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ABSTRACT

New measurements of the heat capacity of $PrCu_2$, between 70 mK and 20 K, are reported. The 7 K anomaly appears to be associated with a Jahn-Teller distortion. A value of $1 + K = 29$ is obtained for the "Knight shift" of the Pr^{3+} nuclei.

We report measurements of the heat capacity of $PrCu_2$ extending from 70 mK to 20 K in zero magnetic field and including some measurements in magnetic fields to 38 kOe. Our zero-field results are qualitatively similar to those reported earlier by Andres et al.[1], but they differ in several details.

Andres et al. observed a heat capacity anomaly in $PrCu_2$ that had a maximum near 7 K and an entropy of approximately Rln2. Associating the anomaly with a singlet first excited crystal-field level, they estimated $\Delta/k = 13$ K for its separation from the ground level. The small value of the hyperfine heat capacity and an almost temperature independent susceptibility between 1 and 7 K both suggested that the ground level was also a singlet. Below 0.5 K, the heat capacity increased with decreasing temperature, and below 90 mK the increase was faster than T^{-2}. The susceptibility showed a maximum near 54 mK. These results were interpreted as associated with a cooperative antiferromagnetic nuclear ordering.

Our results for the heat capacity between 0.3 and 10 K are shown in Fig. 1. To provide a rough estimate of the lattice and electronic heat capacities, we have also shown the heat capacity of $LaCu_2$. (The structures of $LaCu_2$ and $PrCu_2$ differ[2], but are related by a small distortion,[3] and the density of $LaCu_2$ is approximately 10% lower[2] than that of $PrCu_2$). The 7 K anomaly is somewhat higher and sharper than reported by Andres et al.[1] and, as shown in Fig. 1, it is much sharper than a two-level Schottky anomaly. The shape of the anomaly shows that it cannot be produced by fixed crystal-field levels, but suggests instead that it arises from a cooperative transition. Similar anomalies have been observed, for example, in Tm AsO_4[4] and $TmVO_4$[5], and attributed to Jahn-Teller distortions. In each of these cases the data are accurately reproduced by a molecular field calculation. In the present case, the transition is somewhat broadened, possibly by strains and inhomogeneities, and we have introduced a gaussian distribution of critical temperatures into the calculation to account for this. The solid curve in Fig. 1 represents a molecular field calculation for the case of two levels that are degenerate (or nearly degenerate) at high temperatures, but with the degeneracy lifted by a Jahn-Teller distortion. The mean critical temperature is 7.5 K and the half width in critical temperature is 0.35 K. Fig. 1 shows that there must be an additional contribution to the heat capacity at temperatures near 8 K and above, and our data at higher temperatures suggests that it persists to above 20 K. A third crystal-field state that produces a Schottky anomaly with a peak near 12 or 13 K would give a contribution of the correct magnitude. Thus, although we have not made a detailed calculation, it seems probable that the crystal-field heat capacity between 1 and 20 K is the sum of that associated with a Jahn-Teller distortion involving the two lowest states, which have a separation $\Delta/k \leqslant 7K$ at $T \gtrsim 7K$ and $\Delta_\circ/k \approx 2T_c = 15K$ at 0K, and that arising from a third state near 25 K.

Our heat capacity results below 1 K are shown in Fig. 2. The zero-field results are similar to those of Andres et al.,[1] but they are lower in magnitude--only 60% as great at 0.1 K--and show a weaker temperature dependence--only the few points below 80 mK increase as rapidly as T^{-2}. For $0.3 \leqslant T \leqslant 0.6$ and $0 \leqslant H \leqslant 20$ kOe, our results are represented by $C = 0.029\,T + (0.003 + 7.94 \times 10^{-5} H^2)\,T^{-2}$ J/mole K, where H is the field in kOe. In this region it is reasonable to associate the field-dependent term with a hyperfine-enhanced nuclear contribution,

$$C = R\,[(I + 1) / 3I][(1 + K)\,\mu H/k\,T]^2.$$

This gives 1 + K = 29 ± 1 for the Knight shift. This value is in good agreement with that obtained by Andres et al.[1], 1 + K = 32 ± 10, from an analysis of the susceptibility.

In summary, our interpretation of the crystal-field heat capacity differs from that given in Ref. 1, but is consistent with a singlet ground state for the Pr^{+3} ions at $T \ll 7K$. Our value for 1 + K agrees with that derived from susceptibility measurements to within the uncertainty of that value. Our measurements show no indication of a cooperative nuclear ordering, but they do not extend to the temperature of the susceptibility maximum.

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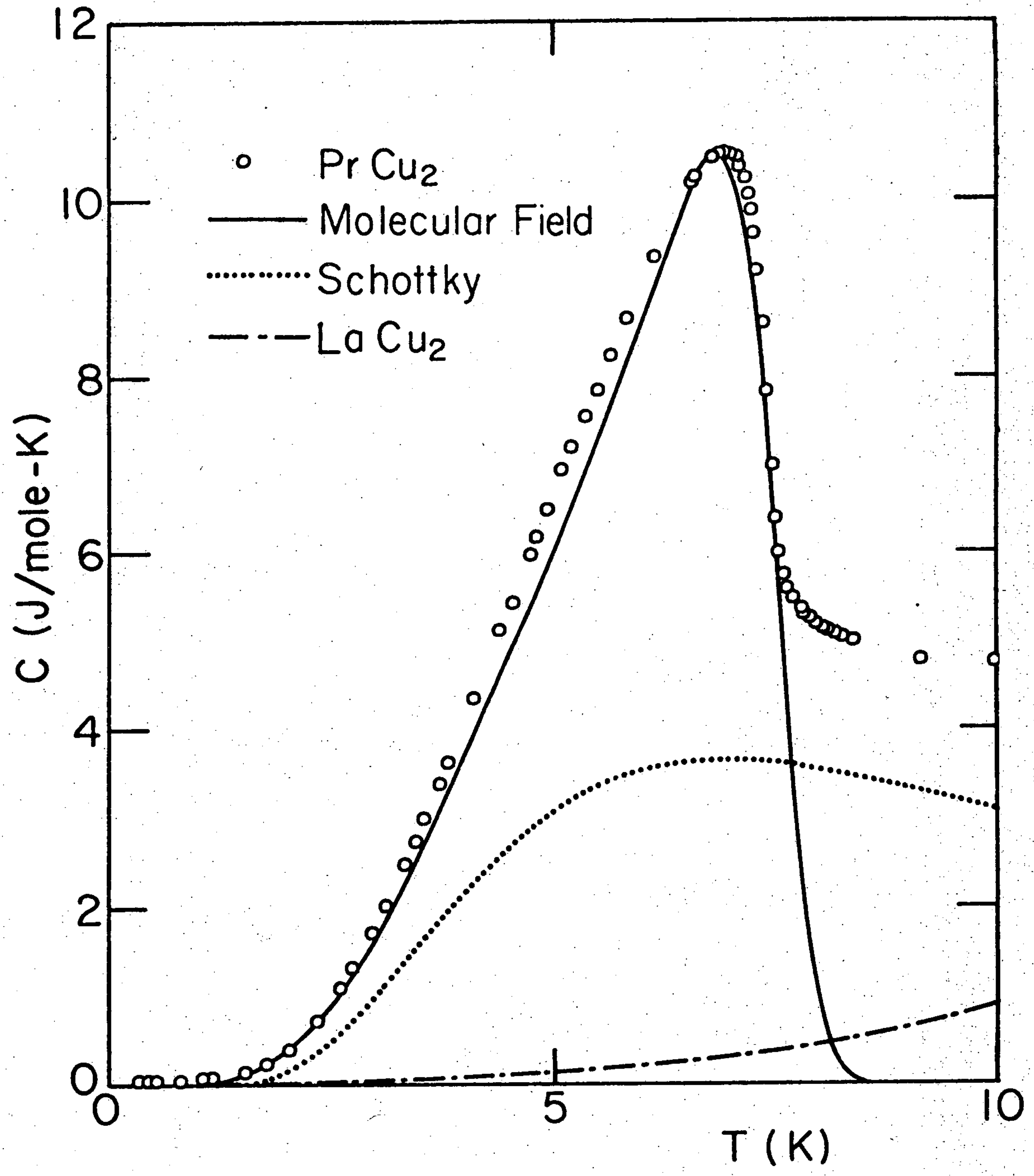

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Fig. 1. The zero-field heat capacity of $PrCu_2$ between 0.3 and 10K.

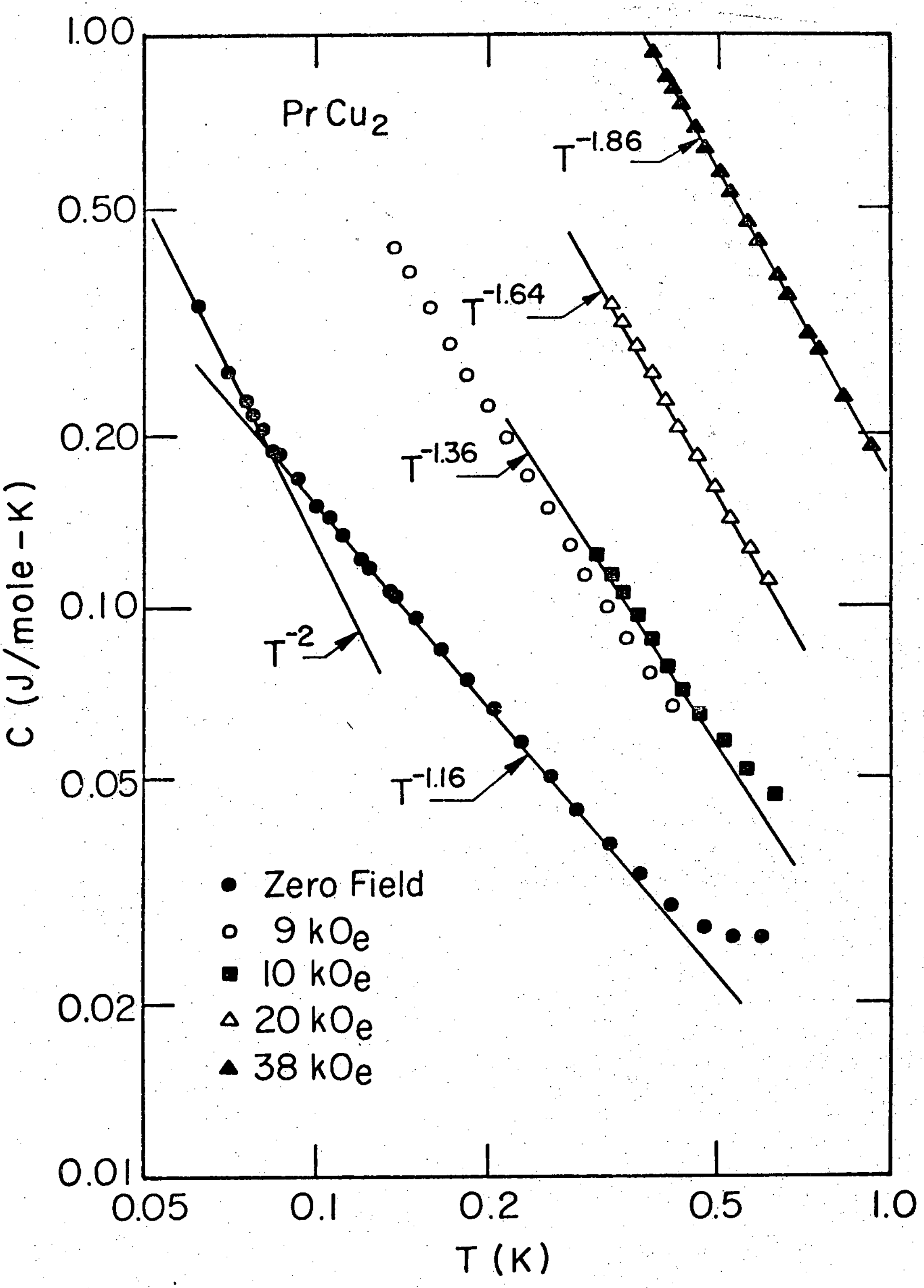

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Fig. 2. The heat capacity of $PrCu_2$ at temperatures below 1K.

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