

Reversible magnetic properties of $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_4$ single crystals with $0.05 \leq x \leq 0.10$

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Systematic magnetization measurements were made to study reversible magnetic properties of a series of high-quality, large single crystals of $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_4$ over a composition range of $x = 0.05-0.10$, with magnetic fields parallel to the c axis. A method of data analysis, based on the variational model developed by Hao and Clem, was used for the determination of various superconducting parameters of this family. We fit the temperature dependence of the thermodynamic critical field $H_c(T)$ data, which was obtained in the temperature region where the thermodynamic fluctuation is insignificant, with the BCS or an empirical function for $H_c(T)$. We found that a crystal ($x = 0.077$), which has the highest T_c (≈ 35.0 K), shows a maximum in the value of $H_c(0)$ ($= 2510 \pm 80$ Oe) and a minimum in the value of $\lambda_{ab}(0)$ ($= 2545 \pm 40$ Å) for the range of x that we have studied. We also observed rather close values of coherence length in the ab plane [$\xi_{ab}(0) \sim 32$ Å] for all $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_4$ single crystals that we studied.

I. INTRODUCTION

Although a large effort has been devoted to the measurements of both the normal and the superconducting properties of $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_4$ (hereafter called LSCO), rather few studies have encompassed measurements of the reversible magnetic properties of the superconducting LSCO family over a wide range of Sr content. This is partly due to the difficulties in synthesizing high-quality crystals and complexities associated with extracting reliable data for subsequent data analysis due to the large crystallographic anisotropy in these layered superconductors. Earlier, following the discovery of high- T_c superconductors, the measurements of the superconducting parameters of the LSCO system, were primarily based on polycrystalline samples with the composition of $(\text{La}_{0.925}\text{Sr}_{0.075})_2\text{CuO}_4$. Widely varying results were reported with lack of agreement due to the grain-boundary effects, the anisotropy, and compositional inhomogeneities.¹⁻⁵ Growth of high-quality large LSCO single crystals using a traveling solvent floating zone technique^{6,7} permits a systematic study of the magnetic properties of LSCO with various Sr contents.

It is known that all of the superconducting parameters can be determined through a complete measurement of the reversible magnetization of a superconductor as a function of magnetic field and temperature.⁸ In this paper, we present the results of our reversible magnetization measurements on a series of high-quality large (a few mm along the c axis) LSCO single crystals over a composition range from $x = 0.05$ to $x = 0.10$. We report the superconducting parameters for these crystals which are determined by a modified use of the Hao-Clem variational model⁹ for the magnetization of high- κ type-II superconductors.

The reversible magnetization of a type-II superconduc-

tor was first quantitatively studied by Abrikosov,¹⁰ in the magnetic field range $H - H_{c1} \ll H_{c1}$ and $H_{c2} - H \ll H_{c2}$, where H_{c1} and H_{c2} are the lower and upper critical fields, respectively. Conventionally, some superconducting parameters, like $H_{c2}(T)$ and κ , are determined from the traditional extrapolation method based on the linear Abrikosov formula [Eq. (1)] for magnetization at high field near H_{c2} ,

$$-4\pi M = \frac{H_{c2}(T) - H}{(2\kappa^2 - 1)\beta_A}, \quad (1)$$

where β_A is 1.16 for a hexagonal array of vortices. The temperature region, for which Eq. (1) is valid, is usually taken to be between $T_c(H)$ and $T_A(H)$, where $T_c(H)$ is the mean-field transition temperature at H and T_A is the temperature at which the applied field $H \approx 0.4H_{c2}(T_A)$. However, this temperature region for high- T_c cuprates is quite limited because of their large values of $dH_{c2}(T)/dT$. Furthermore, as recently reported,^{11,12} the large fluctuation effect near T_c in high- T_c superconductors significantly distorts the Abrikosov-like behavior of magnetization in this temperature region so that the conventional extrapolation method becomes inapplicable for the determination of superconducting parameters for many of the high- T_c superconductors, e.g., $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (Ref. 11) and $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$.¹² In the case of the LSCO system, we also observed a significant deviation of magnetization from the Abrikosov behavior, as it will be discussed in Sec. III.

In the intermediate field $H_{c1} \ll H \ll H_{c2}$, the London model¹³ provides a simple description for the mixed state of extreme type-II superconductors ($\kappa \gg 1$). κ is defined as λ/ξ , where λ is the magnetic penetration depth, and ξ is the superconducting coherence length. Since the coherence length of a high- κ superconductor is normally

much smaller than the spacing between vortex cores, the energy associated with the depression of the order parameter to zero on the axis of vortices is normally not accounted for by the London model. Within this assumption, the penetration depth $\lambda(T)$ of a high- T_c superconductor can be obtained from the slope of straight lines of $4\pi M$ versus $\ln(H)$ via the following equation,¹⁴

$$\frac{d[4\pi M(T)]}{d(\ln H)} = \frac{\phi_0}{8\pi\lambda_{ab}^2}, \quad (2)$$

where H is parallel to the c axis, and ϕ_0 is the flux quantum $hc/2e$. One of the shortcomings of the London approximation is that many other important superconducting parameters (for instance, $H_c(T)$, ξ , etc.) cannot be determined from magnetization measurements. Moreover, it was shown that the London approximation generally overestimates the value of $\lambda(T)$ by 6–20%.^{14,15,16}

In order to improve this situation, Hao and Clem^{9,15} recently proposed a variation model for the reversible magnetization of type-II superconductors as a function of the magnetic field for the entire mixed state. Within this model, the depression of the order parameter at the vortex core is explicitly taken into account for calculating magnetization. The two fitting parameters in this model are $H_c(T)$ and κ . Many fundamental superconducting parameters, such as ξ , $\lambda(T)$, and $H_{c2}(T)$, can be determined from these two variables. Furthermore, we have demonstrated in our earlier work^{12,17} that this model, which is basically a three-dimensional anisotropic Ginzburg-Landau theory, is still applicable for studying the mixed-state magnetic properties of quasi-two-dimensional superconductors like $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$, at least in magnetic fields applied perpendicular to the CuO layers. However, the Hao-Clem model does not address the thermodynamic fluctuation of order parameters in high magnetic fields¹⁸ so that the strong fluctuation effect could make this model inapplicable, particularly near T_c , as reported previously in the study of the reversible magnetic properties of a superconducting $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$.¹⁷ In order to avoid the complications in the analysis of the magnetization data caused by the fluctuation effects, we have proposed a new data analysis technique,¹² which enabled us to determine many of the fundamental superconducting parameters for $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$, which avoid

fluctuation effects. The basis of this technique is to determine the temperature region where the fluctuation effects make negligible contributions to the total magnetization, then apply the Hao-Clem model to the reversible magnetization data taken in this temperature region. A detailed description of this technique can be found in Ref. 12. In Sec. III we will demonstrate the use of this technique in determining superconducting parameters of the LSCO family.

II. EXPERIMENTS

A series of LSCO single crystals, up to 5 mm along the c axis, and over a wide range of Sr content, were grown by a traveling solvent floating zone technique reported previously.⁶ The four specimens used in this study were cut from these crystals. The Sr content of each crystal was analyzed by the inductively coupled plasma (ICP) technique. Table I lists the nominal compositions and Sr contents for the four crystals: *A* (51.2 mg, 3.20 mm along c axis), *B* (29.0 mg, 2.00 mm along c axis), *C* (13.85 mg, 1.4 mm along c axis), and *D* (32.5 mg, 1.4 mm along c axis), respectively. The existence of a single domain in each of the four crystals was confirmed by various techniques reported before.⁶ Low-field dc magnetization shows a sharp transition for each crystal, with the full shielding volume observed at low temperatures.¹⁶ This suggests that a rather uniform Sr content was achieved in these large crystals. In Table I, we also listed the value of T_c at the onset, and the transition width ΔT_c for each sample.

The magnetization measurements were carried out in magnetic fields applied parallel to the c axis by using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design). The magnetometer was calibrated with pure MnF and Al standards (National Institute of Standard and Technology). Field inhomogeneity is estimated to be no greater than 0.05% for the 3-cm scan length with which all the magnetization data were taken.

The irreversible temperature, $T_i(H)$, for magnetic fields applied parallel to the c axis was measured first to define the reversible magnetization region. All the reversible magnetization data were taken by measuring the magnetic moments as a function of temperature at fixed

TABLE I. Composition, T_c , Ginzburg-Landau parameter κ , thermodynamic critical fields $H_c(0)$, upper critical fields $H_{c2}(0)$, $-dH_{c2}/dT$ near T_c , penetration depth $\lambda_{ab}(0)$, and coherence length $\xi_{ab}(0)$ of $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_4$ single crystals.

Sample	Sr content, x		T_c (K)		$H_c(0)$		$H_{c2}(0)$ (T)		$-dH_{c2}/dT^c$		$\lambda_{ab}(0)^d$	$\xi_{ab}(0)^e$
I.D.	Nominal	ICP	Onset	ΔT_c	Mean field	κ	(Oe)	Clean ^a	Dirty ^b	(T/K)	(Å)	(Å)
<i>A</i>	0.05	0.046	26.2	0.9	28.23±0.54	119	1430	31±2	33±3	1.60±0.15	4400±20	32±3
<i>B</i>	0.0625	0.059	32.4	3.7	32.10±0.21	85	1430	22±2	23±2	1.00±0.10	3730±30	38±3
<i>C</i>	0.085	0.077	35.1	2.6	34.39±0.14	70	2510	32±2	34±2	1.35±0.06	2545±40	32±2
<i>D</i>	0.10	0.090	29.8	1.8	29.81±0.09	75	2170	31±4	33±4	1.50±0.16	2830±20	33±4

^a $H_{c2}(0)$ in clean limit (Ref. 20) is obtained from equation (Ref. 20), $H_{c2}(0)=0.5758[\kappa_1(0)/\kappa]T_c[-dH_{c2}(T)/dT]_{T_c}$, where $\kappa_1(0)/\kappa=1.26$.

^b $H_{c2}(0)$ in dirty limit (Ref. 22) is obtained from the same equation above with $\kappa_1(0)/\kappa=1.20$.

^c $-dH_{c2}(T)/dT$ near T_c is obtained from the temperature fit of $H_c(T)$ data, via equation $H_{c2}(T)=\kappa\sqrt{2}\cdot H_c(T)$.

^dObtained through the relation $\sqrt{2}\cdot H_c=\kappa\phi_0/2\pi\lambda_{ab}^2$, where ϕ_0 is one flux quantum, equal to 2.07×10^{-7} G cm².

^eObtained through the relation $H_{c2}(0)=\phi_0/2\pi\xi_{ab}(0)^2$.

magnetic fields in the temperature region from T_c up to 85 K, with a 10-min delay after each temperature change to stabilize the system. The background signals and normal-state magnetization were carefully subtracted by using the extrapolation of a curve fitted to the measured magnetic moment-versus-temperature data between 60 and 85 K, a method similar to that used in a study of a c -axis-oriented $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ tape.¹² Thus, the magnetization data shown in this paper are of the superconducting state of LSCO free of background.

III. RESULTS AND DISCUSSIONS

Shown in Figs. 1(a) and 1(b), are two plots of $4\pi M$ -versus- T data for (a) crystal C with $x=0.077$ and (b) crystal D with $x=0.090$, respectively. The presence of large fluctuation effects near T_c are clearly demonstrated by the fluctuation-induced diamagnetization in the vicinity of T_c at high fields, for instance 50 000 Oe, as well as the crossover in the $4\pi M$ -versus- T curves for various fields. It was also observed that the effect of the fluctuation is enhanced with the decreasing Sr content as illustrated in Figs. 1(a) and 1(b). The relative intensity of the fluctuation effect on the magnetization as a function of the Sr content can be assessed by comparing the values of

the diamagnetization ($4\pi M^*$) at the crossover point of various $4\pi M(T)$ curves as shown in Fig. 1. For crystal A ($x=0.046$), we found the largest $4\pi M^* \approx -1.22$ G, while for crystal B ($x=0.059$), C ($x=0.077$), and D ($x=0.090$), the values of $4\pi M^*$ are approximately -0.61 , -0.49 , and -0.17 G, respectively. These values are related to the electronic mass anisotropy factor γ . The larger the value of $-4\pi M^*$ is, the greater the anisotropy is. Furthermore, from $-4\pi M^*$ value, it is possible to deduce the value of γ .¹⁹ This aspect of the study will be reported elsewhere.

The thermodynamic fluctuations in high- T_c superconductors, which are intrinsic to the layered structures, can significantly distort the Abrikosov-like mean-field behavior of magnetization near T_c .^{11,12} However, it was also expected, and found, that as temperature decreases away from T_c , the effect of fluctuation diminishes. At sufficiently low temperatures, the induced diamagnetism from the fluctuation eventually becomes negligibly small as compared to the total magnetization.¹² In the temperature region where the fluctuation has negligible effects on the magnetization, the mean-field theory can still be applied to study the reversible magnetic properties. Determination of this region can be accomplished by examining the temperature dependence of κ , and this is the starting point of our new data analysis technique.¹²

To obtain the temperature dependence of κ , Eq. (20) of Ref. 9, together with $-4\pi M = H - B$, is used to fit the data of magnetization $4\pi M(H)$ -versus- H at each fixed temperature, via the Hao-Clem variation method.⁹ The fitting parameters are κ and $H_c(T)$. The experimental value of κ at each temperature is determined to give the smallest deviation in $H_c(T)$. The result of the fitting to the reversible $4\pi M(H)$ data for crystal C is shown in the inset of Fig. 2 where κ is plotted as a function of temperature. The value of κ is nearly constant at low temperatures with a value of 68.95 at 26.2 K, and then slowly increases at temperatures above 28.2 K. Finally, κ diverges

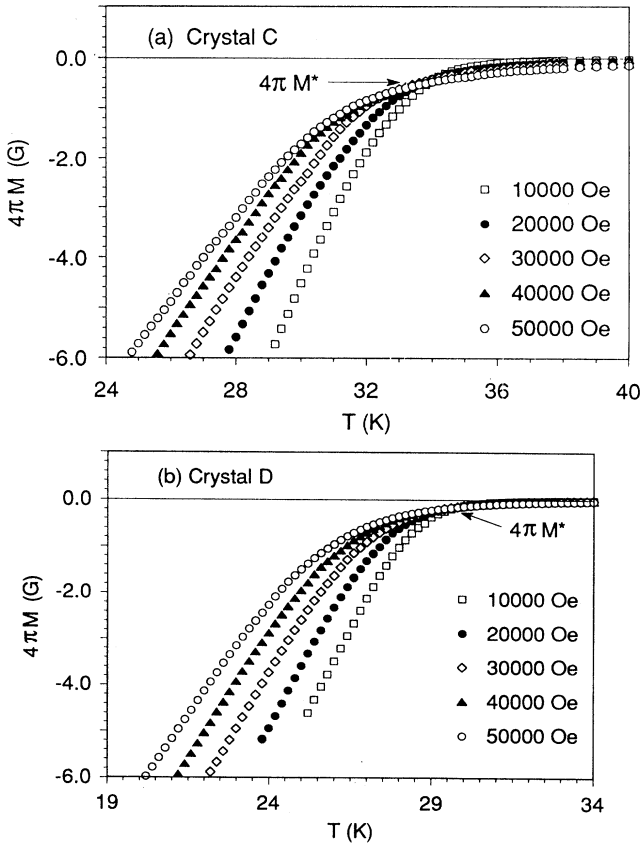


FIG. 1. Temperature dependence of magnetization for LSCO single crystals (a) C and (b) D , measured in various magnetic fields applied parallel to the c axis.

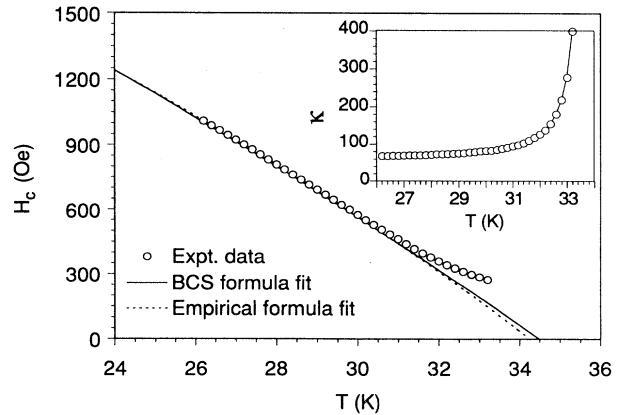


FIG. 2. Temperature dependence of the experimental data $H_c(T)$ (shown as open circles) for single crystal C , obtained using the Hao-Clem variational model with a κ value of 69.8. The solid line shows a BCS formula fit, and the dashed line shows an empirical formula fit. The inset shows the temperature dependence of κ for the same crystal.

around 33.2 K, which is the temperature of the crossover point of all $4\pi M$ -versus- T curves in Fig. 1(a). Essentially, similar behavior of κ as a function of temperature was observed for all of the crystals we studied, and is also very similar to that for $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ reported in Ref. 12. In conventional type-II superconductors, the value of κ slowly increases with decreasing temperature.²⁰ This peculiar temperature dependence of κ observed here reflects the influence of fluctuation on the magnetization of the high- T_c superconductors. More detailed discussions on this matter can be found in Refs. 12 and 16. At temperatures below 28.4 K for crystal C, the constant value of κ indicates that fluctuations have a negligible effect on the reversible magnetization data at the fields up to 50 000 Oe, such that the Hao-Clem model can be applied to determine some of the superconducting parameters of crystal C.

The solid circles shown in Fig. 2 are the values of $H_c(T)$ of crystal C derived from the Hao-Clem fitting using the average value of κ (69.8) between 26.2 and 28.2 K. It must be pointed out here that the values of $H_c(T)$ determined in this manner are affected very little by the small change of κ . We found that a change of κ value from 69.8 to 71.8 only increases the derived value of $H_c(T)$ by 0.7%. Thus, we believe that a small error in κ (a few percent may come from the fitting uncertainty) does not lead to a significant change in the value of $H_c(T)$ reported here. In order to determine $H_c(0)$ and the mean-field $T_c(0)$, we fitted the $H_c(T)$ data between 26.2 and 28.2 K for crystal C with the BCS temperature-dependent formula for $H_c(T)$ given by Clem,²¹

$$\frac{H_c(T)}{H_c(0)} = 1.7367 \left[1 - \frac{T}{T_c} \right] \left[1 - 0.2730 \left[1 - \frac{T}{T_c} \right] - 0.0949 \left[1 - \frac{T}{T_c} \right]^2 \right], \quad (3)$$

and an empirical formula

$$\frac{H_c(T)}{H_c(0)} = 1 - \left[\frac{T}{T_c} \right]^2. \quad (4)$$

The results are shown in Fig. 2. The fitting procedure yields $H_c(0) = 2588 \pm 6$ Oe with $T_c(0) = 34.50 \pm 0.02$ K for the BCS expression and $H_c(0) = 2436 \pm 3$ Oe with $T_c(0) = 34.27 \pm 0.01$ K for the empirical expression. As shown in this figure, the fits by the BCS formula and the empirical formula for $H_c(T)$ are very similar. But above 30.6 K small deviations between the two fits appear, and the derived values of $H_c(T)$ are higher than those from both fitting curves. This is because the fitting curves of $H_c(T)$ are based on the data taken at low temperatures ($T \leq 28.2$ K), where fluctuation effects are negligibly small. Near T_c , fluctuation induces excessive diamagnetic moments, which introduces an increased "apparent" condensation energy [$\propto H_c^2(T)$] into the system. Thus, the experimental values of $H_c(T)$ (including the fluctuation effects) near T_c are higher than the corresponding value of $H_c(T)$ (Fig. 2), represented by the fitting lines which were obtained as if there were no fluctuation effect

involved.

The thermodynamic critical fields $H_c(T)$ and mean-field $T_c(0)$ for the other three single crystals are determined in the same way as for crystal C. The temperature dependence of $H_c(T)$ for other crystals was also found to be well fitted with either the BCS formula or the empirical one. Due to the short reversible magnetization regime for the LSCO family, the $H_c(T)$ data does not cover sufficient temperature region to determine which model would be more appropriate to describe the actual temperature dependence of $H_c(T)$ of superconducting LSCO. However, for each of the four crystals that we studied, the values of $H_c(0)$ and $T_c(0)$, which are obtained from fitting both the BCS and the empirical formula to the experimental data of $H_c(T)$, are rather close. For instance, the difference in the value of $H_c(0)$ for crystal C determined from these two temperature fittings is less than 6%, while the difference in $T_c(0)$ is less than 0.25 K. Thus, the average values obtained from these two fittings are taken as our experimentally determined values of $H_c(0)$ and the mean-field $T_c(0)$ for each of the four crystals. These values are listed in Table I, where the errors include the difference in the results of the two fitting methods.

The determined values of $H_c(T)$, the mean-field $T_c(0)$, and κ for each LSCO crystal enable us to estimate many other superconducting parameters of the LSCO system, such as the upper critical fields $H_{c2}(0)$, $-dH_{c2}/dT$ near T_c , penetration depth $\lambda_{ab}(0)$, and coherence length $\xi_{ab}(0)$. These values for each crystal are also listed in Table I. It is interesting to note that our derived value of $\lambda_{ab}(0)$ of crystal C with a Sr content at $x = 0.077$ is quite close to the value of $\lambda(0)$ (≈ 2500 Å) estimated by Aeppli *et al.*¹ from a muon-spin-relaxation measurement of a LSCO polycrystalline sample with a Sr content of $x = 0.075$. Also, our derived values of $H_{c2}(0) = (33 \pm 3)$ T, $-dH_{c2}/dT = 1.35 \pm 0.06$ T/K at T_c , and $\xi_{ab}(0) = (32 \pm 2)$ Å for the same crystal are in rather good agreement with the corresponding values for a polycrystalline sample with Sr content $x = 0.075$ [$H_{c2}(0) \approx 36$ T, $-dH_{c2}/dT \approx 1.51$ T/K near T_c , and $\xi_{ab}(0) \approx 34 \sim 51$ Å], which are deduced from a transport measurement by Kwok *et al.*⁵ The apparent agreement between the present results and some earlier ones is surprising considering the fact that the earlier studies had employed polycrystalline specimens. Particularly for the transport measurements, there are a number of difficulties in deducing the superconducting parameters from polycrystalline specimens, such as the weak intergranular coupling and the rather wide temperature range of the reversible vortex motion which lead to the broadening of the resistive transition in high magnetic fields. Thus, the apparent good agreement is likely to be accidental due to some compensating factors in the earlier measurements.

The traditional extrapolation method using the linear Abrikosov regime of magnetization near H_{c2} was generally found inapplicable in obtaining superconducting parameters, such as $H_{c2}(T)$ and κ , of the LSCO system, because of the severe deviation of the measured magnetization data from the linear Abrikosov-like behavior. As we

mentioned in the Introduction, the application of the linearized Abrikosov Eq. (1) is limited to the field region near H_{c2} ($0.4H_{c2} \leq H \leq H_{c2}$), or in the equivalent temperature region of $T_A(H) < T < T_c(H)$ at fixed field H , where $H \approx 0.4H_{c2}(T_A)$. For crystal D , as an example, this temperature region is found to be from $T_A = 27.96$ K to $T_c = 29.03$ K at 10 000 Oe, where we have used both the Hao-Clem-model-derived values of $-dH_{c2}/dT \approx 1.5$ T/K near T_c and the mean-field $T_c(0) = 29.8$ K, which coincides with the measured T_c at onset. Similarly, such a temperature region at 30 000 Oe is from 24.22 to 27.60 K, while at 50 000 Oe it is from 20.08 to 26.13 K. As illustrated in Fig. 3, these temperature regions for applied fields between 10 000 and 50 000 Oe are marked by the shaded area. Also shown in the figure are the straight lines representing the traditional extrapolation method used to determine the values of $H_{c2}(T)$. However, it is clear that the magnetization data taken at fields up to 40 000 Oe within the so-called linear Abrikosov regions (shaded area) do not display the linear temperature dependence because of the severe broadening due to the large fluctuation effects involved. Although a set of straight lines can be drawn for $4\pi M(T)$ (see Fig. 3), all of the lines except for 50 000 Oe are actually drawn outside the temperature range of the applicability of the linear Abrikosov region. In addition, it should be noted that the specimen D , which was used for this illustration, is the most three-dimensional-like superconductor among the LSCO specimens. Thus, the linear extrapolation method would be even less appropriate for other crystals which we studied.

In order to further illustrate this point, in Fig. 4, we plotted the values of $H_{c2}(T)$ for crystal D determined both by the Hao-Clem model (open circles) and by the traditional extrapolation method (solid squares) illustrated in Fig. 3 with the use of Eq. (1). Two lines, also shown in Fig. 4, are the BCS and the empirical temperature fits to the experimental values of $H_{c2}(T)$, obtained by the Hao-Clem model, at temperatures between 24.0 and 25.4

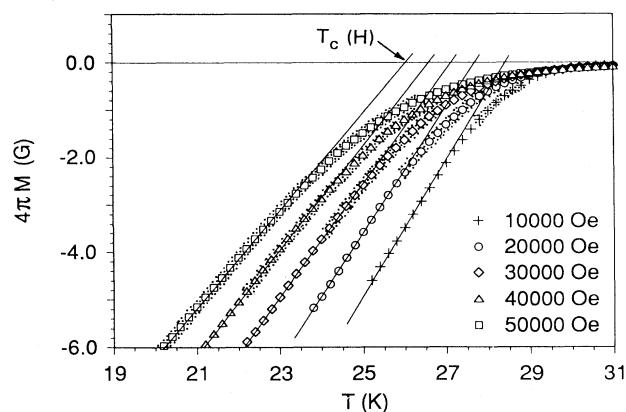


FIG. 3. Magnetization data of crystal D taken at various fields plotted against temperature, where the shaded areas represent the linear Abrikosov region, and the straight lines show the traditional extrapolation method used for the determination of $H_{c2}(T)$, based on Eq. (1).

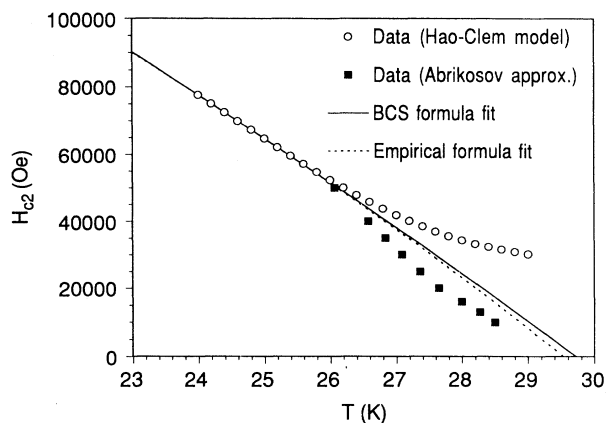


FIG. 4. Comparison of experimentally determined values of $H_{c2}(T)$ for crystal D from the Hao-Clem model (open circles) and from the extrapolation method based on the linear Abrikosov formula. The lines are the BCS and the empirical formula fits to the data obtained from the Hao-Clem model.

K, within which the value of κ for crystal D is found to be nearly constant. The upturn behavior of the data $H_{c2}(T)$ determined by the Hao-Clem model near T_c is due to the fluctuation-induced diamagnetic moment near T_c , as discussed above for crystal C . As for the $T_c(H)$, or $H_{c2}(T)$ data obtained by the linear extrapolation method, we can see that it generally underestimates the value of $H_{c2}(T)$ for crystal D in the field range of our experiments except at 50 000 Oe. This is simply because these extrapolations are primarily based on data actually taken outside the linear Abrikosov temperature regions as illustrated in Fig. 3. The most important issue raised from this comparison of the extrapolation method with our new data analysis technique is that two requirements must be satisfied in order to use the extrapolation method. First, the magnetization data taken at one particular field should be in the valid linear Abrikosov temperature region at that field. Additionally, the magnetization data must display a reasonably linear temperature dependence within the Abrikosov region. In the case of crystal D , only the data taken in the temperature region between 20.0 and 23.0 K at 50 000 Oe satisfies the above requirements. Thus, it is not surprising to see that the value of $H_{c2}(T)$ obtained at 50 000 Oe by the extrapolation method is very close to that determined by our data analysis technique. Also, the value of κ which is obtained by taking the slope of the extrapolation line of $4\pi M(T)$ at 50 000 Oe, and calculated via Eq. (1) is about 70; and it is quite close to the value derived from the Hao-Clem model ($\kappa = 75$) as expected from the above discussion.

In the case of the other three crystals, we have found that the magnetization data taken at all fields near T_c did not satisfy these two requirements. This is due to the fact that the magnetization data taken in the linear Abrikosov region are significantly affected by large fluctuations such that it does not exhibit a linear temperature dependence. Thus, the traditional extrapolation method is not applicable at all for the other three compositions of LSCO.

Furthermore, it was observed that this fluctuation effect on the magnetization increases as the Sr content decreases, presumably due to the progressive weakening of the coupling between the adjacent superconducting CuO planes, i.e., greater anisotropy at low Sr contents which enhance fluctuation effects at high fields.¹⁸

In Fig. 5, the data of $H_c(0)$ and $T_c(0)$ for the superconducting LSCO family are shown as a function of Sr content. At Sr content $x = 0.077$, both $H_c(0)$ and T_c reach the maximum value, while the value of $\lambda_{ab}(0)$ is the lowest (see Table I). We recall that $\lambda(0)$ in the simplest form is proportional to (m^*/n_s) , where m^* is the effective mass of the charge carrier, and n_s is the density of the superconducting carriers. Then, although the data is limited, it suggests that the number of holes contributing to superconductivity in this system decreases beyond the Sr content $x \sim 0.075$ for the maximum in the T_c , if m^* is independent of the Sr content. This is in contrast to the results of the measurements of the hole density in the normal state as a function of Sr, using the techniques such as the Hall coefficient,²³ the electronic-spin susceptibility,^{24,25} and the x-ray absorption spectra at the oxygen K edge.^{26,27} All of these measurements indicated that the carrier density continues to increase to a Sr content well beyond that required for the maximum in T_c . However, the detailed measurements of the polarization dependence of the oxygen K -edge absorption spectra in the LSCO system²⁸ showed a significant increase of the absorption intensity ratio of apical O $2p_z$ to in-plane O $2p_{x,y}$ for a Sr content $x > 0.075$. This suggests that the hole density in the ab plane may actually decrease beyond the Sr content for the T_c maximum. Our observation, although based on a limited compositional range, is consistent with such a conclusion. However, the assumption of the constancy of m^* with the Sr content must be examined further before the above conclusion is finalized.

In Fig. 5, it was also noted that the value of $H_c(0)$ for crystal B is anomalous and equal to that for crystal A . It is possible that a relatively large transition width for crystal B ($\Delta T_c = 3.7$ K), as opposed to $\Delta T_c = 0.9$ K for crystal A , may indicate some inhomogeneity inside this particular crystal. This could result in reduced total diamagnetization from some weak (or non-) superconducting regions at high fields so as to give a lower experimental value of $H_c(T)$ which was computed for the entire crystal. It should also be noted that at this composition a well-known anomalous dip or plateau in T_c has been documented.^{23,29,30} Although the physical cause, such as a structural phase transition, has not been determined, this observed anomaly in $H_c(0)$, as well as in other parameters, is likely to be caused by the same phenomenon as that responsible for the anomaly in T_c . Moreover, this present measurement confirms that this anomaly is a bulk effect rather than some unknown surface phenomena.

Finally, the Ginzburg-Landau parameter κ for LSCO family along the c axis is around 75 for the composition near the peak value $x = 0.075$, which falls between the κ value of $\text{YBa}_2\text{Cu}_3\text{O}_7$ ($\kappa = 57$, determined by Hao *et al.*⁹) and that of Bi-based superconductors (for instance, $\kappa = 170$ for $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$).¹² This suggests that the

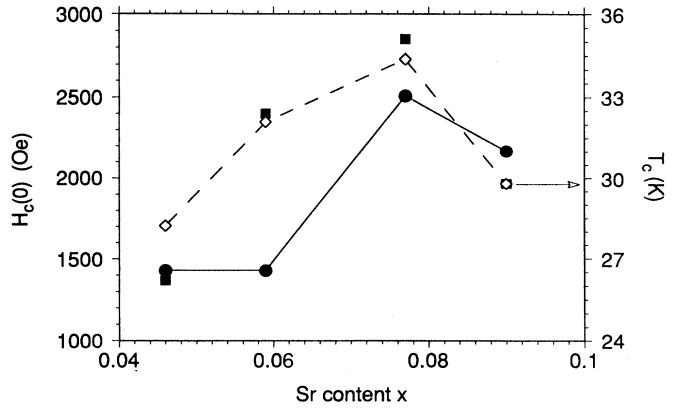


FIG. 5. Experimental data of $H_c(0)$ (represented by solid circles) and T_c for superconducting LSCO system are plotted as a function of Sr content x . The open diamonds represent the results of derived mean-field $T_c(0)$ from a BCS fit to the experimental data of $H_c(T)$, while the solid squares show the values T_c at onset determined from low-field shielding measurements.

strength of superconducting coupling of the adjacent CuO planes in the LSCO system is between that of $\text{YBa}_2\text{Cu}_3\text{O}_7$, which is a three-dimensional system in nature,³¹ and that of the Bi-based family (a quasi-two-dimensional system¹⁷). A thorough study on the dimensionality related properties of the LSCO system is currently underway.

IV. CONCLUSION

Superconducting parameters of the $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_4$ system, over the composition range of $x = 0.05$ – 0.10 , have been determined by a data analysis technique based on the Hao-Clem variational model. We have shown that the large fluctuation effects near T_c significantly distort the Abrikosov-like behavior of the magnetization near $H_{c2}(T)$ such that the traditional extrapolation method based on the linear Abrikosov formula is practically inapplicable for this system except at very high fields ($\geq 50\,000$ Oe) for a high-Sr-content specimen. Also, in a special case where the linear Abrikosov formula is applicable, we found that the Hao-Clem model gives essentially the same result as that based on the traditional extrapolation method in the determination of some of superconducting parameters of LSCO system.

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- ¹G. Aeppli, R. J. Cava, E. J. Ansaldo, J. H. Brewer, S. R. Kreitzman, G. M. Luke, D. R. Noakes, and R. F. Kiefl, *Phys. Rev. B* **35**, 7129 (1987).
- ²S. Uchida, H. Takagi, K. Kishio, K. Kitazama, K. Fueki, and S. Tanaka, *Jpn. J. Appl. Phys.* **26**, L443 (1987).
- ³M. Tarascon, L. H. Greene, W. R. McKinnon, G. W. Hull, and T. H. Geballe, *Science* **235**, 1373 (1987).
- ⁴T. P. Orlando, K. A. Delin, S. Foner, E. McNiff Jr., J. M. Tarascon, L. H. Greene, W. R. McKinnon, and G. W. Hull, *Phys. Rev. B* **35**, 5347 (1987).
- ⁵W. K. Kwok, G. W. Crabtree, D. G. Hinks, D. W. Capone, J. D. Jorgensen, and K. Zhang, *Phys. Rev. B* **35**, 5343 (1987).
- ⁶T. Kimura, K. Kishio, T. Kobayashi, Y. Nakayama, N. Motohira, K. Kitazawa, and K. Yamafuji, *Physica C* **192**, 247 (1992).
- ⁷I. Tanaka and H. Kijima, *Nature* **21**, 337 (1989).
- ⁸M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).
- ⁹Z. Hao, J. R. Clem, M. W. McElflesh, L. Civale, A. P. Malozemoff, and F. Holtzberg, *Phys. Rev. B* **43**, 2844 (1991).
- ¹⁰A. A. Abrikosov, *Zh. Eksp. Teor. Fiz.* **32**, 1442 (1957).
- ¹¹P. H. Kes, C. J. van der Beek, M. P. Maley, M. E. McHenry, D. A. Huse, M. J. V. Menken, and A. A. Menovsky, *Phys. Rev. Lett.* **67**, 2383 (1991).
- ¹²Q. Li, M. Suenaga, J. Gohng, D. K. Finnemore, T. Hikata, and K. Sato, *Phys. Rev. B* **46**, 3195 (1991).
- ¹³P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).
- ¹⁴V. G. Kogan, M. M. Fang, and S. Mitra, *Phys. Rev. B* **38**, 11 958 (1988); S. Mitra, J. H. Cho, W. C. Lee, D. C. Johnston, and V. G. Kogan, *ibid.* **40**, 2674 (1989); W. C. Lee and D. M. Ginsburg *ibid.* **44**, 2815 (1991).
- ¹⁵Z. Hao and J. R. Clem, *Phys. Rev. Lett.* **67**, 2371 (1991).
- ¹⁶Q. Li, M. Suenaga, T. Kimura, and K. Kishio, *Phys. Rev. B* **47**, 2854 (1993).
- ¹⁷Q. Li, M. Suenaga, T. Hikata, and K. Sato, *Phys. Rev. B* **46**, 5857 (1992).
- ¹⁸Z. Tesanovic, L. Xing, L. Bulaevskii, Q. Li, and M. Suenaga, *Phys. Rev. Lett.* **69**, 3563 (1992).
- ¹⁹L. Bulaevskii (private communication).
- ²⁰A. L. Fetter and P. C. Hohenberg, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), p. 817.
- ²¹J. R. Clem, *Ann. Phys. (N.Y.)* **40**, 268 (1966).
- ²²N. R. Werthamer, E. Helfand, and P. C. Hohenberg, *Phys. Rev.* **147**, 295 (1966).
- ²³H. Takagi, T. Ido, S. Ishibashi, M. Uota, and S. Uchida, Y. Tokura, *Phys. Rev. B* **40**, 2254 (1989); T. Nagano, Y. Nakayama, T. Tomioka, K. Kishio, and K. Kitazawa (unpublished).
- ²⁴M. Oda, T. Ohguro, N. Yamada, and M. Ido, *J. Phys. Soc. Jpn.* **58**, 1137 (1989).
- ²⁵J. B. Torrance *et al.*, *Phys. Rev. B* **40**, 8872 (1989).
- ²⁶C. T. Chen *et al.*, *Phys. Rev. Lett.* **66**, 104 (1991).
- ²⁷D. A. Fischer, A. R. Moodenbaugh, Y. Xu, K. W. Dennis, and R. W. McCallum (unpublished).
- ²⁸C. T. Chen, L. H. Tjeng, J. Kwo, H. L. Kao, P. Rudolf, F. Sette, and R. M. Flemming, *Phys. Rev. Lett.* **68**, 2543 (1992).
- ²⁹M. K. Crawford, W. E. Farneth, E. M. McCarron III, R. L. Harlow, and A. H. Moudden, *Science* **250**, 1390 (1990).
- ³⁰M. Oda, T. Nakano, N. Yamada, and M. Ido, *Physica C* **183**, 234 (1991).
- ³¹U. Welp, S. Fleshler, W. K. Kwok, R. A. Klemm, V. M. Vinokur, J. Downey, B. Veal, and G. W. Crabtree, *Phys. Rev. Lett.* **67**, 3180 (1991).