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Magnetic-field dependence of the low-temperature specific heat of $La_{2-x}Sr_xCuO_4$

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Recently, possible evidence from the low-temperature specific heat (LTSH) for the lines of nodes in the superconducting order parameter of cuprate superconductors has attracted much attention and is still debated. To clarify this issue and to cover the studies in different carrier doping regimes, we have measured LTSH of $La_{2-x}Sr_xCuO_4$ ($x=0.10$, 0.16, and 0.22) both in zero and applied magnetic fields. In all doping regimes, it is found that the increase in the linear-T coefficient γ is proportional to $H^{1/2}$, consistent with d-wave superconductivity. The data shows clear evidence for an αT^2 term at zero magnetic field in LTSH of $La_{1.78}Sr_{0.22}CuO_4$. Furthermore, our results are compared with the recently proposed scaling theory. [S0163-1829(98)50246-9]

After many efforts made to clarify the pairing symmetry in high-temperature superconductors (HTSC's), a growing consensus has emerged in recent years that the symmetry of hole-doped HTSC's is d wave.[1] One of the key experiments which helped to establish this consensus is the low-temperature specific heat (LTSH). In d-wave superconductors, at zero magnetic field $H=0$, the electronic specific heat C_e is proportional to T^2 rather than $\exp(-\Delta/T)$ as in conventional s-wave superconductors, where Δ is the superconducting gap. In magnetic fields, $C_e=\gamma(H)T$ at low temperatures with γ proportional to $H^{1/2}$, as first proposed by Volovik[2] and later by others.[3] Several papers[4–8] on LTSH $C(T,H)$ of $YBa_2Cu_3O_{7-\delta}$ (YBCO) have reported experimental results which in general agree with the d-wave predictions, however with controversies on the existence of the T^2 term at $H=0$. All LTSH experiments on YBCO investigated only the optimally doped and slightly overdoped samples. On the other hand, LTSH of $La_{2-x}Sr_xCuO_4$ (LSCO) at $H=0$ was measured and an obvious T^2 term has been identified.[9,10] Besides cuprates, studies on $C(T,H)$ of other possibly unconventional superconductors were reported.[11,12] In order to extend the test of the d-wave model to different doping regimes, we have measured LTSH of $La_{2-x}Sr_xCuO_4$ for $x=0.10$ to 0.22 both in zero and applied magnetic fields. In addition, it was pointed out that the $H^{1/2}$ dependence in $\gamma(H)$ might be a general phenomenon due to flux-line interactions near H_{c1}.[13] Since LSCO possesses smaller thermodynamics critical fields than YBCO, LTSH experiments on LSCO may be elucidating to this question. Our results show that the change of $\gamma(H)$ can be well described by Volovik's predictions in all doping regimes. Furthermore, the data reveal convincing evidence of both $C_e=\alpha T^2$ at $H=0$ and $C_e=AH^{0.5}T$ at $H\neq 0$ in the same sample, where α and A are constants. Comparison of our results with the recent scaling theory[14,15] is also presented.

Polycrystalline samples of $La_{2-x}Sr_xCuO_4$ with $x=0.10$ to 0.22 were carefully prepared from La_2O_3, $SrCO_3$, and CuO powder of 99.999% purity. Details of the preparation were described in Refs. 9 and 10 and references therein. The powder x-ray-diffraction patterns of all samples used in the experiments show a single T phase with impurity less than 3%. The transition width (90 to 10% by the resistivity drop) of T_c is 3 K or less, suggesting a decent homogeneity of the samples. $C(T)$ was measured from 0.6 to 10 K with a ^{3}He relaxation calorimeter using the heat-pulse technique in the magnetic field up to $H=8$ T. The precision of the measurement in the temperature range is about 1%. To test the accuracy of the field dependence of specific heat, $C(T,H)$ of a copper sample with mass 21.69 mg was measured at $H=0$, 1, and 8 T, respectively. The scatter of data in different magnetic fields is 3% or better. A fit of data below 7 K results in a linear term coefficient $\gamma=0.704\pm 0.003$ mJ/mol K^2 and the Debye temperature $\Theta_D=324$ K, both of which basically show no dependence on H. In this paper, we report $C(T,H)$ measurements for $H=0$ to 8 T on three LSCO samples of doping level $x=0.10$, 0.16, and 0.22 with $T_c=33$, 39, and 29 K, respectively.

The $C(T,H)$ data of samples with $x=0.16$, 0.10, and 0.22 are shown in Fig. 1. The zero-field data for all samples are fit to $C(T,0)=\gamma(0)T+\alpha T^2+C_{\text{lattice}}$, where $C_{\text{lattice}}=\beta T^3+\delta T^5$ represents the phonon contribution. Data in magnetic fields are fit to $C(T,H)=\gamma(H)T+C_{\text{lattice}}+nC_{\text{Schottky}}(g\mu H/k_BT)$, where the third term is a two-level Schottky anomaly $C_{\text{Schottky}}(x)=x^2e^x/(1+e^x)^2$. The anharmonic T^5 term for the phonon contribution is usually negligible at low temperatures, though inclusion of this term sometimes improves the quality of fit. Both the individual-field and global fit have been executed. It is found that both give similar results and do not change any conclusion of this paper. The fit of data below 7 or 8 K does not result in any significant change, either. In the following, the results of individual-field fit for $C(T,H)$ data from 0.6 to 7 K will be discussed.

The fitting results for all samples are described by the solid lines in Fig. 1, and the important resulting parameters are listed in Fig. 2 and Table I. For $x=0.16$ sample, $\gamma(0)=0.77$ mJ/mol K^2 and $\beta=0.230$ mJ/mol K^4. The corresponding Debye temperature is $\theta_D=389$ K, which agrees with the literature value determined from this temperature

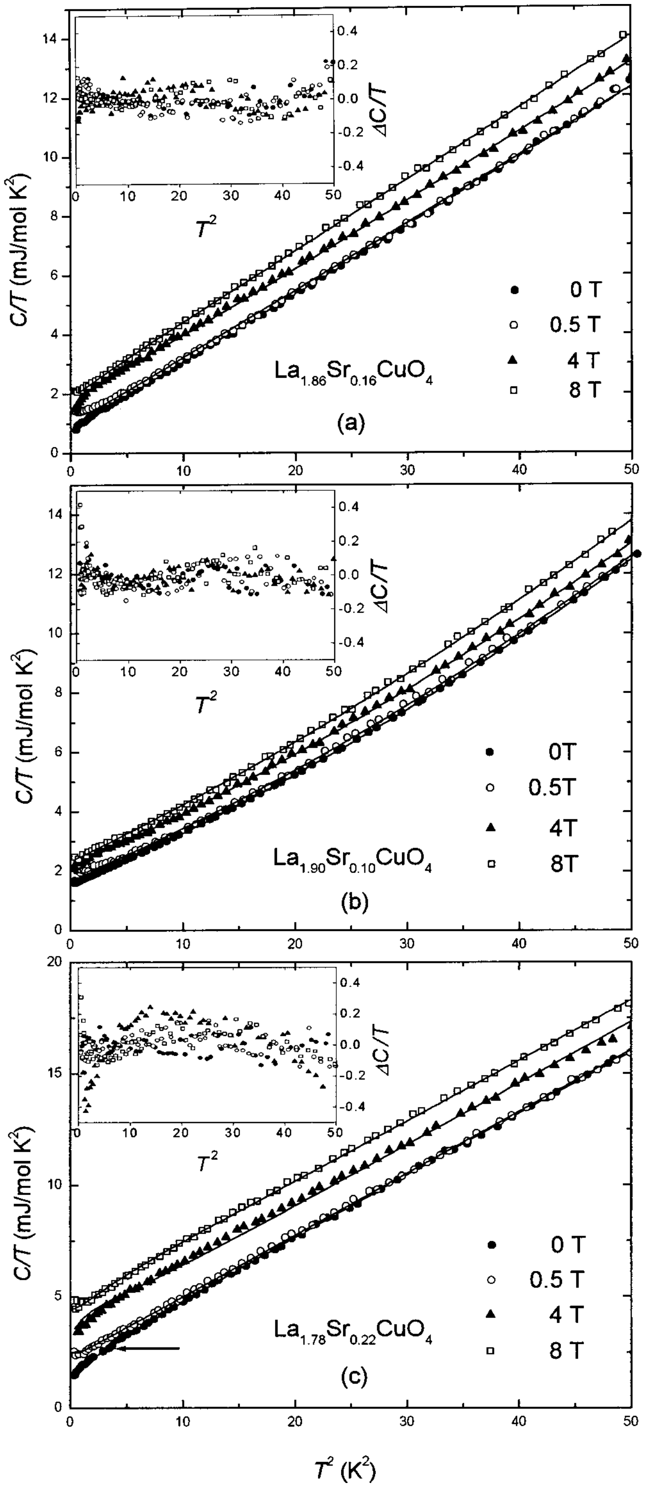

FIG. 1. C/T vs T^2 of (a) $x=0.16$, (b) $x=0.22$, and (c) $x=0.10$ LSCO. For clarity, only data at $H=0$, 0.5, 4, and 8 T are shown. The solid lines are from the fit described in text. The change from $C_e=\alpha T^2$ at $H=0$ to $C_e=\gamma T$ at $H\neq 0$ is emphasized in (c) by the arrow. Insets: the difference between data and the fit $\Delta C(=C_{\rm data}-C_{\rm fit})/T$ vs T^2.

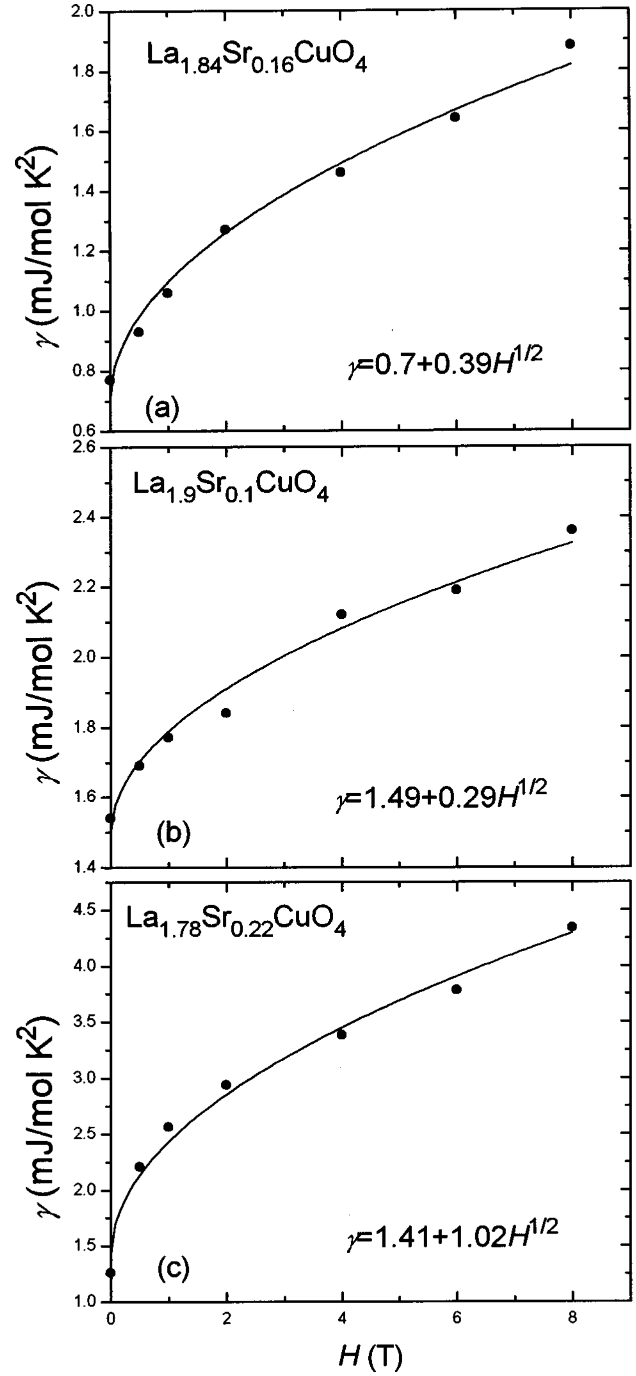

FIG. 2. Coefficient of the linear-T term γ for (a) $x=0.16$, (b) $x=0.10$, and (c) $x=0.22$ samples. The solid lines represent the fits of $\gamma=\gamma(0)+AH^{1/2}$.

range.[16] The small but nonzero $\gamma(0)$ was observed in all three samples, and remains unexplained within the clean d-wave model.[5] For both the $x=0.16$ and 0.10 samples, no apparent T^2 term at $H=0$ was observed as in Ref. 6 for YBCO. $\gamma(H)$ from the fit of data at $H\neq 0$ is shown in Fig. 2. The increase in $\gamma(H)$ can be well described by $AH^{1/2}$, which is a manifestation of the lines of nodes in the gap. It is interesting to note that the group at Berkeley obtained $A=0.47$ mJ/mol K^2 $T^{1/2}$ for $x=0.15$ LSCO,[7] which is comparable with our results. For $x=0.22$, at $H=0$ C/T vs T^2 shows an obvious downward curve at low temperatures rather than a straight line, as marked by the arrow in Fig. 1(c). The fit results in a significant αT^2 term. Since this αT^2 term is ~20% of total zero-field specific heat at 4 K and exceeds the γT term above this temperature, its identification is unambiguous (see Fig. 3). At $H=0.5$ T, this downward curve becomes a straight line except below 1 K where the

TABLE I. Comparison of LTSH experimental results with the estimates of *d*-wave model.

	$x=0.10$	$x=0.16$	$x=0.22$
T_c (K)	33	39	29
γ_n (mJ/mol K^2)	5	10	12
H_{c2} (T)		~50	
Estimated α (mJ/mol K^3)	0.17	0.26	0.41
Observed α (mJ/mol K^3)	<0.06	<0.01	0.31±0.02
Estimated A (mJ/mol K^{2}T$^{1/2}$)		0.96	
Observed A (mJ/mol K^{2}T$^{1/2}$)	0.29	0.39	1.02

contribution from the Schottky anomaly is important. The existence of the αT^2 term at $H=0$ and its disappearance in magnetic fields are both consistent with the predictions for the *d*-wave superconductivity. The increase in $\gamma(H)$ is again well described by $AH^{1/2}$. The obtained n from the fit is of the order 1 mJ/mol K for all samples, and the estimated concentration of spin-$\frac{1}{2}$ impurities per Cu atom is lower than 0.01%, one order of magnitude smaller than in the previous experiments.[4–6] Probably due to this reason, no upturn in C/T vs T^2 was observed down to 0.6 K for all samples. Through the paper, we fix $g=2.0$ to fit data.[4–6] However, it is found that the results are insensitive to the values of g from 1.5 to 2.5, partly because of the small magnetic contribution to C.

With C_e both at zero and nonzero magnetic fields qualitatively well described by *d*-wave superconductivity, it is interesting to compare values of α and A with the theoretical estimates, and the results are summarized in Table I. The coefficient α is estimated to be $\alpha \approx \gamma_n / T_c$, neglecting factors assumed to be of order 1. For the $x=0.22$ sample, taking $\gamma_n = 12$ mJ/mol K^2 for the corresponding T_c from Ref. 10, the estimate gives $\alpha = 0.41$ mJ/mol K^{-3}, comparable to the observed $\alpha = 0.31$ mJ/mol K^3. Similar estimates give much

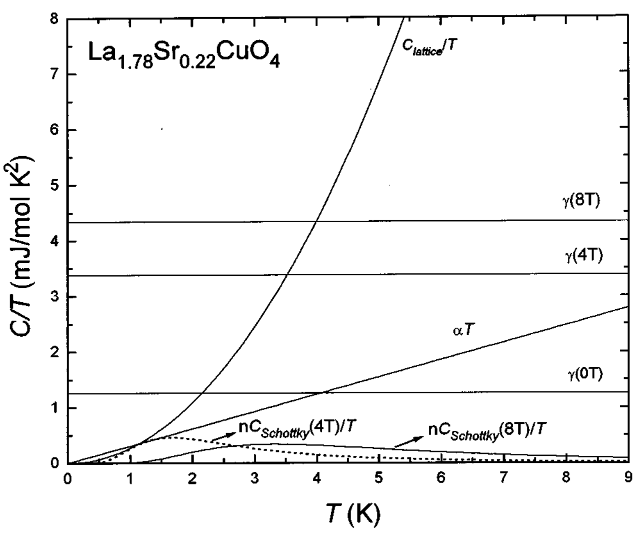

FIG. 3. The components of LTSH of the $x=0.22$ sample.

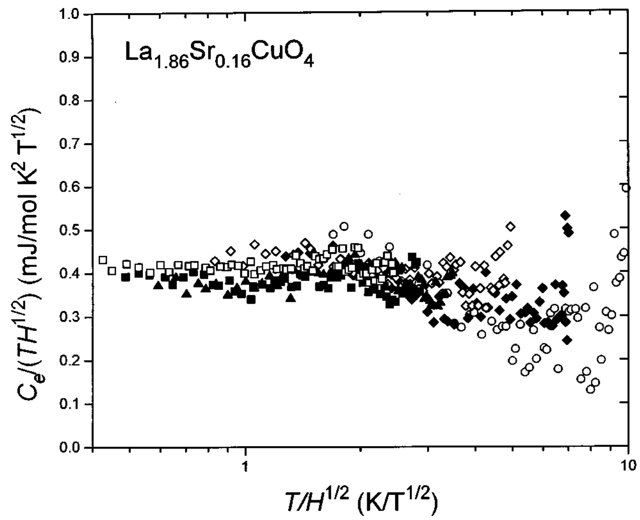

FIG. 4. Scaling plots of $C_e/(TH^{12})$ vs $T/H^{1/2}$ for $x=0.16$. Symbol: ○ = 0.5 T; ◆ = 1 T; ◇ = 2 T; ▲ = 4 T; ■ = 6 T; □ = 8 T.

smaller values of α for $x=0.10$ and 0.16 than the estimated α of $x=0.22$ (see Table I). Therefore, it is possible that the T^2 term cannot be confirmed by our measurements for $x=0.10$ and 0.16 may be either due to the intrinsic small values of α, or due to the effects of a small amount of defects or impurities which will be discussed later. In the same *d*-wave scenario, $A = k\gamma_n / H_{c2}^{1/2}$, where H_{c2} is the upper critical field and k is a factor of order 1.[2,4,5] H_{c2} of LSCO is much less known than that of YBCO. Taking $H_{c2} = 50$ T for $x=0.16$ with H parallel to the c axis,[17] $A \approx 0.96$ mJ/mol K^2 T$^{1/2}$ after averaging the anisotropy of H_{c2}. Given the uncertainties in such as k and H_{c2}, the observed $A=0.39$ agrees reasonably well with the theoretical estimate. It would be valuable to carry out the same comparison for $x=0.10$ and 0.22. However, H_{c2} of $x=0.10$ or 0.22 is not available to our best knowledge. According to $A = k\gamma_n / H_{c2}^{1/2}$ and assuming that H_{c2} is roughly proportional to T_c, one should expect that A of $x=0.10$ is close to that of $x=0.16$ and A of $x=0.22$ is much larger. Our results do show this trend.

According to the recent scaling theory,[14,15] $C_e \propto TH^{1/2}$ is exact only when $T/H^{1/2} \ll T_c / H_{c2}^{1/2}$. When H is around the c axis, roughly speaking, this criterion is satisfied in our experiments, especially at high fields (see Fig. 4). Since H_{c2} is anisotropic, $C_e \propto TH^{1/2}$ might not be valid when H is parallel to the planes. For polycrystalline samples, however, when the contribution to C_e from all directions is averaged, most of the contribution comes from around the c axis where H_{c2} is smaller. Therefore, a polycrystalline sample should still show the $TH^{1/2}$ term.[18] To compare with the scaling theory, $C_e/(TH^{1/2})$ vs $T/H^{1/2}$ for $x=0.16$ sample is shown in Fig. 4. Here C_e is obtained by $C_e = C - nC_{\mathrm{Schottky}} - C_{\mathrm{lattice}}$. Consistent with the scaling theory, all data at various T and H collapse into one scaling line for each sample, and the scaled data show a crossover to a stronger $T/H^{1/2}$ dependence at large $T/H^{1/2}$ as proposed by the scaling theory. For polycrystalline samples, one interesting question is whether the scaling relation holds for H parallel to the planes. The very recent theoretical work suggests that there exists a scaling function for H parallel to the planes, which form could depend on whether H is along the node.[18] Although it is still not clear theoretically, the experimental data seem to suggest that there is some approximate scaling relation for polycrystalline samples. The scaled data for $x=0.10$ and 0.22 are similar and will be discussed elsewhere.

In addition to the intrinsic small values of α for $x=0.10$ and 0.16, another alternative to explain the absence of αT^2 in these samples is the impurity effect. Should there be some impurities or defects in CuO_2 planes, the impurity scattering rate Γ could convert the T^2 term to the linear T term. Furthermore, $\gamma(H)=\gamma(0)[1+D(H/H_{c2})\ln(H_{c2}/H)]$, where $D \approx \Delta/32\Gamma$.[19,20] This expression can be tried to describe $\gamma(H)$ of $x=0.16$, and results in $\Gamma/\Delta=0.006$. In principle, the scaling in Fig. 4 should break down with the presence of impurity scattering. However, with such a small Γ/Δ, the breakdown might be too small to resolve by the measurements.[19] Trying the same expression on the $x=0.22$ sample fails to obtain a satisfactory fit, consistent with the presence of the αT^2 term. It is therefore compelling that the $x=0.22$ sample represents a clean d-wave superconductor. Whether the impurity scattering causes the absence of the αT^2 term in the other two samples and the validity of the expression of $\gamma(H)$ with impurities cannot be concluded by the present data. More studies on systematic impurity doping are underway to elucidate these questions.

In conclusion, by $C(T,H)$ measurements, we have shown that the superconducting state of LSCO is consistent with the d-wave scenario for the underdoped, optimum doped, and overdoped samples. No matter how different and curious their normal state might be,[21] LSCO samples of all doping levels seem to possess the same superconducting state.

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