

# Effect of pressure on the specific heat of $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$

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Measurements of the specific heat of  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  under pressure have been carried out over the temperature interval 2–4 K. It has been observed that  $T_N$  increases from 2.23 to 2.265 K under a pressure  $\sim 5$  kbar, with  $dT_N/dP = 7 \times 10^{-6}$  K/bar.

It was reported previously that measurements of the specific heat in the  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  system revealed a maximum at a temperature of 2.23 K, which was apparently associated with a transition of the compound to an antiferromagnetic state (Ref. 1; see also Refs. 2 and 3). At  $T < T_N = 2.23$  K the sample remained superconducting. Consequently, a coexistence of superconductivity and an antiferromagnetic order is observed in  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$ . There is considerable research interest in the mutual effects of superconducting and magnetic subsystems and also in the nature of the magnetic ordering for high-temperature superconductors, such as  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$ . Important information can be found by carrying out measurements under pressure (Refs. 4 and 5, for example). In the present letter we report measurements of the specific heat of the compound  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  under pressure over the temperature interval  $2 \text{ K} < T < 4 \text{ K}$ , i.e., near  $T_N$ . The specific heat was measured in the calorimeter described in Ref. 6. For the measurements under pressure, a small, high-pressure bomb of a special nonmagnetic beryllium bronze was designed and constructed. The empty bomb had a mass of about 52 g. An estimate of the concentration of paramagnetic impurities from the results of the measurements of the heat capacity of the empty bomb showed that their concentration did not exceed  $10^{-2}$  at. %. As the medium we used a mixture of kerosene and transformer oil. The pressure was measured on the basis of the shift of  $T_c$  of the tin pressure gauge.

A  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  sample, synthesized by a procedure like that of Ref. 1, consisted of essentially a single phase. The mass of the sample during the measurements under pressure was 0.355 g. To determine the specific heat of the sample, we measured the heat capacity of the bomb containing the sample under pressure and the heat capacity of the empty bomb. Figure 1 shows the results of the measurements of the specific heat of the  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  sample at standard pressure and at a high pressure. The inset shows the temperature dependence of the heat capacities of the bomb with the sample under pressure and of the empty bomb. We see that at  $T = T_N = 2.265$  K the heat capacity of the sample is nearly an order of magnitude greater than that of the bomb. It follows from this figure that the temperature at which the specific heat reaches a maximum increases under pressure and has the value  $T_N = 2.265$  K at a pressure of about  $^1) 5$  kbar (at  $P = 0$  the corresponding temperature

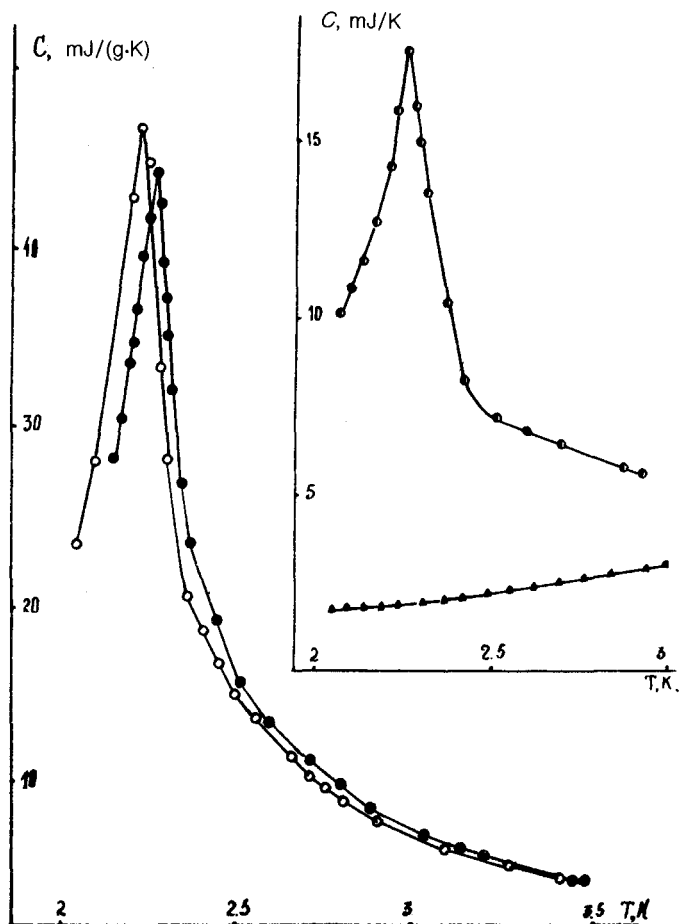


FIG. 1. ○—Specific heat of  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  at standard pressure; ●—specific heat of  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  at a pressure  $\sim 5$  kbar; ◐—heat capacity of the bomb containing the sample; ▲—heat capacity of the bomb without the sample.

is  $T_N = 2.23$  K). The relative error in the determination of  $T_N$  does not exceed 0.01 K here. The imposition of a pressure did not cause a broadening of the peak in the specific heat. The time scale of the relaxation to a thermal equilibrium for the sample with the bomb in the calorimeter did not exceed the corresponding time for the sample without the bomb. The 0.035-K shift of the specific-heat maximum up the temperature scale is evidence of an increase in the temperature of the magnetic ordering in the compound  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  under pressure. An estimate of the entropy of the transition under pressure yields a value in good agreement with the entropy of a transition without an applied pressure.

The magnetic entropy found from the specific heat is 11.7 J/(mole·K), or about

70% of the quantity

$$\Delta S_m = R \ln(2J + 1) = 16.7 \text{ J/(mole} \cdot \text{K)}, J = 7/2,$$

in other words, at least 70% of the sample goes into a magnetically ordered state.

The present study thus reveals an increase in the temperature of the antiferromagnetic ordering of  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  as the pressure is raised. The value of the derivative  $dT_N/dP$  is  $7 \times 10^{-6}$  K/bar. We should point out that as the pressure is raised, the superconducting transition temperature  $T_c$  of the compound  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  also increases. In a system with the magnetic rare-earth ion gadolinium the increase in  $T_c$  under pressure occurs more rapidly than in the system with the nonmagnetic ion yttrium.<sup>7</sup>

Opinion is divided on the nature of the ordering in the  $\text{REBa}_2\text{Cu}_3\text{O}_{7-x}$  compounds. Comparing the values of  $T_N$  of 1–2–3 compounds with various rare earths, Ramirez *et al.*<sup>2</sup> and Van der Menlen *et al.*<sup>3</sup> conclude that the interaction between the magnetic RE ions in these compounds is predominantly of an exchange nature. On the other hand, several papers, e.g., Ref. 8, point out that it is possible to explain the ordering in the  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  system in terms of a purely dipole-dipole interaction between gadolinium ions. If, by analogy with the approach taken for rare-earth calco-genides of molybdenum,<sup>4,5</sup> we estimate the value of the derivative  $|\partial \ln T_N / \partial \ln V| = |\kappa^{-1} d \ln T_N / dP|$  from the results of the present study, and if we use the compressibility value<sup>9</sup>  $\kappa = 0.65 \times 10^{-6} \text{ bar}^{-1}$ , we find a value  $\sim 5$ . On the other hand, the value of this derivative for the case of a dipole-dipole interaction should be close to 1, since in this case we have<sup>4</sup>  $T_N \sim \mu_{\text{eff}}^2 / \alpha^3 \sim \mu_{\text{eff}}^2 / V$ . It can thus be concluded from the results of these measurements of the specific heat of  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  under pressure that the interaction which leads to the antiferromagnetic ordering of the gadolinium ions in this compound is of an exchange nature. The results of measurements of the ESR of  $\text{Gd}^{3+}$  provide further evidence in favor of an exchange nature of the interaction.<sup>11</sup>

<sup>1</sup>The specific heat of the compound  $\text{GdBaSrCu}_3\text{O}_{7-x}$  at standard pressure was also measured in this study. For this compound we observed a significant broadening of the  $\lambda$  transition. In contrast with Ref. 10, the temperature at which the maximum is reached shifts downward:  $T_N \sim 2.1$  K.

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