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Specific Heat of (Y{sub 1-x}Pr{sub x})Ba{sub 2}Cu{sub 3}O{sub 7}; Magnetic Ordering; Magnetic Hyperfine Fields

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# Materials & Chemical Sciences Division

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### Specific Heat of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ ; Magnetic Ordering; Magnetic Hyperfine Fields

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#### SPECIFIC HEAT OF (Y<sub>1-x</sub>Pr<sub>x</sub>)Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>: MAGNETIC ORDERING; MAGNETIC HYPERFINE FIELDS

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The specific heat of  $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$  has been measured for x=0, 0.1, 0.2 and 0.3, 0.3 $\leq T \leq 120$ K and H=0 and 7T. The coefficient of the linear term is  $\gamma = 200$  mJ/mole Pr·K<sup>2</sup>. The Pr ions are characterized by a previously unrecognized <u>low-temperature</u> ordering, and an effective hyperfine field that is enhanced by an applied field.  $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$  has the orthorhombic structure, but is superconducting only for  $x \le 0.6$ , with  $T_c(x)$  following the Abrikosov-Gorkov relation for pair breaking by magnetic moments (1,2). Earlier measurements of the specific heat (C) were interpreted as showing very high values of the coefficient ( $\gamma$ ) of the "linear" term  $\gamma$  T, reaching, in the case of high values of x,  $\gamma = 750$  mJ/mole Pr·K<sup>2</sup> (3-5). As reported here, extension of the measurements to lower temperatures and to magnetic fields has shown that magnetic ordering contributed to C in the temperature interval in which  $\gamma$  was determined, and the values of  $\gamma$  that appear to be relevant to the conduction electron system are, although still high, substantially lower. The measurements also give values of the effective hyperfine field (H<sub>h</sub>) at the Pr nuclei.

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In Fig. 1 (x=0 and 0.3) magnetic ordering for x=0.3 is evident from the broad Schottkylike anomalies near 1 and 5K, respectively, for H=0 and 7T. The low temperature upturns in C/T are indicative of hyperfine contributions, primarly from <sup>141</sup>Pr. Although Meissner effect measurements showed a transition to the superconducting state at  $T_c$ =42K (2), no obvious discontinuity was observed in C.

A least-squares fit of the 7-T data by  $C(7T) = D(7T)T^2 + \gamma(7T)T$ , shown in Fig. 2, gives  $\gamma(7T) = 9.0, 32, 47$  and 66 mJ/mole·K<sup>2</sup> and D(7T) = 0.26, 9.4, 15.7 and 29.7 mJ·K/mole for x=0, 0.1, 0.2 and 0.3, respectively, and H<sub>h</sub>(7T) = 100±5T. The values of  $\gamma(7T)$  are well represented by  $\gamma(7T) = 9+200x$  mJ/mole·K<sup>2</sup>; determination of  $\gamma(0)$  is precluded by the Schottky anomalies except for x=0, in which case  $\gamma(0) = 7.6$  mJ/mole·K<sup>2</sup>.

The values  $H_h(H)$  derived from D(H), are shown for x = 0.3 in Fig. 3. The zero-field value is 44T, and there is a strong non-linear enhancement with increasing H.  $H_h(7T)$  is essentially independent of x; the field dependence was not determined for other values of x. At least

qualitatively,  $H_h(7T)$  is consistent with either a  $Pr^{4+}$ ,  $J_z = \pm 1/2$  ground state and low-lying states of higher  $J_z$ , or a  $Pr^{3+}$  singlet ground state with exchange-induced mixing of higher states.

Figure 4 shows the magnetic ordering and electronic contributions to C for x=0.3, obtained by subtracting the hyperfine contribution and (for the "background" contribution) the total specific heat for x=0. The upper horizontal line is the value of  $\gamma(7T)$  determined below 1K, and the lower horizontal line represents the high-temperature limiting value. The temperature dependence of the electronic contribution, represented in the low- and high-temperature limits by these lines, is indeed reminescent of the heavy-fermion behavior that was suggested (6) on the basis of high  $\gamma$  values. The small feature near 42K may be associated with the transition to the superconducting state. The entropies associated with the magnetic ordering anomalies are ( $\frac{1}{2}$ )Rln2, based on moles of Pr. The same values were obtained for x=0.1 and 0.2.The insert to Fig. 4 illustrates the origin of higher  $\gamma$  values when the usual C/T vs T<sup>2</sup> plot is used without taking account of the magnetic ordering. Fig. 5 shows the anomaly in C/T near T<sub>c</sub>=86K for x=0.1 which is very much attenuated compared with those observed for x=0.

The work at LBL was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Dept. of Energy under contract DE-AC03-76SF00098, and at LLNL and Davis under the auspices of the U.S. Dept. of Energy under contract W-7405-ENG-48. Additional support for A.A. was provided by a grant from the Swiss National Science Foundation.

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## FIGURE 1

C/T vs T for x=0 and 0.3





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 $H_h$  vs H for x=0.3



FIGURE 4

Magnetic ordering and electronic contributions to C.

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FIGURE 5

C/T vs T for x=0.1

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