# Electronic specific heat and low-energy quasiparticle excitations in the superconducting state of La<sub>2-r</sub>Sr<sub>r</sub>CuO<sub>4</sub> single crystals

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Low-temperature specific heat has been measured and extensively analyzed on a series of La2-xSrxCuO4 single crystals from underdoped to overdoped regime. From these data the quasiparticle density of state in the mixed state is derived and compared to the predicted scaling law  $C_{vol}/T\sqrt{H}=f(T/\sqrt{H})$  of d-wave superconductivity. It is found that the scaling law can be nicely followed by the optimally doped sample (x=0.15) in quite a wide region of  $(T/\sqrt{H} \leq 8K/\sqrt{T})$ . However, the region for this scaling becomes smaller and smaller toward more underdoped region: a clear trend can be seen for samples from x=0.15 to 0.069. Therefore, generally speaking, the scaling quality becomes worse on the underdoped samples in terms of scalable region of  $T/\sqrt{H}$ . This feature in the underdoped region is explained as due to the low-energy excitations from a second order (for example, antiferromagnetic correlation, d-density wave, spin-density wave, or charge-density wave order) that may coexist or compete with superconductivity. Surprisingly, deviations from the d-wave scaling law have also been found for the overdoped sample (x=0.22), while the scaling law is reconciled for the overdoped sample, when the core size effect is taken into account. An important discovery of present work is that the zero-temperature data follow the Volovik's relation  $\Delta \gamma(T=0) = A \sqrt{H}$  quite well for all samples investigated here; although the applicability of the *d*-wave scaling law to the data at finite temperatures varies with doped-hole concentration. We also present the doping dependence of some parameters, such as the residual linear term  $\gamma_0$ , the  $\alpha$  value, etc. It is suggested that the residual linear term ( $\gamma_0 T$ ) of the electronic specific heat observed in all cuprate superconductors is probably due to the inhomogeneity, either chemical or electronic in origin. The field-induced reduction of the specific heat in the mixed state is also reported. Finally, implications on the electronic phase diagram are suggested.

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# I. INTRODUCTION

One of few points with consensus in the cuprate superconductors is the  $d_{x^2-y^2}$  pairing symmetry in the hole-doped region. This has been supported by tremendous experiments<sup>1</sup> both from surface detection<sup>2-6</sup> and bulk measurements.<sup>7-14</sup> In a *d*-wave superconductor with line nodes in the gap function, the quasiparticle density of state (DOS) N(E) rises linearly with energy at the Fermi level in zero field, i.e., N(E) $\propto |E - E_F|$ , resulting in<sup>15</sup> an electronic specific heat  $C_e = \alpha T^2$ , where  $\alpha \propto \gamma_n / T_c$  and  $\gamma_n$  is the specific heat coefficient, which is proportional to the DOS at the Fermi level of the normal state. In the mixed state with the field higher than a certain value, the DOS near the Fermi surface becomes finite, therefore, the quadratic term  $C_e = \alpha T^2$  will be surpassed and substituted by both the localized excitations inside the vortex core and the delocalized excitations outside the core. Volovik<sup>16</sup> pointed out that for *d*-wave superconductors in the mixed state, supercurrents around a vortex core lead to a Doppler shift to the quasiparticle excitation spectrum, which strongly affects the low-energy excitation around the nodes. It was shown that the contribution from the delocalized part (outside the core) will prevail over the core part and the specific heat is predicted to behave  $as^{15,16} C_{vol} = k\gamma_n T \sqrt{H/H_{c2}}$  with k in the order of unity. This prediction has been verified by many measurements, which were taken as the evidence for d-wave symmetry.<sup>8–14,17</sup> In the finite temperature and field region a scaling law is proposed<sup>18</sup> as

$$C_{vol}/T\sqrt{H} = f(T/\sqrt{H}) \tag{1}$$

with  $T/\sqrt{H} \leq T_c/\sqrt{H_{c2}(0)}$ . This scaling law can be further converted into the form of  $C_{vol}/H=g(T/\sqrt{H})$  or  $C_{vol}/T^2$  $=y(T/\sqrt{H})$ , here f(x) or g(x) or y(x) are unknown scaling functions. This scaling law has been proved in YBCO<sup>8-10</sup> and in LSCO.<sup>11-14</sup> It remains, however, unclear whether this scaling law is still valid in the very overdoped region because the vortex core size  $\xi$  grows up. In the underdoped region, inelastic neutron scattering reveals that an antiferromagnetic order emerges when the superconductivity is suppressed.<sup>19,20</sup> It is thus also interesting to check whether the *d*-wave scaling law proposed by Simon and Lee is applicable in underdoped regime. In addition, the Simon-Lee scaling law is in agreement with the calculations as proposed by Kopnin and Volovik<sup>15</sup> and Volovik<sup>16</sup> in two extreme conditions of temperature. In the low-temperature limit, the scaling law  $C_{vol}/T^2 = y(T/\sqrt{H})$  becomes the Volovik's relation  $C_{vol} = AT\sqrt{H}$ . When the temperature is increased, another relation  $C \propto aT^2 + bH$  is reached. The boundary between these two regions is  $T/\sqrt{H} = T_c/\sqrt{H_{c2}}$  according to Volovik and Kopnin.<sup>21</sup> These theoretical models can be quantitatively tested by experiments on samples with different doping concentrations.

Another important but controversial issue is the vortex core state in the cuprate superconductors. By solving the mean-field Bogoliubov-de Gennes (BdG) equation theoretically, it is suggested that a zero-bias conductance peak (ZBCP) exists in the vortex core.<sup>22,23</sup> However, this is in sharp contrast with the experimental observations,<sup>24-29</sup> mainly on optimally doped samples. The absence of a ZBCP within the vortex cores was attributed to the presence of  $id_{rv}$ or is components<sup>28</sup> or the competing orders (see Sec. III D). In this paper we show that the DOS, because of vortex quasiparticle excitations, deviates from Simon-Lee scaling law for the overdoped sample, but follows rather well with the optimally doped sample. The deviations for the overdoped sample are found to be induced by the vortex core size effect. In the extremely underdoped region, it is found that the Simon-Lee scaling law fails, except for in very lowtemperature regions. This can be understood as being due to the competing order emerging within or nearby the vortex cores.

### **II. EXPERIMENT**

The single crystals measured in this work were prepared by the traveling-solvent floating-zone technique. Samples seven different doping with concentrations (p=0.063,0.069,0.075,0.09,0.11,0.15,0.22) have been investigated. The sample with p=0.15 and 0.22 are from CRIEP, and others are from NLSC(IOP). Part of the data for all samples, will be presented, for example, the field-induced change of  $\gamma$  at zero K, the residual linear term  $\gamma_0$ , and the  $\alpha$ value in the pure d-wave expression  $C_{DOS} = \alpha T^2$  when H = 0(see Sec. III A). However, for clarity we mainly show the data and analysis on three typical samples with x=0.22 ( $T_c$ =27.4 K, overdoped), x=0.15 ( $T_c=36.1$  K, close to optimal doping point), and p=0.069 ( $T_c \approx 12$  K, underdoped, x =0.063 originally) as characterized by AC susceptibility and DC magnetization (shown by the insets in Figs. 3 and 8 and in the main panel of Fig. 12). The quality of our samples has also been characterized by x-ray diffraction patterns, and R(T) data showing a narrow transition  $\Delta T_c \leq 2$  K. For some samples, the full width at the half maximum (FWHM) of the rocking curve of the (008) peak is only 0.10°. The overdoped sample has a mass of about 28.55 mg and  $3.66 \times 2.3$  $\times 0.5 \text{ mm}^3$  in dimension. The optimally doped sample weighs about 23.6 mg with dimensions of  $3.1 \times 3$  $\times 0.5$  mm<sup>3</sup>. For the underdoped sample with nominal concentration x=0.063 before annealing, it has a superconducting transition temperature of about 12 K and a mass of about 32.89 mg and  $3.75 \times 2.75 \times 0.5$  mm<sup>3</sup> in dimensions. By fitting to the empirical relation  $T_c/T_c^{max} = 1 - 82.6 \ (p - 0.16)^2$ with  $T_c^{max} = 38$  K the maximum  $T_c$  at the optimal doping point p=0.16, we estimate that the hole concentration of this



FIG. 1. Temperature dependence of the heat capacity from the addenda with 110  $\mu$ g Wakefield grease (bottom solid line for H = 0 and dashed line for H=12 T), and three typical samples (filled symbols for H=0 and open symbols for H=12 T, lines are guides to the eye).

sample is around p=0.069. After annealing in flowing Ar gas for 48 h, the  $T_c$  drops down from about 12 K to 9 K, indicating that the sample becomes more underdoped. Note that  $T_c=9$  K is expected exactly by the empirical relation at x = p=0.063.

The heat capacity presented here was taken with the relaxation method based on an Oxford cryogenic system Maglab. The sample is put onto a microchip on which there is a tiny Cernox temperature sensor and a film heater. The microchip, together with the sample, are hung up by golden wires in vacuum. These golden wires are the only thermal links between the microchip and the thermal sink, whose temperature is well controlled. The temperature of the microchip is controlled by the onboard small film heater and measured by the onboard thermometer. When the temperature of the microchip is stable, a heating power with fixed current is sent to the film heater on the chip and the time dependence of the chip temperature is measured simultaneously. The change in temperature is fitted to an exponential relation  $\Delta T = \Delta T_0$  $\times [1 - \exp(-t/\tau)]$ , and heat capacity is determined by  $\tau = (C + \tau)$  $+C_{add})/\kappa_w$ ; here, C and  $C_{add}$  are the heat capacity of the sample and addenda [including a small sapphire substrate, small printed film heater, tiny Cernox temperature sensor,  $\phi$ 25  $\mu$ m gold wire leads, and Wakefield thermal conducting grease (about 100  $\mu$ g)], respectively, where  $\kappa_w$  is the thermal conductance between the chip and the thermal link. The value  $C_{add}$  has been measured and subtracted from the total heat capacity, thus the C value reported here is only that from the sample. In Fig. 1 we present the temperature and field dependence of the heat capacity from the addenda and three typical samples. It is clear that the heat capacity of the addenda is much smaller than the value of the samples. In addition, the data of C/T for the addenda extrapolates to zero at T=0 K showing the only existence of a phonon part. One can also see that almost no field dependence can be observed for the addenda. However, for all samples, there is a clear finite intercept at T=0 K, which gives rise to a residual linear term  $\gamma_0$ . Meanwhile the field-induced change can be easily observed for all samples, even for the very underdoped sample. The intercrossing of the data at H=0 and H=12 T at



FIG. 2. Temperature dependence of C/T for samples (p=0.069, 0.075, 0.09, 0.11, 0.15, and 0.22) at zero field. The solid lines are fits to Eq. (2), and the parameters derived here are listed in Table I.

about 6 K for the undedoped sample is understandable and will be discussed later.

In all measurements taken for the present work, the magnetic field H is always parallel to the c-axis of single crystals, and the data are collected in the warm-up process after it is cooled under a field [field-cooling process (FC)]. In the data treatment, we use  $\Delta \gamma = (C_{H\parallel c} - C_{H=0})/T$  instead of using  $\Delta \gamma$  $=(C_{H\parallel c}-C_{H\perp C})/T$ . The latter may inevitably involve the unknown DOS contributions from another kind of vortice (for example, Josephson vortices) when  $H \perp C$ . The field dependence of the Cernox thermometer has been calibrated well by Oxford before the shipment. The true temperature has been derived automatically by the software with a calibration table with magnetic fields at 0 T, 1 T, 2 T, 4 T, 8 T, and 12 T. The values at other fields are also obtained automatically by software by doing linear interpolations between two nearby fields. Therefore, the readout from the machine directly gives the true temperature value with the field effect corrected.

### **III. RESULTS AND DATA ANALYSIS**

#### A. Fitting to the zero field data

Before showing the field-induced change of the heat capacity, we present (in Fig. 2) the temperature dependence of C/T for some samples at zero field. As mentioned previously, for a d-wave superconductor in the superconducting state, it is known that  $C_{DOS} = \alpha T^2$  when H = 0. In addition, as observed in other cuprate superconductors, the curve at zero field extrapolates to a finite value ( $\gamma_0$ ) at 0 K instead of zero. This was interpreted as a potential scattering effect due to a small amount impurities or disorders.<sup>8,30</sup> We will argue that this residual linear term may also reflect physics beyond the simple argument of impurity scattering (see Sec. III E). As also observed by other groups for the La-214 system, the anomalous upturn of C/T due to the Schottky anomaly of free spins is very weak.<sup>11–14</sup> This avoids the complexity in the data analysis. Together with the phonon contribution  $\beta T^3 + \delta T^5$ , we have

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TABLE I. Values of  $\gamma_0$ ,  $\alpha$ ,  $\beta$ , and  $\delta$  determined by fitting the data at H=0 to Eq. (2).

р	$T_c$	$\gamma_0$	α	β	δ
0.22	27.4	2.19	0.463	0.186	0.00054
0.15	36.1	1.90	0.177	0.120	0.00093
0.11	29.3	1.70	0.065	0.137	0.00096
0.09	24.4	2.64	0.158	0.145	0.00110
0.075	15.6	3.72	0.131	0.177	0.00110
0.069	12.0	4.06	-0.077(?)	0.157	0.00117

The above equation is used to fit the data at H=0 for some samples. The fitting results are shown in Fig. 2 and listed in Table I, where the units for  $\gamma_0$ ,  $\alpha$ ,  $\beta$ , and  $\delta$  are mJ mol<sup>-1</sup> K<sup>-2</sup>, mJ mol<sup>-1</sup> K<sup>-3</sup>, mJ mol<sup>-1</sup> K<sup>-4</sup>, and mJ mol<sup>-1</sup> K<sup>-6</sup>, respectively. One can see that  $\alpha$  decreases quickly toward underdoping and  $\beta$  (and thus the Debye temperature  $\Theta_D$ ) does not change too much with doping. The sudden drop of  $\alpha$  at p=0.11 may be induced by the well-known  $\frac{1}{8}$  problem. The residual linear term  $\gamma_0$  increases rapidly toward underdoping, which will be discussed later. The  $\alpha$  values are also comparable to those found by other groups.<sup>13,14</sup>

#### **B.** Overdoped sample with x = 0.22

Figure 3 shows C/T as a function of  $T^2$  at magnetic fields ranging from 0 to 12 T for the overdoped sample. The separation between each field can be well determined. In a low-temperature region the curves are rather linear, showing that the major part is due to phonon contribution  $C_{ph} = \beta T^3$ +  $\delta T^5$ . It is known that the phonon part is independent on the magnetic field, this allows one to remove the phonon contribution by subtracting the C/T at a certain field with that at zero field. The results after the substraction are shown in Fig. 4. The subtracted values  $\Delta \gamma = \gamma(H) - \gamma(0) = [C(T,H) - C(T,H=0)]/T$  exhibit a rather linear T dependence in the



FIG. 3. Specific heat C/T vs  $T^2$  of the overdoped sample (x = 0.22) at magnetic fields ranging from 0 to 12 *T*. The inset shows the diamagnetic transition at around 27.4 K determined by the crossing point of the extrapolating line of the most steep part with the normal state background M=0.



FIG. 4. Temperature dependence of  $\Delta \gamma = \gamma(H) - \gamma(0) = [C(H,T) - C(0,T)]/T$  of the overdoped sample (*x*=0.22). A linear behavior is clearly seen in the low-temperature region with a field-dependent slope, which is not in accord with the proposed scaling law by Simon and Lee (see text). The straight lines in the low-temperature region are guides to the eyes. From these lines one can determine the zero-temperature intercept  $\Delta \gamma$  and the slope  $d\Delta \gamma/dT$  shown in Fig. 5.

low-temperature region. One can also see that the negative slope is actually field dependent. In the following we will show that the field-dependent slope of the linear part in the low-temperature region shown in Fig. 4 directly deviates from the Simon-Lee<sup>18</sup> scaling law.

According to the Simon-Lee scaling law  $C_{vol}/T\sqrt{H} = f(T/\sqrt{H})$ , in the low-temperature region, the Volovik's relation is restored; thus, one has  $C_{vol}/T = A\sqrt{H}$  which further leads to

$$\Delta \gamma = [C(H) - C(0)]/T = A\sqrt{H - \alpha T}.$$
(3)

This clearly shows that there is a negative slope for  $\Delta \gamma$  versus *T*, but the slope  $\alpha$  is a constant. However, from Fig. 4 one can see that the slope changes slightly with the magnetic field *H*. This indicates that the Volovik's relation alone is not enough to interpret the data. In the following, we will take both the core-size and finite-temperature effects into account. The former has not been considered in the original Simon-Lee scaling law because the size of the vortex core was thought to be small, and the contribution from that small region is negligible. If the vortex core size becomes bigger, this should be reconsidered when counting the DOS due to the Doppler-shift effect.

Let us first consider only the finite-temperature effect. Suppose that we are in the crossover region between the lowand high-temperature limits suggested by Volovik and Kopnin,<sup>21</sup> making Taylor's expansion to the right-hand side of the Simon-Lee scaling law lead to

$$C_{vol} = b_0 H + b_1 T \sqrt{H} + b_2 T^2 + o(T^3), \qquad (4)$$

where  $b_0=0$  because  $C_{vol}/T$  should not diverge when T=0and  $H \neq 0$ , b1=A. Since  $o(T^3)$  is very small in the lowtemperature region, one therefore has  $C_{vol}/T=b_1\sqrt{H}+b_2T$ . Interestingly, one can see that this simple formula contains the results both in the low-temperature limit

 $C_{nol} = b_1 T \sqrt{H^{15,16,31}}$  and in the high-temperature limit,<sup>21,31</sup>  $C_{vol} = b_2 T^2$ . This is not surprising because a scaling function should be more general and cover most possible cases. When  $H_{c1} \ll H \ll H_{c2}$ , the total specific heat contains four parts: Doppler shift term from the region outside the core  $C_{vol}$ , the inner vortex core term  $C_{core} \propto HT$ , the residual linear term  $\gamma_0 T$ , and the phonon term  $C_{ph}$ . Here it is assumed that the heat capacity contributed by the core region is equal for each vortex and independent on the external magnetic field. Thus,  $C_{core}$  depends only on the vortex density, which is proportional to H. The local DOS measured by STM<sup>26</sup> revealed that the low-energy DOS within the vortex core differs only slightly from the case for a *d*-wave superconductivity (outside and far away from the vortex core). When changing the external magnetic field, the low-energy DOS within the vortex core is not expected to vary too much. At zero field, the total specific heat contains three parts:  $\gamma_0 T$  and  $C_{ph}$  and a quadratic term  $\alpha T^2$  due to the thermal excitation near the nodal region. Thus,  $\Delta \gamma$  can be written as

$$\Delta \gamma = \gamma(H) - \gamma(0) = b_1 \sqrt{H} + (b_2 - \alpha)T + b_{core}H.$$
 (5)

From Eq. (5) one can see that  $\Delta \gamma$  depends on *T* through the second term, however, the slope  $b_2 - \alpha$  is still field independent by definition. This clearly indicates that the Simon-Lee scaling law is still not enough to interpret the field dependent slope of  $\Delta \gamma$  versus *T* as shown in Fig. 4.

Let us keep going, still based on Eq. (5), we propose that the core-size effect may have a sizable influence on the total vortex quasiparticle excitations. This is actually reasonable because the vortex core with size  $\xi \propto \hbar v_F / \Delta_s$  grows up in the overdoped side due to a smaller superconducting gap value,<sup>32</sup> where  $v_F$  is the Fermi velocity and  $\Delta_s$  is the superconducting gap. By taking the vortex core size  $(2\xi)$  into account, i.e., deducting the normal core area away from the Volovik term, one can rewrite  $\Delta \gamma$  as

$$\Delta \gamma = (b_1 \sqrt{H} + b_2 T) \times (1 - \xi^2 / R_a^2) - \alpha T + b_{core} H, \qquad (6)$$

where  $\xi$  is the radius of the normal core and  $R_a$  is the outer radius of a single vortex where the supercurrent is flowing, thus  $R_a^2 = \phi_0 / \pi H$ . Reorganizing all terms in Eq. (6) leads to

$$\Delta \gamma = b_1 \sqrt{H} \times \left( 1 - \frac{\pi \xi^2}{\phi_0} H \right) + (b_2 - \alpha)T - b_2 \frac{\pi \xi^2}{\phi_0} HT + b_{core} H.$$
(7)

One can see that the third term in Eq. (7) is just what we need for interpreting the difficulty as mentioned above. It is necessary to recall that the core-size correction is proportional to  $\xi^2$ , for example, it will be four times when  $\xi$ doubles. Thus increase of  $\xi$  in the overdoped side will have a sizable effect on the total DOS, and core size effect should be considered. Next let us make a closer inspection of the data and derive some parameters. At zero temperature, only the first and last terms are left in Eq. (7). The values of  $\Delta \gamma(T=0)$  are determined from the extrapolation of the linear lines in Fig. 4 to 0 K and presented in Fig. 5. The data  $\Delta \gamma(H, T=0)$  are also determined by doing a linear fit to the raw data C/T versus  $T^2$  between 2 K and 4 K, and then subtracting the zero-temperature value  $\gamma_0$ . The results are



FIG. 5. Field-induced DOS at 0 K of the overdoped sample (x =0.22). The solid line is a theoretical curve  $\Delta \gamma = 1.995 \sqrt{H}(1 - 0.0121H)$ . The dotted line represents the best fit to the case at the unitary limit (Ref. 30). The inset shows the slope  $d\Delta \gamma/dT$  of the straight lines shown in Fig. 4 in the low-temperature region. The dashed line is a linear fit to the data at fields above 1 tesla. The intercept and the slope of the dashed line give rise to the prefactors of the second and third terms in Eq. (7).

quite close to each other using these two different methods. The solid line in Fig. 5 is a fit to the data using the first term in Eq. (7), yielding  $b1 = 1.995 \pm 0.046$  mJ K<sup>-2</sup>T<sup>-1/2</sup> and  $\pi \xi^2 / \hat{\phi}_0 = 0.012 \pm 0.003$  and, thus,  $\xi = 28.2$  Å. The value  $\xi$ =28 Å derived here is quite close to that found in Nernst<sup> $3\overline{3}$ </sup> and STM measurements<sup>26,34</sup> (20 Å for the optimally doped Bi-2212 sample). We also tried to use the first term together with the last term to fit the data, but found that the contribution from the last term is extremely small. The first term here describes the zero temperature data very well, indicating the absence of a second component of the order parameter, such as  $id_{xy}$ , or is since otherwise the Fermi surface would be fully gapped and the Doppler shift had very weak effect on the quasiparticle excitations. The inset of Fig. 5 shows the field dependence of the slope of the linear part in Fig. 4. It is clear that the slope increases roughly linearly with Habove 1 T. This can be exactly anticipated by the second and third terms in Eq. (7). From the inset of Fig. 5 one  $\alpha - b_2 = 0.305 \text{ mJ mol}^{-1} \text{ K}^{-3}$ and obtains  $b_2\pi\xi^2/\phi_0$ =0.00238 mJ mol<sup>-1</sup> K<sup>-3</sup>T<sup>-1</sup>. By taking  $\xi$ =28.2 Å, we obtain the following values:  $\alpha$ =0.501 mJ mol<sup>-1</sup>K<sup>-3</sup> and  $b_2$ =0.196 mJ mol<sup>-1</sup> K<sup>-3</sup>. The value of  $\alpha$ =0.501 mJ mol<sup>-1</sup> K<sup>-3</sup> found here is quite close to the value obtained by fitting the zero field data to Eq. (2) (0.465 mJ/mol  $K^2$ , see Table I). Since the contribution from the core region [last term in Eq. (7)] is negligible compared to the Volovik term, from Eq. (7) one understands that the failure of using the Simon-Lee scaling law in a very overdoped sample is because of the coresize effect. This is actually quite reasonable because the core size in the overdoped region grows up. We do not yet know whether the negligible contribution from the core region is because of the gapped feature within the core region, as found in optimally and underdoped samples,<sup>24-29</sup> or if it is naturally small compared to the contributions from the Doppler-shift effect of the surrounding superfluid. This casts an interesting issue for future STM measurement on the tun-



FIG. 6. Scaling of the raw data  $[C(H) - C(0)]/T\sqrt{H}$  vs  $T/\sqrt{H}$  for the overdoped sample based on the Simon-Lee scaling law. Clearly, no good scaling can be obtained.

neling spectrum within the vortex cores in very overdoped regions.

It is necessary to estimate how much of the field-induced delocalized DOS is contributed by the impurity scattering in our present sample. At the unitary limit at zero energy, i.e., when T=0, Kübert and Hirschfeld<sup>30</sup> predict that the fieldinduced relative DOS is  $\delta \gamma / \gamma_0 = P_1(H/P_2)\log(P_2/H)$ , where  $P_1 = 0.322 (\Delta_0/\Gamma)^{1/2}$ ,  $\Delta_0$  the gap maximum,  $\Gamma$  the impurity scattering rate, and  $P_2 = \pi H_{c2}/2a^2$ ,  $a \approx 1$ . The dotted line in Fig. 5 represents the best fit of this relation to our data yielding  $\Gamma/\Delta_0 \approx 0.00039$  (close to the clean limit). In addition, the value of  $H_{c2}$  derived here is about 21.53 T, which is too small for the present sample. It is clear that the fit has poor quality compared to the better fit in the clean limit (solid line). Furthermore, the formula considering the impurity effect does not predict a field-dependent slope for the linear relation  $\Delta \gamma$  versus T in the low-temperature region. Therefore, together with the extremely small  $\Gamma/\Delta_0$  found in the present case, we believe that the field-induced DOS in our sample comes mainly from the Doppler-shift effect on the supercurrent outside the cores. The residual linear term  $(\gamma_0 T)$ of electronic specific heat will be discussed separately in the Sec. III C.

In order to show the inapplicability of the Simon-Lee scaling law for the overdoped sample, we present the raw data  $[C(H) - C(0)]/T\sqrt{H}$  versus  $T/\sqrt{H}$  in Fig. 6. Clearly, the scaling looks very poor. From the above discussion, we conclude that the failure of the Simon-Lee scaling law in the very overdoped region is due to the quite large vortex core size, which needs to be corrected.

The nice fit in Fig. 5 with only the first term of Eq. (7) suggests that the core region makes a very small contribution to the DOS, otherwise the last term  $b_{core}H$  should be sizeable. This implies that the low-energy DOS inside the vortex core is very small. Based on this idea, we write a different scaling law as follows:

$$\frac{\Delta C/T^2 + \alpha}{1 - \pi\xi^2 H/\phi_0} = \frac{\sqrt{H}}{T} f\left(\frac{T}{\sqrt{H}}\right)$$
(8)

One can use this equation to test the idea about the vortex core-size correction. We present the data of  $(\Delta C/T^2 + \alpha)/$ 



FIG. 7. Plot of the data  $C_{scal} = (\Delta C/T^2 + \alpha)/(1 - \pi \xi^2 H/\phi_0)$  vs  $T/\sqrt{H}$ . It is clear that the data collapse onto one main branch, which is expected by the theoretical expression with core-size correction [Eq. (8)].

 $(1 - \pi \xi^2 H / \Phi_0)$  versus  $T / \sqrt{H}$  in Fig. 7 with  $\alpha$ =0.501 mJ/mol  $K^3$  and  $\pi\xi^2/\phi_0=0.0121$  derived above. The data collapse on one branch and show good consistency with the expected theoretical curve. The slight scattering or deviation from the main scaling branch is due to the simple assumption made for the core-size correction  $(1-\xi^2/R_a^2)$  and the rough estimation for  $\alpha$  value. It is worth noting that to have this nice data collapse and be consistent with the theoretical curve, we need to take  $b_{core} \approx 0$ , again showing a small contribution from the inner vortex core. The nice data collapse using Eq. (8) suggests that the Simon-Lee scaling law can be reconciled by considering the vortex core-size effect. It is interesting to note that the electronic thermal conductivity derived by Sun et al.35 is not consistent with the Volovik's expression in the low-temperature region for the overdoped sample, rather it shows a plateau when the field is high. However, the  $H^{1/2}$  law is followed very well in the low-temperature region for the optimally doped sample. Our core-size correction picture may provide an alternative interpretation to this discrepancy.

#### C. Optimally doped sample (x=0.15)

In order to have a comparison with the overdoped sample, in this section we present the data from an optimally doped



FIG. 8. Raw data of C(H)/T vs  $T^2$  for the optimally doped sample. The inset shows the diamagnetic transition measured in the ZFC mode at 20 Oe.



FIG. 9. Scaling of the raw data  $[C(H) - C(0)]/T\sqrt{H}$  vs  $T/\sqrt{H}$  for the optimally doped sample (x=0.15). The scaling looks rather good.

sample with x=0.15. The raw data of specific heat for the optimally doped sample is shown in Fig. 8. The separation between each field can also be easily distinguished in the low-temperature region. Again here the curve at zero field extrapolates to a finite value ( $\gamma_0$ ) at 0 K instead of zero. This will be discussed in the forthcoming Sec. III D. It is found that the linear behavior of  $\Delta \gamma$  versus T for the overdoped sample (shown in Fig. 4) is absent here. This may be due to the much smaller  $\alpha$  value (see Table I). We then check whether the *d*-wave scaling law is applicable here. If the Volovik (Doppler shift) effect really dominates here, one can expect that  $C(H) - C(0) = T^2 y(T/\sqrt{H}) - \alpha T^2$ , thus, [C(H)] $-C(0)]/T^2$ , or  $[C(H)-C(0)]/T\sqrt{H}$  should scale with  $T/\sqrt{H}$ . In Fig. 9 we present the result of  $[C(H) - C(0)]/T\sqrt{H}$  versus  $T/\sqrt{H}$ . It is clear that the scaling is rather good compared to that of the overdoped sample [Fig. 6]. Here the value of  $\Delta \gamma / \sqrt{H}$  in the zero-temperature limit gives the prefactor A in the Volovik's relation  $C_{vol} = AT\sqrt{H}$ , which is about 0.55 to 0.6 mJ mol<sup>-1</sup> K<sup>-2</sup>T<sup>-1/2</sup>. In Fig. 10 we present the Simon-Lee scaling law as  $[C(H) - C(0)]/T^2$  versus  $T/\sqrt{H}$ . One can see that the scaling is reasonably good. All data below about 10 K collapse onto one branch. We have been aware that Nohara et al.13 successfully used the Simon-Lee



FIG. 10. Scaling of the raw data  $[C(H) - C(0)]/T^2$  vs  $T/\sqrt{H}$  for the optimally doped sample based on the Simon-Lee scaling law. Clearly, the scaling quality is quite good.



FIG. 11. Zero-temperature specific heat  $\Delta \gamma(H)$  of the optimally doped sample. The solid line is a theoretical expression  $\Delta \gamma(H) = 0.577 \sqrt{H} \text{ (mJ/mol } K^2)$ , which fits the data very well. The dashed line is a fit to the first term of Eq. (7), yielding a small and unreasonable negative value for  $\pi \xi^2 / \phi_0$ .

scaling law for the overdoped sample x=0.19, but failed for the optimally doped sample. The failure of using the Simon-Lee scaling law in Nohara's experiment for the optimally doped sample is in contradiction to the reports from many other groups.<sup>11,12,14</sup> This may be caused by the way in which they derived  $\Delta \gamma$ . As stressed in Sec. III B, we use  $\Delta \gamma$ = $[C_{H\parallel c} - C_{H=0}]/T$  instead of using  $\Delta \gamma = [C_{H\parallel c} - C_{H\perp C}]/T$  to derive the field-induced change of  $\gamma$ . The latter (as used by Nohara et al.) may inevitably involve the unknown DOS contributions from another kind of vortice (for example, Josephson vortices), when  $H \perp C$ . For La-214 system, since the Schottky anomaly is very weak, it is not necessary to derive  $\Delta \gamma$  in the second way. While Nohara *et al.*<sup>13</sup> obtained a relatively good scaling for the overdoped sample we would not comment on the validity of this successful scaling at x=0.19. One reason for the discrepancy between their results and ours may be because of the different doping levels; our sample (x=0.22) is more overdoped, and the vortex core size is certainly larger and needs to be corrected.

The data  $\Delta \gamma(H,T=0)$  is determined by doing a linear fit to the raw data C/T versus  $T^2$  between 2 K and 4 K, and then subtracting the zero-temperature value  $\gamma_0$ . The results are shown in Fig. 11. We tried to fit the zero-temperature data in Fig. 11 to the first term in Eq. (7) in terms of coresize correction (shown by the solid line). It turns out that the correction term  $\pi \xi^2 H/\phi_0$  is very small and negative, which is certainly unreasonable. This actually indicates that  $\Delta \gamma(T)$ =0) can be nicely fitted to the theoretical expression  $\Delta \gamma(H) = 0.577 \sqrt{H} (\text{mJ/mol K}^2)$ . Using  $C_{vol}/T = k \gamma_n \sqrt{H/H_{c2}}$ , we have  $k\gamma_n = 0.577 \sqrt{H_{c2}}$ . A similar value (0.49) was derived by Fisher *et al.*<sup>11</sup> for La-214 sample with x=0.15. Taking  $\dot{H}_{c2} \approx 100 \ T^{32}$  and  $k \approx 0.74^{17}$ , we have  $\gamma_n \approx 7.8 \text{mJ/mol K}^2$ , which is very close to the reported values for optimally doped La-214 sample.<sup>13,14</sup> It is worth of noting here that the field-induced extra DOS at 0 K can be nicely fitted with the Volovik's relation  $C_{vol} \propto T \sqrt{H}$ , albeit the residual linear term  $\gamma_0$  is quite large. This suggests that the residual linear term  $\gamma_0$  observed commonly in cuprate superconductors may originate from some other properties, such as inhomogeneity. It may not be induced by the small-scale impurity scattering, otherwise the  $H^{1/2}$  law should not be followed so well. Since both the Simon-Lee scaling law and Volovik's  $\sqrt{H}$  are fol-



FIG. 12. AC susceptibility and DC magnetization of the underdoped sample with p=0.069. The bottom curve is the real part x' of the ac susceptibility, and the upper one is the imaginary part x''. By extrapolating the most steep transition portion of the real part of the AC susceptibility to the normal state background (x'=0), the  $T_c$ =12 K is determined here. The inset shows the DC magnetization measured in the FC and ZFC processes. Below about 7.5 K, the magnetization measured in FC mode is rather stable. The slight temperature dependence of the magnetization measured in ZFC mode is induced by the flux penetration.

lowed very well for the optimally doped sample, the coresize effect seems to be very weak.

## **D. Underdoped sample**

In this section the low-temperature specific heat of underdoped La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (p=0.069, 0.075, 0.09, and 0.11) single crystals is reported in magnetic fields up to 12 *T*. It is found that the Volovik's relation  $C_{vol}=ATH^{1/2}$  is still satisfied in the zero-temperature limit, but the proposed Simon-Lee scaling law, i.e.,  $C_{vol}/T^2 = f(T/\sqrt{H})$ , is not followed so well, except for at very low temperatures (below about 3–4 K).

Figure 12 shows the temperature dependence of the AC susceptibility and DC magnetization of the underdoped sample  $La^{2-x}Sr_xCuO_4$  (*p*=0.069). The transition temperature drops from about 12 K to 9 K after extracting some oxygen out of the sample (not shown here) by annealing the sample in Ar gas for 48 hr. Then  $T_c$  keeps stable on further annealing in Ar gas. The  $T_c$  is increased again when the sample is treated in flowing oxygen. The DC magnetization measured in the FC process shows a transition width of about 2.5 K. Below about 7.5 K the M(T) curve keeps flat. The magnetization measured in ZFC mode shows a slight increase with temperature induced by the easy flux penetration in the very underdoped region. Specific heat has been measured in the FC mode as done for all other samples. This mode provides a vortex system that is close to equilibrium state and, thus, relatively uniform.<sup>36</sup> Presented in Fig. 13 is the specific heat C/T as a function of  $T^2$  at magnetic fields ranging from 0 to 12 T for this underdoped sample before annealing (estimated p=0.069). In the low-temperature region the curves are rather linear, showing that the major part is due to the phonon contribution  $C_{ph} = \beta T^3 + \delta T^5$ , and have no slight upturn due to the Schottky anomaly of free spins. The curve at zero field extrapolates to a finite value ( $\gamma_0$ ) at 0 K, again



FIG. 13. Raw data of C(H)/T vs  $T^2$  for the underdoped sample p=0.069. One can see that the field-induced change of specific heat becomes much smaller than that of the optimally and overdoped sample.

showing the existence of a residual linear term, which will be discussed later. As mentioned before the phonon part is independent of the magnetic field, this allows one to remove the phonon contribution by subtracting the C/T at a certain field with that at zero field, one has  $\Delta C = C(H) - C(0) = C_{vol}$  $-\alpha T^2$  and  $\Delta C/T^2 = C_{nol}/T^2 - \alpha$ . The results after the subtraction are shown in Fig. 14. One can see that the linear part with negative slope, as appearing for the overdoped sample, is absent here. This is understandable when  $\alpha$  value (or  $\alpha T^2$ ) term) is very small compared to the field-induced change of total specific heat. Therefore, for this underdoped sample, no apparent  $T^2$  term at H=0 was observed, which can be found easily in the overdoped LSCO sample. This is consistent with the data shown in Table I and experimental results from other groups on LSCO.<sup>13,14</sup> The disappearance of this  $\alpha T^2$ term was usually interpreted as being due to either the impurity scattering, which smears out the nodal effect, or the small value of coefficient  $\alpha$  of  $T^2$ . We will show that this is induced by a much smaller  $\gamma_n$  value (thus smaller  $\alpha$ ) just above  $T_c$ .

Next let us have a look at the field-induced DOS at T = 0 K. The data  $\Delta \gamma(H, T=0)$  is determined by doing a linear fit to the raw data C/T versus  $T^2$  between 2 K and 4 K, and



FIG. 14. The subtracted data [C(H)-C(0)]/T vs *T* for the underdoped sample (p=0.069). It is clear that no linear part with negative slope of  $\Delta \gamma$  vs *T* as appearing for the overdoped sample can be observed here. This may be induced by the much smaller  $\alpha$  value.



FIG. 15. Field-induced change of  $\gamma$  at 0 K for the underdoped sample with p=0.069. The solid and dashed lines are fits to the theoretical relation of Volovik effect at the clean limit and the impurity scattering at the unitary limit, respectively.

then subtracted the zero-temperature value  $\gamma_0$ . The results are shown in Fig. 15. In order to compare to the theoretical predictions, the increase in  $\gamma(H,T=0)$  was fitted with  $\Delta \gamma(H, T=0) = AH^B$ , and the value of B is 0.52 and A is about 0.28. The value B, derived here from free fitting, is very close to 0.5 as predicted by the Volovik theory,<sup>16</sup> which may manifest the existence of a line node in the gap function. We fix B=0.5 and find out that can also A =0.282 mJ/mol  $K^2T^{1/2}$ . This is also compatible with the results of other groups.<sup>14</sup> For the zero-temperature data we also considered the core-size correction, i.e., tried to use the first term of Eq. (7) to fit the zero-temperature data. But it gives rise to a small and negative value of  $\pi \xi^2 / \phi_0$ , which is certainly unreasonable.

For the underdoped sample, we used the Simon-Lee scaling law to scale our data. The results of  $[C(H)-C(0)]/T\sqrt{H}$  versus  $T/\sqrt{H}$  are shown in Fig. 16. The data fan out showing a poor scaling quality. Clearly, the data cannot be scaled using the Simon-Lee scaling law except for at very low temperatures. We plot also the data of  $[C(H)-C(0)]/T^2 = C_{DOS}/T^2 - \alpha$  versus  $T/\sqrt{H}$  in Fig. 17. One can again see the poor scaling in a wide temperature region. The Simon-Lee



FIG. 16. Scaling of the raw data  $\Delta C/T\sqrt{H}$  vs  $T/\sqrt{H}$  for the sample p=0.069 based on the Simon-Lee scaling law. Clearly, no good scaling can be obtained, except for at very low temperatures.



FIG. 17. Scaling of the raw data  $\Delta C/T^2 = [C(H) - C(0)]/T^2$  vs  $T/\sqrt{H}$  for the sample p=0.069 based on the Simon-Lee scaling law. Good scaling can be found only at very small values of  $T/\sqrt{H}$ .

scaling law has been applied to all samples investigated in this work (p=0.069, 0.075, 0.09, 0.11, 0.15, 0.22). It is easy to find that the scaling quality becomes better and better when the doping concentration is increased from 0.069 to 0.15. One can even see the gradual change among these underdoped samples (p=0.069, 0.075, 0.09, 0.11); the scaling curves fan out like that in Fig. 17 for samples with p=0.069, 0.075, but the scaling pattern becomes narrower toward higher doping. The scaling behaviors are shown in Figs. 18–20 for samples with p=0.075, p=0.09, and p=0.11. A clear trend for a better scaling at a higher doping can be easily seen here.

There are several possibilities for the failure of using Simon-Lee scaling law in an underdoped region. One possibility is due to the impurity scattering effect as suggested by Kübert and Hirschfeld.<sup>30</sup> Thus we use the dirty limit formula  $\gamma(H) = \gamma(0)[1+D(H/H_{c2})ln(H_{c2}/H)]$  to fit the data at 0 K, where  $D \approx \Delta/32\Gamma$ . For simplicity, we show here only the fit to the data of the sample p=0.069. It is found that the data can also be roughly fitted by the relation with impurity scattering (as shown by the dashed line in Fig. 15). The obtained results for the sample with p=0.069 are  $H_{c2}=45.6 T$ ,  $\gamma(0)$ =4.03 mJ/mol K<sup>2</sup>,  $\Gamma/\Delta=0.046$ . Thus, it seems that one can-



FIG. 18. Scaling of the raw data  $\Delta C/T^2 = [C(H) - C(0)]/T^2$  vs  $T/\sqrt{H}$  for the sample p=0.075 based on the Simon-Lee scaling law. Good scaling can be found only at very small values of  $T/\sqrt{H}$ .



FIG. 19. Scaling of the raw data  $\Delta C/T^2 = [C(H) - C(0)]/T^2$  vs  $T/\sqrt{H}$  for the sample p = 0.09 based on the Simon-Lee scaling law.

not rule out the possibility of impurity scattering to play a dominant role in the field-induced change of  $\gamma$  in very underdoped samples. However this speculation cannot interpret the nice  $\sqrt{H}$  dependence of the field-induced DOS at 0 K, as shown in Fig. 15. It is worth noting that the dirty limit formula of Kübert and Hirschfeld<sup>30</sup> is a more flexible fit to the data than the simple  $\sqrt{H}$  relation. One needs to seek an alternative way to clarify this discrepancy.

The second possibility is that of the core size effect appearing in the overdoped sample. We then try to scale the data by using Eq. (8) and leaving both  $\alpha$  and  $\pi \xi^2/\phi_0$  as free-fitting parameters. Unfortunately, no good scaling can be found by choosing any values for  $\alpha$  and  $\pi \xi^2/\phi_0$ . This is inconsistent with the fact that an unreasonable negative value for  $\pi \xi^2/\phi_0$  is obtained if we fit the zero-temperature data in Fig. 15 to the first term of Eq. (7). Both indicate that the failure of the Simon-Lee scaling law here is not due to the core-size effect. One may argue that the data are scalable within only a very narrow scaling region of  $T/\sqrt{H}$ , for example, from Figs. 16–18, the scalable region is about  $T/\sqrt{H} \leq 1.5 \text{ KT}^{-0.5}$ . This is, of course, possible because we do not know the precise value for many parameters. However, we can have a rough estimation to check whether this is



FIG. 20. Scaling of the raw data  $\Delta C/T^2 = [C(H) - C(0)]/T^2$  vs  $T/\sqrt{H}$  for the sample p=0.11 based on the Simon-Lee scaling law. Now the fanning-out of the scaling curves are strongly constrained, showing a better scaling behavior.

reasonable. Provided the scalable region is  $T/\sqrt{H} \leq T_c/\sqrt{H_{c2}(0)} = 1.5$ , inputting  $T_c = 12$  K, one has  $H_{c2}(0) = 64$  T, which seems too big for this very underdoped sample.

Another possible reason for the failure of the scaling law is that the sample is in the underdoped region with a pseudogap in the normal state. When the sample is in the mixed state, some competing or coexisting order, such as the short-range antiferromagnetic order<sup>19,20,37-41</sup> or the SDW order<sup>42</sup> or a *d*-density-wave (DDW) order<sup>43</sup> is enhanced, and this enhanced order will certainly contribute to the total specific heat. For example, for the 2D AF correlation, it is known that  $C_{AF} \propto T^2$ . Therefore, qualitatively, the failure of the Simon-Lee scaling law in the underdoped region can be understood in the following way. By increasing the magnetic field, a second order is generated or enhanced within the vortex core and nearby regions (about 100 Å). On one hand this region is gapped, leading to a decrease of the total DOS at the Fermi level simply by reducing the region where the supercurrent can flow. On the other hand, the AF or SDW or DDW region will contribute a different term to the total specific heat due to spin or other-type excitation. The relevant competing order under a magnetic field, according to both neutron scattering<sup>19,20</sup> and NMR measurement,<sup>7</sup> may be the AF order. STM measurement by Hoffman et al.44 indicates a checkerboardlike modulation with periodicity of 4a of the LDOS. This was regarded as the direct observation of the strong electronic correlation with the underlying competing which was predicted by many theoretical order, works.<sup>38–41,45–48</sup> This qualitative picture calls for further detailed analysis and evidence from other experiments. Since the heat capacity from the enhanced second order has a temperature dependence of  $T^{\epsilon}$  with  $\epsilon > 1$ , at zero temperature the specific heat from this term is zero, thus the  $H^{1/2}$  law from the Doppler shift of the *d*-wave superconductivity is restored. This may be the reason for that the zero-temperature data follows the  $H^{1/2}$  law, but the data at finite temperatures do not satisfy the Simon-Lee scaling law very well.

## E. The residual linear term $\gamma_0$

Almost in all cuprate superconductors, a residual linear term of electronic specific heat  $\gamma_0$  has been observed in the low-temperature limit  $T \rightarrow 0$ , even in the best samples to date. In YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> single crystals, Moler *et al.*<sup>8</sup> found that  $\gamma_0$  is larger for the twinned samples than for the detwinned ones. Meanwhile, they further found that  $\gamma_0$  increases when the sample becomes more underdoped. Clearly, one can conclude that  $\gamma_0$  is related to the impurities or disorders in the samples. While quite surprisingly, for many samples with quite different  $\gamma_0$  values; it is found that the zero-temperature data can be expressed as  $\gamma(H, T=0) = \gamma_0 + A\sqrt{H}$ , showing evidence for *d*-wave pairing. This may suggest that  $\gamma_0$  is not mainly induced by the impurity scattering, otherwise the field-induced extra DOS should not follow the relation  $\Delta C(H,T=0)/T=A\sqrt{H}$  so well. In Fig. 21, we present the field dependence of  $\Delta \gamma = [C(H, T=0) - C(0, T=0)]/T$  normalized to the value for each sample at about 12 T. Meanwhile we show the  $H^{1/2}$  law by the solid line. One can see



FIG. 21. Field dependence of the field-induced extra  $\gamma$  normalized by the value at 12 *T* at 0 K. The solid line represents the theoretical curve  $H^{1/2}$ . It is found that the data from different samples at different doping levels follow the  $H^{1/2}$  law reasonably well.

that for almost all samples, the field-induced extra DOS at 0 K follows the  $H^{1/2}$  relation reasonably well despite the  $\gamma_0$ value highly disperses. This feature was also discovered by Chen et al.<sup>14</sup> on three typical samples (x=0.10, 0.16, and0.22). Nohara *et al.*<sup>13</sup> measured three single crystals (x=0.10, 0.16 and 0.19) and empirically found that the optimally doped sample (x=0.16) has the lowest value of  $\gamma_0$  $(\gamma_0 = 2.8, 1.5, \text{ and } 2.2 \text{ mJmol}^{-1} \text{ K}^{-2} \text{ for } x = 0.10, 0.16, \text{ and}$ 0.19, respectively, obtained from Fig. 1 of Ref. 13). Chen et  $al.^{14}$  found similar behavior among three samples with x =0.10, 0.16, and 0.22 ( $\gamma_0$ =1.49, 0.7, and 1.41 mJmol<sup>-1</sup> K<sup>-2</sup>, respectively). This raises the question of the origin of this residual linear term and its correlation with the field-induced quasiparticle DOS. As mentioned before, if the field-induced DOS is related to the impurity scattering, another relation<sup>30</sup>  $\delta \gamma / \gamma_0 = P_1(H/P_2) log(P_2/H)$  is expected. This is sometimes contradicting to the experiment result (see dotted line in the main panel of Fig. 5). In addition, the  $\sqrt{H}$  dependence of the field-induced change of  $\gamma$  is certainly not obtained by accident, since it is found on different samples from different groups, even on polycrystalline samples.<sup>14</sup> From the view point of chemistry, it is not true that the optimally doped sample is the cleanest one because, in most cases, the underdoped samples can be more easily grown with high quality. In this sense, the residual linear term may be related to some other properties rather than the impurity scattering.

In Fig. 22 the  $\gamma_0$  values for different single crystals measured in our experiment from underdoped to overdoped are shown. The value of  $\gamma_0$  is obtained by fitting the zero field data to Eq. (2) (see Table I). It is clear that the minimum  $\gamma_0$ is found in the region around 0.11 or 0.125. The value for  $\gamma_0$ found from our data are more close to the data of Nohara *et al.*<sup>13</sup> on single crystals, but clearly higher than that obtained on polycrystalline samples.<sup>14</sup> Thus far, we do not know the reason for this discrepancy. For our extremely underdoped sample (x=0.063) investigated here, although the data at finite temperatures cannot be treated with the Simon-Lee scaling law, the data in the low-temperature limit  $T \rightarrow 0$ , however, can still be nicely expressed by  $\gamma(H, T=0) = \gamma_0 + A \sqrt{H}$ ,



FIG. 22. The doping dependence of  $\gamma_0$  of La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> crystals. It is clear that the minimum  $\gamma_0$  value is found in the region around p=0.11 to 0.125.

even the absolute increase of  $\gamma(H,T=0)$  by field is much smaller than  $\gamma_0$ . Therefore, it is reasonable to conclude that the field-induced part is mainly contributed by the Dopplershift effect on the supercurrent outside the vortex cores, while the residual linear term  $\gamma_0$  is mainly contributed by some small normal regions, which weakly depends on the magnetic field. Similar explanations to the origin of  $\gamma_0$  have been suggested many times in the past.<sup>49</sup> This may be understood in the following way. In underdoped Bi-2212 single crystals, scanning-tunneling-microscopic (STM) measurement indeed reveal a mixture of superconducting regions with sharp quasiparticle coherent peaks on the tunneling spectrum, and the nonsuperconducting regions with pseudogaplike tunneling spectrum.<sup>50</sup> In the overdoped side, the tiny normal cores as proposed in the Swiss-cheese model,<sup>51</sup> or the mesosscopic normal regions suggested by Fukuzumi et al.<sup>52</sup> and Wen et al.,<sup>53</sup> will contribute a residual term  $\gamma_0$  which does not show an apparent increase with the field. As proposed by Fukuzumi et al.52 that the domelike electronic phase diagram may be formed by the mixture of three phases: the antiferromagnetic phase in the extremely underdoped region, a d-wave superconducting region with the robust superconductivity near the optimal doping point, and a nonsuperconducting Fermi liquid in the overdoped region. According to this simple picture the  $\gamma_0$  should increase in the underdoped and overdoped regions, which is just the case as shown by the data in Fig. 22. Therefore, we would argue that the residual linear term may be mainly contributed by some nonsuperconducting regions due to phase separation, either chemical or electronic in origin. This interesting argument needs certainly to be further checked with data obtained by different techniques on different systems.

# F. Field-induced reduction of specific heat in high-temperature regions

In above analysis, we concentrate on the data below 10 K (below 6 K for the very underdoped sample). This is also the temperature region in which most of the low-temperature specific heat data was reported in the literature. Now we



FIG. 23. Temperature dependence of the field (12 T) induced change of  $\Delta \gamma = [C(12T) - C(0T)]/T$  vs T for three typical samples (x=0.22, overdoped: filled circles; x=0.15, optimally doped: diamonds; x=0.069, underdoped: triangles). The horizontal axis is normalized to  $T_c$  of each sample. In the high-temperature regime below  $T_c$ , the field-induced change of DOS becomes negative.

report another phenomenon: field-induced reduction of specific heat in the mixed state. In Fig. 23, we present the temperature dependence of the field (12 T) induced change of  $\gamma$ for three typical samples analyzed above, here  $\Delta \gamma$ =[C(12T)-C(0T)]/T. Although the data are strongly scattered, one can still see that: (1) the field-induced change  $\Delta \gamma$ becomes negative at about  $0.5-0.7 T_c$ ; (2) the curves have a similar shape:  $\Delta \gamma$  is positive in low-temperature region, then it becomes negative and finally comes back to zero in hightemperature region (near  $T_c$  for optimal and overdoped sample). For the overdoped sample, the  $\Delta \gamma$  keeps negative above 0.5 T<sub>c</sub> until T<sub>c</sub> at which  $\Delta \gamma$  suddenly goes back to zero. For the optimally doped sample, the  $\Delta \gamma$  is negative above about 0.7  $T_c$  up to the highest temperature we measured here (30 K). However for the underdoped sample, it shows that the  $\Delta \gamma$  keeps negative until 1.5 T<sub>c</sub>. Similar data were obtained by Fisher *et al.*<sup>11</sup> on samples with x=0.15. Our data near  $T_c$  is more scattered because our setup can only measure samples with maximum mass of 50 mg. This feature, namely, the negative  $\Delta \gamma$  in the high-temperature region, is a consequence of entropy consideration, which has been observed in all types of superconductors. In the lowtemperature region, when a magnetic field is applied, vortices will be generated leading to higher DOS near the Fermi surface, so that  $\Delta \gamma = \gamma(H) - \gamma(0)$  is positive. When the temperature is increased to satisfy the field-independent entropy above  $T_c$ , in a certain region below  $T_c$ ,  $\Delta \gamma$  should be negative. The most interesting point is that for the underdoped sample here, even above  $T_c$ , one clearly sees a magnetic field-induced change of entropy. This implies an abnormalnormal state, which is far from a conventional metal. For a conventional s-wave superconductor, the field-induced change of  $\gamma$  can be negative near  $T_c$ . It is difficult to understand the field-induced reduction of specific heat well below  $T_c$ , since the normal core region always gives rise to a higher DOS of quasiparticle. Outside the vortex core, the DOS is almost negligible. However, this field-induced reduction of

specific heat well below  $T_c$  is found to be a general feature of all LSCO crystals we investigated thus far. This may be related to the intrinsic properties of cuprate superconductors. In a *d*-wave superconductor, theoretically it is predicted that there is a ZBCP within the vortex core, which should also contribute a quite high DOS<sup>22,23</sup> in the mixed state. Besides, a high DOS will be generated by the Doppler-shift effect of the supercurrent surrounding the vortex core. Normally the sum of these two terms are larger than the zero-field term  $C_{DOS} = \alpha T^2$ , leading to a field-induced enhancement of DOS in the low-temperature region. When the temperature is high, the Doppler-shift effect will be smeared out by strong thermal excitation, and, finally,  $\Delta \gamma$  becomes negative. As far as we know, no quantitative theoretical expression about  $\Delta \gamma$ has been reported thus far for a *d*-wave superconductor in a wide temperature region. We cannot have a quantitative understanding to our data. However, this field-induced reduction of specific heat well below  $T_c$  may be understood as being due to the anomalous feature of the vortex core state, i.e., a gapped vortex core as seen by the STM,<sup>26</sup> or based on the assumption that the contributions from the core region is much smaller than the outside region where either the Doppler shift or the strong thermal excitation dominates. Actually, the Simon-Lee scaling law becomes a  $C_{vol} \propto T^2$  relation in the high-temperature region. In this case the quasiparticle excitation outside the vortex core is almost the same  $(\alpha T^2)$ with or without applying a magnetic field. However, since the vortex core region is gapped or contributes a negligible part to the total DOS, one needs to take the core region away from the total area in calculating  $\Delta \gamma$ , naturally leading to a negative value of  $\Delta \gamma$ .

## **IV. DISCUSSION**

In the low-temperature region, our analysis indicates that the field-induced quasiparticle DOS can be well described by Volovik's theory or the Simon-Lee scaling law, although a correction due to the core-size effect is needed for the overdoped sample. This means that the prerequisite for the theory, i.e., the  $d_{x^2-y^2}$  pairing symmetry is well satisfied. Therefore, it naturally rules out the presence of a secondorder parameter, such as  $id_{xy}$  or is, either due to overdoping<sup>6</sup> or to the field effect<sup>54</sup> in all samples investigated here. Meanwhile, for the overdoped sample, another interesting phenomenon is that the vortex core region contributes very little (at least much smaller than that induced by the Doppler shift if the supercurrent would flow in the same area) to the total DOS. We have also tried to analyze the data of the optimally doped and underdoped samples in a way that for the overdoped one, for example, to fit the data in Figs. 11 and 15 to the first term in Eq. (7). It turns out, however, that the correction term  $\pi \xi^2 H/\Phi_0$  derived is small and negative, which is unreasonable. For the optimally doped sample, it is quite easy to understand because the vortex core becomes very small. However, for the underdoped sample, it is quite hard to understand because the core size tends to grow up too.<sup>32</sup> The negligible contribution from the vortex core region may suggest that the ZBCP is absent within the cores, even in the overdoped region. This suggestion inferred from the specific



FIG. 24. The doping dependence of the prefactor A in  $\gamma(T=0) = \gamma_0 + AH^B$ ,  $B \approx 0.5$ . It is evident that the A-value increases with the hole concentration, monotonously. The data measured on polycrystalline samples are somewhat smaller, which is perhaps induced by the random orientation of the grains. For some grains the field is not parallel to the *c*-axis, leading to a smaller contribution to the field-induced change of  $\gamma$ .

heat measurement about the ZBCP within the vortex core is consistent with the tunneling results<sup>24–28</sup> and certainly clears up the concerns about the surface conditions in the STM measurement. Recent results from NMR also show the absence of a ZBCP inside the vortex core.<sup>29</sup> In this sense our data together with the earlier NMR data present a piece of evidence from bulk measurements for an anomalous vortex core. Interestingly, it is widely perceived that the normal state in overdoped region shows a Fermi-liquid behavior even when the superconductivity is completely suppressed.<sup>55</sup> If this is the case, the mean-field frame of BdG theory based on the conventional *d*-wave superconductivity seems not enough to interpret the anomalous vortex core state in HTS. For the underdoped sample, the Simon-Lee scaling fails except for in very low-temperature regions. This is interpreted as due to the presence of a second (gapped) order, such as AF, SDW, or DDW, within and nearby the vortex core. However, one needs more theoretical and experimental efforts to show the justification for this argument.

By fitting the field-induced extra DOS at zero temperature to the relation  $\Delta \gamma = AH^{1/2}$ , we obtained the prefactor A in the wide-doping regime, where  $A = 0.74 \gamma_n / \sqrt{H_{c2}}$ .<sup>17</sup> The results are presented in Fig. 24. It is seen that the A-value increases with the doping concentration, monotonously. This can be understood in the following way: by increasing doping the normal state value  $\gamma_n$  will increase,<sup>56</sup> the  $H_{c2}$  will drop down (at least it is the case in the overdoped region). Therefore the A value will increase monotonously in the overdoped side. One can see from the data that the A-value keeps almost constant in the extremely underdoped region, which means that  $\gamma_n$  and  $H_{c2}$  should both decrease with underdoping. This indicates that the  $H_{c2}$  becomes smaller, and the coherence length  $\xi$  becomes larger toward more underdoping. This is consistent with the recent conclusion drawn by Wen et al.<sup>32</sup> by analyzing the data about the low-temperature flux dynamics. This conclusion about the coherence length calls for a direct check to the vortex core size by using scanning tunneling microscopy in the future.

# V. CONCLUDING REMARKS

In conclusion, the field-induced change of the electronic specific in the mixed state of a series  $La_{2-r}Sr_rCuO_4$  single crystals has been measured and extensively analyzed. It is found that the field-induced DOS of the optimally doped sample fits the predicted Simon-Lee scaling law for a *d*-wave superconductor very well, while deviations have been found for the overdoped sample. However, it is reconciled for the overdoped sample if one considers the core-size effect provided the contribution from the inner vortex core is small compared to that due to the Doppler shift in the same area. The Simon-Lee scaling law is applicable in the underdoped region only in very low-temperature regions. We attribute this to the appearance of a second competing order (such as AF, SDW, or DDW) within and nearby the vortex core. The negligible contribution from the vortex core region may suggest the absence of the ZBCP in the vortex core, even in the overdoped region, although it is expected by the Bogoliubov-de Gennes theory for a *d*-wave superconductor. Finally, we present the doping dependence of the residual linear term  $\gamma_0$  commonly observed in cuprate superconduct-

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ors. It is argued that this linear term may be related to inhomogeneity (either electronic or chemical), rather than be simply explained as due to the small-scale impurity scattering as usually thought. This conclusion is made because the field-induced extra DOS at zero temperature follows the Volovik's  $\sqrt{H}$  law reasonably well in all doping regime. It is hard to believe that this nice consistency is obtained by accident. Our results generally conclude a *d*-wave pairing symmetry for the hole-doped La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> samples, although some competing orders may coexist with the superconductivity, and an anomalous feature (missing of the ZBCP) may appear within the vortex core.

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