

# Low-Temperature Specific Heat of overdoped Bi2201 Single Crystals

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## Abstract

Low-temperature specific heat of heavily overdoped  $\text{Bi}_2\text{Sr}_2\text{CuO}_6$  single crystals was studied. The specific heat under zero magnetic field included a linear- $T$  term, suggesting the presence of a large pair breaking effect, which we attribute to impurities because their influence on  $d$ -wave superconductors increases when the energy gap decreases with carrier over-doping. The field dependence of the electronic specific heat followed well the prediction for an impure  $d$ -wave superconductor.

*Key words:* specific heat;  $d$ -wave symmetry; pair breaking; impurity effect

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It is now more or less established that the order parameter of high temperature superconductors has a  $d$ -wave symmetry. The properties of high temperature superconductors, however, depend strongly on the carrier density. We think that the heavily overdoped region, where the superconductivity is fading out by approaching a paramagnetic metal phase, needs an especially careful investigation, if spin fluctuation is critical for the appearance of high temperature superconductivity with a  $d$ -wave symmetry. Moller *et al.* were the first who showed experimentally that the field dependence of electronic specific heat helps to distinguish the symmetry of a superconductor [1,2]. Here, we report the low-temperature specific heat of very heavily overdoped  $\text{Bi}_2\text{Sr}_2\text{CuO}_6$  (Bi2201) single crystals.

The crystals were grown by the floating zone (FZ) method using sintered powders with a nominal composition of  $\text{Bi}_{1.74}\text{Pb}_{0.38}\text{Sr}_{1.88}\text{CuO}_y$ . The carrier density was carefully adjusted by annealing the crystals under various atmospheres at different temperatures. The specific heat was measured using a relaxation method (Oxford Instruments MLHC9H) down to 0.5 K. The addenda were carefully determined by measuring first the heat capacity of the substrate with a small amount of grease. The sample was then mounted on the sub-

strate without adding or removing any of the grease. The addenda measurement was performed for every sample and every magnetic field.

Figure 1 shows the specific heat of one of the Bi2201 single crystals measured with 0 and 6 T. The superconducting transition temperature ( $T_c$ ) of this sample was 19 K. First of all, it is very obvious that the extrapolation of the zero-field data to  $T=0$  gives a non-zero value, implying a contribution from a linear- $T$  term to the specific heat at  $H=0$ . This is unexpected for a superconductor regardless of the symmetry, and suggests the presence of a large pair-breaking effect. It should be noted here that even non-magnetic impurities can cause pair-breaking in a  $d$ -wave superconductor, and their influence increases when the ratio of the gap energy to the density of impurities decreases [3]. Therefore, the effect of impurities cannot be neglected when the gap becomes small in the heavily overdoped region. In a separate paper, we will make a quantitative estimation based on the pair-breaking effect observed for samples with various doping levels, which shows that at least a large part of the pair-breaking effect can be explained by the impurities that are unavoidably included in the crystals because of the purity of the starting materials.

As shown in Fig. 1, applying a magnetic field parallel to the  $\text{CuO}_2$  plane had little effect on the specific

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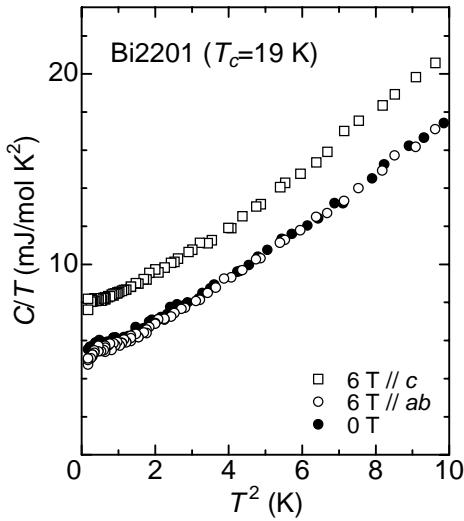


Fig. 1. Specific heat  $C/T$  versus  $T^2$  under zero magnetic field and 6 T with  $H \parallel c$  and  $H \parallel ab$ .

heat, in contrast to the reported data of  $\text{YBa}_2\text{Cu}_3\text{O}_y$  [1,2]. This observation indicates the smallness of the Schottky anomaly due to magnetic impurities in our samples. On the other hand, the specific heat increased when a magnetic field was applied parallel to the  $c$ -axis direction, the field dependence of which is determined by the symmetry of the order parameter. The data were fitted to the following equation:

$$C(T) = (\gamma_0 + \gamma(H))T + \beta T^3 + \delta T^5 + C_{\text{Sch}}(T). \quad (1)$$

Here, the first term expresses the electronic specific heat, the second and third terms the phonon contribution, and the last term is the Schottky anomaly often observed for high temperature superconductors [1,2].

For a  $d$ -wave superconductor with line of nodes, a term proportional to  $T^2$  is expected for  $H=0$ . However, including a term proportional to  $T^2$  in Eq. (1) and fitting it to the zero-field data resulted in a negative coefficient for the  $T^2$  term, indicating that there is no contribution that is proportional to  $T^2$ . The  $T^2$  term is expected to be non-zero for a pure  $d$ -wave superconductor because the density of states is linear to the energy in the vicinity of Fermi level. If there is a pair-breaking effect, however, the density of states is different, and the contribution of low-energy excitations to the specific heat is no more depending on  $T^2$ . Therefore, the absence of the  $T^2$  term is actually consistent with the observation of a large pair-breaking effect.

The field dependence of  $\gamma(H)$  was obviously not a linear function of the field, implying that the symmetry of the order parameter is not  $s$ -wave. While Volovik showed that in a clean  $d$ -wave superconductor with line of nodes  $\gamma(H) \sim \sqrt{H}$  [4], the following expression was derived for an impure superconductor in the unitarity limit [3].

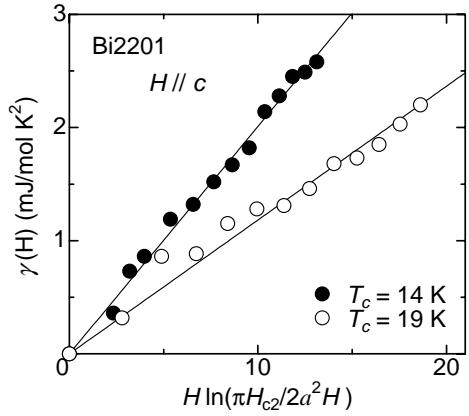


Fig. 2. The field dependent part of the coefficient of electronic specific heat plotted as a function of  $H \ln(\pi H_{c2}/2a^2 H)$ .

$$\frac{\gamma(H)}{\gamma_n} = \frac{a^2}{4.88} \sqrt{\frac{\Delta_0}{\Gamma}} \frac{H}{H_{c2}} \ln \left[ \frac{\pi}{2a^2} \frac{H_{c2}}{H} \right] \quad (2)$$

Here,  $a=0.465$  for a triangular vortex lattice,  $\Gamma$  is the impurity scattering rate proportional to the concentration of impurities,  $\Delta_0$  the gap maximum,  $H_{c2}$  the upper critical field, and  $\gamma_n$  the coefficient of electronic specific heat in the normal state.

Figure 2 shows  $\gamma(H)$  as a function of a quantity proportional to the right hand side of Eq. (2). The data of a crystal with  $T_c=14$  K is also indicated in the same figure. The  $H_{c2}$  values used for the plots are 18.4 T and 7.36 T for the samples with  $T_c=19$  K and 14 K, respectively, which were determined by fitting Eq. (2) to the data. The plots in Fig. 2 are both fairly well a linear function. In fact, fitting Eq. (2) to the data of  $\gamma(H)$  gave a better result than using the prediction of Volovik for a pure  $d$ -wave superconductor. Therefore, we conclude that both the temperature dependence of the zero-field data and the field dependence of the coefficient of electronic specific heat are consistent with a  $d$ -wave symmetry, and a large pair-breaking effect exists for which impurities that are unavoidably included in the crystals are probably playing an important role.

The authors are indebted to Prof. Y. Tanaka of Nagoya University for fruitful discussion.

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