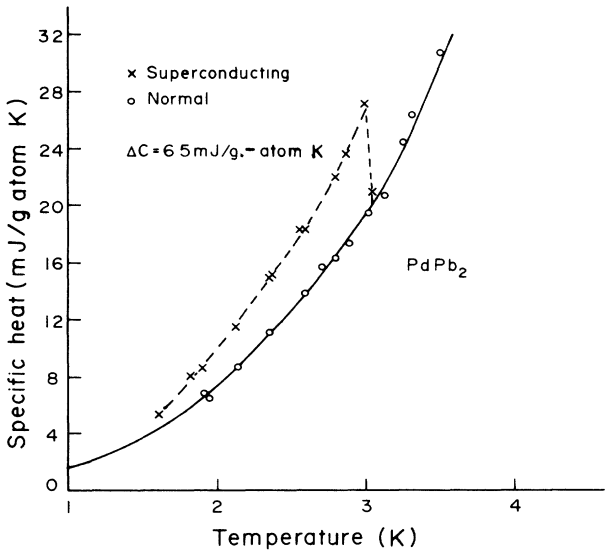

FIG. 1. Specific heat C vs temperature T in the normal and superconducting states.

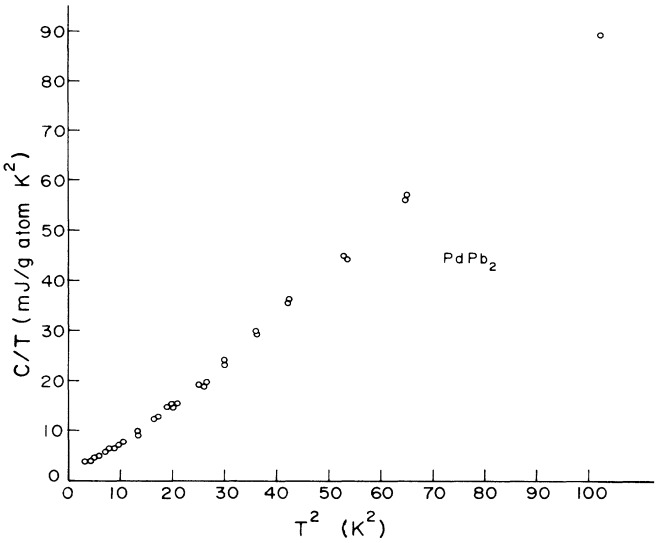

FIG. 2. C/T vs T^2 in the temperature range 1.7–10 K.

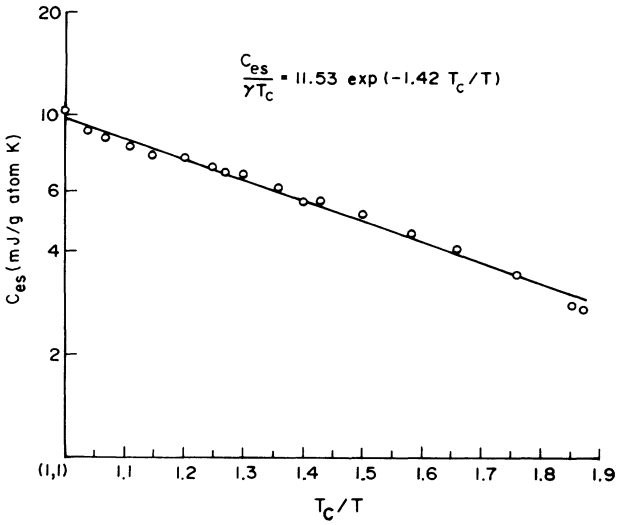

FIG. 4. The electronic specific heat in the superconducting state C_{es} is plotted against T_c/T in a semilog plot. The curve shows the exponential dependence of C_{es} on T_c/T.

ature of the compound by the formula

$$\beta = 234Nk/\Theta_D^3 , \tag{2}$$

where N is the Avogadro number, k the Boltzmann constant, and Θ_D the Debye temperature. The value of the Debye temperature is found to be 147 K. Previous determinations[8] of the Debye temperature were found to be 159 K from the room-temperature elastic constant measurements and 120 K from the low-temperature resistance measurements.

The value of γ for $PdPb_2$, it may be noted, is considerably smaller than those of the elements ($\gamma = 9.42$ and 2.98 mJ/g-at. K^2 for Pd and Pb, respectively). However, it is not possible to discuss the value of γ for $PdPb_2$ in terms of the values for the elements as the compound and the elements have entirely different crystal structures. The small value of γ is also surprising since Havinga, Damsa, and Kanis[3] found a peak in the magnetic susceptibility measurements as a function of electron to atom ratio in $PdPb_2$ while studying $Rh_{1-x}Pd_xPb_2$ and $Pd_{1-x}Au_xPb_2$ alloy systems.

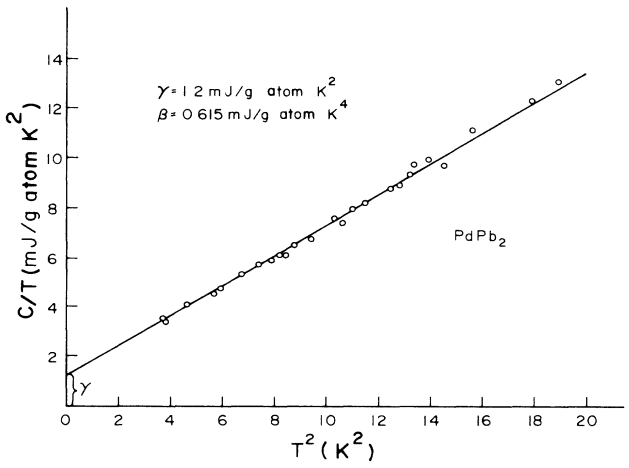

FIG. 3. C/T vs T^2 in the temperature range 1.7–4.5 K. The constants γ and β have been evaluated from this graph.

The value of the specific-heat jump ΔC at T_c, as stated earlier, is 6.5 mJ/g-at. K. This gives us $\Delta C/\gamma T_c = 1.8$ for $PdPb_2$, which can be compared with the Bardeen-Cooper-Schrieffer (BCS) value of 1.43 for weak-coupling superconductors. For strong-coupling superconductors the value of $\Delta C/\gamma T_c$ is generally greater than 2.

T_c and γ are related to the density of states for electrons $n(0)$ at the Fermi surface by

$$T_c = 1.14\Theta_D \exp\{-1/[n(0)V]\} , \tag{3}$$

$$\gamma = (2/3)\pi^2 k^2 n(0) . \tag{4}$$

Equation (3) is based on the assumption of weak coupling where V is the BCS pairing potential. Using values $T_c = 3.02$ K and $\Theta_D = 147$ K, we obtain

$$n(0)V = 0.249 .$$

Usually the value of $n(0)V < \frac{1}{4}$ is considered to be an indication of a weak-coupling superconductor. From Eq. (4) we obtain

$$n(0) = 0.255 \frac{\text{eV}^{-1}}{\text{atom spin}} .$$

Therefore $PdPb_2$ is a material with low electron density of states at the Fermi surface and relatively high pairing potential.

In the superconducting state, the electronic specific heat C_{es}, which is the difference between the total measured specific heat and the lattice specific heat, follows an exponential form as shown in Fig. 4. The curve gives the expression

$$C_{es}/\gamma T_c = 11.53 \exp(1.42\, T_c/T) . \tag{5}$$

The exponential dependence of specific heat C_{es} is an

indication of energy gap. If the gap is independent of temperature then the value of the energy gap is $2\Delta(0) = 2 \times 1.42kT_c = 2.84kT_c$. This can be compared to the BCS weak coupling value $2\Delta(0) = 3.52kT_c$. This comparison, however, is not valid since the gap changes rapidly near the transition temperature. For weak-coupling superconductor BCS gives[9]

$$C_{es}/\gamma T_c = 8.5 \exp(1.44T_c/T) \quad (6)$$

for $2 < (T_c/T) < 7$. In our measurement the maximum value of T_c/T is 2. Experimentally most of the weak-coupling superconductors give the exponential in the specific-heat measurements as from $\exp(-1.4T_c/T)$ to $\exp(-1.5T_c/T)$ (Refs. 10–12) in the range of temperature $1.5 < (T_c/T) < 5$.

We therefore conclude that $PdPb_2$ is a weak-coupling superconductor with a high value for the pairing potential.

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