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Specific heat in the mixed state of superconducting $YBa_2Cu_3O_{7-x}$

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It is shown that the observed magnetic-field dependence of the specific-heat anomaly near the superconducting transition temperature T_c of $YBa_2Cu_3O_{7-x}$ can be understood quantitatively by the Ginzburg-Landau theory of anisotropic type-II superconductors. It is shown, in the London limit, that the change in specific heat by the application of magnetic field B_a (~ 1 T) is approximately linear in $(B_a/T_c)[t/(1-t)]$ for temperatures slightly below T_c, where $t=T/T_c$. It is also predicted that the slope of this linear behavior for $\mathbf{B}_a \perp c$ axis should be $(m_c/m_{ab})^{1/2}$ times lower than that for $\mathbf{B}_a \parallel c$ axis. Here, m_c and m_{ab} are the components of the effective-mass tensor parallel and perpendicular to the c axis, respectively.

The effect of magnetic fields on the specific-heat (C) anomaly near the superconducting transition temperature T_c of the high-T_c superconductors (HTSC) is considerably different from the effect on conventional superconductors.[1-3] These specific-heat measurements in HTSC have been done in applied fields ($\mathbf{B}_a$) of the order of 1 T. It has been found that the specific-heat peak[4] associated with the superconducting transition mainly reduces in amplitude and broadens by the application of magnetic field. This is unlike the conventional type-II superconductors, in which case, the application of magnetic fields shifts the specific-heat jump at T_c to lower temperatures, with practically very little change in the shape of the anomaly.[5] The origin of this unusual behavior in the HTSC is not yet clear.

Salamon *et al.*[2] have measured the specific heat of a $YBa_2Cu_3O_{7-x}$ single crystal, in fields of 1.5–6.0 T applied parallel to the c axis. They have found that the change in specific heat by the application of a magnetic field, $\Delta C = C(0,T) - C(\mathbf{B}_a,T)$, is a function of temperature which shows a peak a few kelvins wide, near T_c. Only the low-temperature side of the peak was found to shift appreciably to lower temperatures by increasing B_a. They also found that ΔC satisfies a scaling relation, which suggests critical fluctuations. They have interpreted this in terms of a broadening of the critical region by the application of magnetic fields. The values of the scaling exponents obtained are, however, unusual. In order to account for the unusual values of the scaling exponents, they have suggested possible renormalization of the critical exponents either by disorder or else by the presence of an order parameter with more than two components. In this context, we note that it is difficult to distinguish, with the available accuracy of the specific heat data,[6] the effect of inhomogeneity from that of intrinsic fluctuations.

Recently, de Jongh[7] has provided another viewpoint to explain the field-dependent broadening of the specific-heat anomaly and other quantities, such as the resistivity and the nuclear-spin-lattice relaxation time. It has been argued that the HTSC can be considered as a system of weakly coupled superconducting layers (represented by the Cu-O layers), belonging to the same universality class as the XY-model with nearest-neighbor interactions. When cooled in the absence of a magnetic field, the average size of the correlated two-dimensional (2D) superconducting regions diverges and eventually long-range 3D superconducting order is obtained. However, for $\mathbf{B}_a \parallel c$ axis, the vortices in the mixed state reduce the interlayer coupling, which gives rise to an assembly of superconducting regions of finite extent along the c axis. The establishment of long-range 3D superconducting order is then prevented and the superconductor shows strong 2D fluctuations at all temperatures. Whereas, for $\mathbf{B}_a \perp c$ axis, the magnetic flux just penetrates between the superconducting layers, since the coherence length along the c axis is much smaller than the interlayer separation. It is thus argued that the broadening of the transition should be absent for $\mathbf{B}_a \perp c$ axis.

These explanations, as they stand at present, can only be considered heuristic. In this Rapid Communication, we show that the field dependence of the specific heat near T_c can be understood quantitatively, using the Ginzburg-Landau (GL) theory of type-II superconductors.[8]

The HTSC are type-II superconductors, having small lower critical field B_{c1} ($\sim 10^{-2}$ T at $T=0$) and high upper critical field B_{c2} ($\sim 10^2$ T at $T=0$). These superconductors, therefore, remain in the mixed state, for $B_a \sim 1$ T. (Hereafter, we will only consider $B_a \sim 1$ T.) Moreover, the intervortex separation (L) in the mixed state satisfies the condition $\xi \ll L \ll \lambda$ over a wider temperature range near T_c compared to conventional superconductors. (Here, ξ and λ are the coherence length and the London penetration depth, respectively.) This is due to the large GL parameter $\kappa = \lambda/\xi (\sim 10^2)$ and the high $T_c (\sim 100$ K) of the HTSC. The comparison of L with ξ and λ, given in Table I, for $YBa_2Cu_3O_{7-x}$, is seen to support this situation (except for t close to 0.99 and $B_a \simeq 6$ T). For $\mathbf{B}_a \parallel c$ axis, the ab-plane lengths λ_{ab} and ξ_{ab} come into the picture, whereas for $\mathbf{B}_a \perp c$ axis, the c-axis lengths λ_c and ξ_c are also relevant. It is generally believed that the HTSC are in the clean limit,[9] mainly because ξ is of the order of the lattice constant. Therefore, in Table I, we have used the following clean-limit GL temperature dependences[8] of ξ and λ, which are valid near T_c:

$$\xi(T) = 0.73\xi(0)(1-t)^{-1/2}, \qquad (1)$$
$$\lambda(T) = 0.71\lambda(0)(1-t)^{-1/2},$$

TABLE I. Comparison of L with ξ and λ in $YBa_2Cu_3O_{7-x}$.

T/T_c	T (K)	ξ_{ab} (Å)	λ_{ab} (Å)	ξ_c (Å)	λ_c (Å)	L (Å) $B_a = 1$ T	2 T	4 T	6 T
0.99	89.1	117	9940	22	31950				
0.98	88.2	83	7029	15	22592	455	322	227	186
0.95	85.5	52	4445	10	14288				
0.90	81.0	37	3143	7	10103				

where $t = T/T_c$. We have used $\xi_{ab}(0) = 16$ Å, $\xi_c(0) = 3$ Å,[10] $\lambda_{ab}(0) = 1400$ Å,[11] $\lambda_c(0) = 4500$ Å,[12] and $T_c = 90$ K. The intervortex separation is obtained from

$$L = (\phi_0/B_a)^{1/2}, \tag{2}$$

where $\phi_0 \simeq 2.07 \times 10^{-15}$ Wb is the flux quantum. Equation (2) gives a good estimate of L in the HTSC, since $B_{c1} \ll B_a$. The GL theory in the London limit provides the appropriate description of the mixed state, when $\xi \ll L \ll \lambda$.[8] The London limit, although it does not treat the vortex cores correctly, is expected to give reliable results since the contribution from the vortex cores ($\propto \xi^2$) constitutes only a small fraction of the total energy in case of the HTSC. Further, we assume the validity of the 3D description of the GL theory, which is justified for temperatures close to T_c.[13] In the following, we will show that the field dependence of the specific heat near T_c can be understood quantitatively in terms of the specific heat of the mixed state in the London limit. For simplicity, we will ignore the effect of thermal fluctuations of the vortex positions.[14] We will make use of the anisotropic formulation of the London limit,[15] since HTSC show large anisotropy in ξ and λ. The twins in the ab plane, however, (especially in the case of $YBa_2Cu_3O_{7-x}$) average out the small anisotropy among the a and b axes.[16] We will, therefore, assume anisotropy only between the c axis and the ab plane. Following Campbell, Doria, and Kogan[17] we write the increase in the free-energy density by the application of magnetic field as

$$\begin{aligned}\Delta F &= F(\mathbf{B}_a,T) - F(0,T) \\ &= B^2/2\mu_0 + \alpha\lambda_m^{-2}(m_{ab}B_x^2 + m_c B_z^2)^{1/2}\ln(B_{c2}/B),\end{aligned} \tag{3}$$

where $\alpha = \phi_0/8\pi\mu_0$. $B_z = B\cos\theta$ and $B_x = B\sin\theta$ are the components of the magnetic induction ($\mathbf{B}$) parallel and perpendicular to the c axis, respectively. θ is the angle between the c axis and $\mathbf{B}$. m_{ab} and m_c are the components of the dimensionless effective-mass tensor and can be defined in terms of the London penetration depths as follows:

$$m_{ab} = \lambda_{ab}^2/\lambda_m^2 \text{ and } m_c = \lambda_c^2/\lambda_m^2, \tag{4}$$

where the mean penetration depth $\lambda_m = (\lambda_{ab}^2\lambda_c)^{1/3}$. As usual, the angular dependence of B_{c2} has been ignored in the derivation of Eq. (3) for ΔF.[17]

The magnetic field $H = (\partial\Delta F/\partial B)$ has been shown to be given by[17]

$$\begin{aligned}H_z &= B_z/\mu_0 + \alpha\lambda_m^{-2}(m_c/m^{1/2})(B_z/B)\ln(B_{c2}/B), \\ H_x &= B_x/\mu_0 + \alpha\lambda_m^{-2}(m_{ab}/m^{1/2})(B_x/B)\ln(B_{c2}/B),\end{aligned} \tag{5}$$

where $m = m_{ab}\sin^2\theta + m_c\cos^2\theta$. Here, H_z and H_x are the components of $\mathbf{H}$ parallel and perpendicular to the c axis, respectively. The prefactor of the natural logarithm is of the order of B_{c1}/μ_0, which is very small compared to $H(=B_a/\mu_0)$. Hence, $\mathbf{B}$ is almost parallel to $\mathbf{B}_a$, and θ can be taken as the angle between $\mathbf{B}_a$ and the c axis. The change in the Gibbs-free-energy density (ΔG) can now be obtained using Eqs. (3) and (5) as

$$\Delta G = \Delta F - BH = -B^2/2\mu_0. \tag{6}$$

Finally, the change in the specific heat (ΔC), by the application of magnetic field can be obtained from

$$\Delta C = C(0,T) - C(\mathbf{B}_a,T) = T(\partial^2\Delta G/\partial T^2). \tag{7}$$

Evaluation of ΔC using Eq. (7) involves only the temperature derivatives of the magnetization M since $(\partial H/\partial T) = 0$. To this end, we make the following approximation. We replace B by B_a in the argument of the natural logarithm in Eq. (5). This is justified, since $\ln(\cdots)$ is a slowly varying function of its argument and the prefactor of ln is very small compared to H. The components of the magnetization parallel (M_z) and perpendicular (M_x) to the c axis can then be written as

$$\begin{aligned}M_z &= -\alpha\lambda_m^{-2}(m_c/m^{1/2})\cos\theta\ln(B_{c2}/B_a), \\ M_x &= -\alpha\lambda_m^{-2}(m_{ab}/m^{1/2})\sin\theta\ln(B_{c2}/B_a).\end{aligned} \tag{8}$$

For $\mathbf{B}_a \parallel c$ axis, $\theta = 0$, $m = m_c$, and $M_x = 0$; and for $\mathbf{B}_a \perp c$ axis, $\theta = \frac{1}{2}\pi$, $m = m_{ab}$, and $M_z = 0$. We first consider the case with $\mathbf{B}_a \parallel c$ axis. So we use $M = M_z$ in Eq. (8) with $\theta = 0$ and the following GL temperature dependences,[8]

$$\begin{aligned}\lambda_m(T) &= \left[\frac{\lambda_m(0)}{\sqrt{2}}\right](1-t)^{-1/2}, \\ B_{c2}(T) &= B_{c20}(1-t),\end{aligned} \tag{9}$$

valid near T_c, in Eq. (7) for ΔC. We obtain

$$\Delta C = Q(B_a/T_c)[t/(1-t)] + O[(L/\lambda_m)^4], \tag{10}$$

where

$$Q = 2\alpha m_c^{1/2}/\lambda_m^2.$$

The maximum value of t in Eq. (10) is limited by $B_a = B_{c20}(1-t)$. Hence divergence of ΔC for $t = 1$ does not occur. The first term of Eq. (10) increases rapidly as $T \to T_c$ and gives rise to the apparent broadening of the specific-heat anomaly in the presence of $\mathbf{B}_a$. The magnitude of the second term of Eq. (10) cannot be estimated reliably since the free-energy expression of Eq. (3) is accurate only to $O[(L/\lambda_m)^2]$. We can consider ΔC to be linear in $(B_a/T_c)[t/(1-t)]$. We now estimate Q using typical values of quantities appropriate for $YBa_2Cu_3O_{7-x}$. We take $\lambda_{ab}(0)=1400$ Å,[11] and $\lambda_c(0)=4500$ Å,[12] and calculate $\lambda_m(0)=2066$ Å, $m_{ab}=0.46$, and $m_c=4.7$. This gives $Q=6.66$ kJ/m^{3}T. For comparison, we have plotted the experimental data of Salamon *et al.*[2] as ΔC vs $(B_a/T_c)[t/(1-t)]$ in Fig. 1. ΔC is seen to be linear in $(B_a/T_c)[t/(1-t)]$, within the scatter of the original data. The linearity is maintained up to about $t=0.985$ and on the low-temperature side, the data merges in the noise at $t \simeq 0.97$. The data for $B_a=6$ T shows a trend quite different from the rest. This can be traced back to the original data, in which ΔC vs T for $B_a=4.5$ and 6.0 T practically coincide. A possible reason can be that $B_{c2} \lesssim 6$ T in this temperature range. We now estimate Q from the data of Salamon *et al.*[2] For this purpose, we take the data for $B_a=3.0$ T as the representative (Fig. 1) and use the least-squares method to fit a straight line in the range 0.74 T/K $< (B_a/T_c)[t/(1-t)] <$ 2.74 T/K (equivalent to $0.95 < t < 0.99$, with $B_a=3.0$ T and $T_c=90.83$ K). The fit gave $Q=7.20 \pm 0.21$ kJ/m^{3}T. This is in good agreement with the theoretically calculated value of 6.66 kJ/m^{3}T. It therefore brings out the main point: the temperature dependence of ΔC can be quantitatively accounted for, solely in terms of the specific heat of the mixed state.

It is seen that there is a systematic deviation from linearity for higher values of $(B_a/T_c)[t/(1-t)]$ or t (Fig. 1). This is due to the fact that the London limit does not hold when the vortex cores occupy much of the volume (i.e., $B_a \simeq B_{c2}$). In such a situation, it can be shown from the GL theory that $(\partial C/\partial B_a)=0$.[8] The maximum value of ΔC should be limited by the specific-heat jump at T_c in the absence of magnetic field, which is about 40 kJ/m^{3}K.[3] It is then possible that the peak in ΔC as a function of temperature arises due to the smeared specific-heat jump at T_c. The negligible shift of the specific-heat peak (or the specific-heat jump at T_c in the case of an ideal transition) by the application of magnetic field is likely due to the large upper critical field. The broadened superconducting transition, due to the sample inhomogeneity and the large field dependence of the specific heat below T_c, can also easily mask the small shifts of the specific-heat peak. As seen before, the large κ combined with the high T_c in the HTSC, gives rise to the large field dependence of the specific heat over a wider temperature interval ($\simeq 4$ K) below T_c, compared to the conventional type-II superconductors. Therefore, the specific-heat peak in the HTSC, appears only to be reduced in amplitude and broadened by the application of a magnetic field, unlike the conventional type-II superconductors.

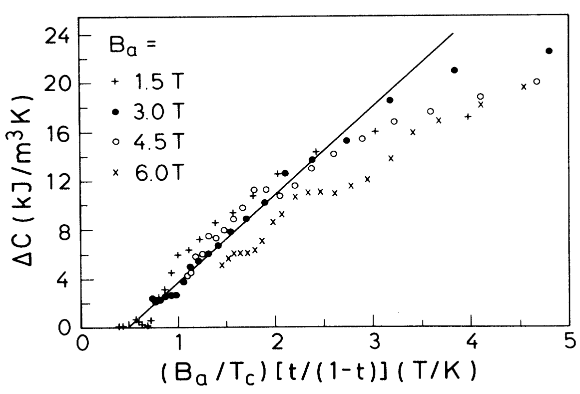

FIG. 1. ΔC vs $(B_a/T_c)[t/(1-t)]$ for various applied fields $\mathbf{B}_a$. The data is obtained from Fig. 2 of Ref. 2 and we have assumed a density of 6.38 g/cm^3 to express ΔC in kJ/m^{3}K. The T_c is 90.83 K. The solid line is the least-squares-fitted straight line through the data for $B_a=3.0$ T, in the range 0.74 T/K $< (B_a/T_c)[t/(1-t)] <$ 2.74 T/K. The slope and the intercept of the fitted line are 7.20 kJ/m^{3}T and -3.52 kJ/m^{3}K, respectively.

We next consider the case with $\mathbf{B}_a \perp c$ axis (i.e., $\theta = \frac{1}{2}\pi$). It can be shown that in this case, one has to only replace m_c by m_{ab} in Eq. (10) to obtain the expression for ΔC. We therefore see that

$$\frac{Q(\mathbf{B}_a \parallel c \text{ axis})}{Q(\mathbf{B}_a \perp c \text{ axis})} = \left(\frac{m_c}{m_{ab}}\right)^{1/2} . \tag{11}$$

Noting that $m_c > m_{ab}$ in $YBa_2Cu_3O_{7-x}$, the field dependence of specific heat near T_c for $\mathbf{B}_a \perp c$ axis should be $(m_c/m_{ab})^{1/2}$ times lower than that for $\mathbf{B}_a \parallel c$ axis. The anisotropy ratio $(m_c/m_{ab})^{1/2}$ is about eight in $YBa_2Cu_3O_{7-x}$.[13] Measurement of specific heat with $\mathbf{B}_a \perp c$ axis has not been carried out so far to our knowledge. However, approximate estimates on grain-aligned $YBa_2Cu_3O_{7-x}$ samples show that the magnetic-field dependence of specific heat for $\mathbf{B}_a \perp c$ axis is about five times lower than that for $\mathbf{B}_a \parallel c$ axis,[18] which is in agreement with our prediction.

In view of our observation that the GL theory satisfactorily explains the magnetic-field dependence of specific heat, we note the opposite conclusion obtained by Reeves *et al.*[19] They have measured the specific heat of sintered $YBa_2Cu_3O_{7-x}$ powder in magnetic fields up to 3 T at low temperatures ($t < 0.1$). It has been found that the GL theory in the London limit fails to completely account for the observed field dependence. Therefore, they have conjectured that the superconductor order parameter does not change slowly over a length characteristic of the atomic structure of this material, leading to the failure of the GL theory. In this context, it is interesting to note that there is strong evidence that a 3D description of the GL theory breaks down below 80 K in $YBa_2Cu_3O_{7-x}$, accompanied by a crossover to a 2D superconducting behavior.[13] This is most likely the reason for the difference in conclusion.

In summary, we have quantitatively explained the observed magnetic-field dependence of the specific-heat anomaly in $YBa_2Cu_3O_{7-x}$ near T_c for $\mathbf{B}_a \parallel c$ axis, using the 3D description of the GL theory in the London limit. The large GL parameter combined with the high T_c in the HTSC, gives rise to the large field dependence of specific heat over a wider temperature interval ($\simeq 4$ K) below T_c, compared to the conventional type-II superconductors. Based on this analysis, we have shown that ΔC is approximately linear in $(B_a/T_c)[t/(1-t)]$ for temperatures slightly below T_c. There is a good agreement between the theoretically calculated value of the slope of this linear behavior and that obtained from experiment. We have also predicted that the slope for $\mathbf{B}_a \perp c$ axis should be $(m_c/m_{ab})^{1/2}$ times lower than that for $\mathbf{B}_a \parallel c$ axis.

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