*c***-axis negative magnetoresistance and fluctuation conductivity of** $Bi_2Sr_2CaCu_2O_x$ **single crystals**

Katsufumi Hashimoto,* Koichi Nakao, Hiroyuki Kado, and Naoki Koshizuka

Superconducting Research Laboratory, International Superconductivity Technology Center,

10-13 Shinonome 1-chome, Koto-ku, Tokyo 135, Japan

 $(Received 15 March 1995)$

We analyze the negative magnetoresistance observed in the *c*-axis transport of $Bi₂Sr₂CaCu₂O_x$ single crystals based on the density of states fluctuation conductivity by taking renormalization effects into account. It is shown that the calculated magnetic field dependence of the *c*-axis conductivity fits to the experimental results below as well as above critical temperature (T_c) reasonably well using a single set of parameters. This gives strong evidence to indicate important roles of fluctuation effects arising from the density of states term in high- T_c superconductors.

I. INTRODUCTION

Recently there has been a proposal that a semiconductorlike temperature dependence observed in the *c*-axis resistivity of $Bi₂Sr₂CaCu₂O_r$ (Bi 2212) superconductors above critical temperature (T_c) is due to a superconducting fluctuation effect. $1-3$ In this explanation the fluctuation conductivity arising from the density of states (DS) term, whose contribution is opposite to that of the conventional Aslamazov-Larkin (AL) term,⁴ is taken into account. It is shown that in highly anisotropic superconductors the coefficient of the AL conductivity along the *c* axis is reduced by the small anisotropic parameter compared to that of the DS conductivity, and effects of the latter contribution appear above T_c to produce a semiconductorlike temperature dependence in the *c*-axis resistivity. A similar explanation based on a Josephson junction model of the Bi system was also proposed by the other authors.5,6 In particular Ref. 5 gave a reasonable fit to experimental results on the semiconductorlike temperature dependence of the Bi system. However, an unusually large amplitude of fluctuation was required in the fit.

One of the most convincing ways to investigate the validity of this proposal is to measure the magnetic field dependence of the *c*-axis resistivity, because superconducting fluctuation effects are sensitively affected as in the case of varying temperatures. The negative contribution of the DS term to the conductivity is reduced by increasing magnetic fields, which might lead to a negative magnetoresistance. Such a feature is clearly seen in the calculated results by Dorin *et al.*,³ though these authors did not mention it explicitly.

We have previously measured the *c*-axis magnetoresistance of Bi 2212 single crystals with fields up to 40 T applied along the *c* axis, and in fact observed the negative magnetoresistance below as well as above T_c .⁷ We have also performed measurements with fields applied perpendicularly to the *c* axis, and found that the negative magnetoresistance almost vanishes in this case. 8 The fact that the phenomenon crucially depends on the orientation of applied magnetic fields indicates that the mechanism by spin scattering is excluded. In the present paper we analyze our experimental results based on the fluctuation conductivity due to the DS term. In order to account for a large broadening of the superconducting transition under magnetic fields we include renormalization effects in the fluctuation propagator. $9,10$ We show that the calculated magnetic field dependence of the *c*-axis conductivity fits to the experimental results below as well as above T_c reasonably well using a single set of parameters, namely, T_c/T_{c0} = 0.84 (T_c ₀ is the mean field critical temperature), the in-plane coherence length ξ_{ab} =16 Å, and the effective mass m_{ab} =3.6. The same set of parameters gives also good agreement between the AL conductivity and experimental results obtained in the low field region below *T_c*. These results provide the first strong evidence to indicate importance of fluctuation effects due to the DS term in understanding physical properties of high- T_c superconductors. On the other hand, the semiconductorlike temperature dependence above T_c observed in our sample with high c -axis resistivity is not explained by the DS term alone using the deduced parameters.

II. FIELD DEPENDENCE OF THE DENSITY OF STATES FLUCTUATION CONDUCTIVITY

The fluctuation conductivity due to the DS term was introduced by Ioffe *et al.*¹ in order to give an account for a semiconductorlike temperature dependence of the *c*-axis resistivity of high- T_c superconductors. The first order perturbation result was applied in Ref. 2 to analyze experimental results on the Bi system, and in Ref. 3, Dorin *et al.* extended the result for cases with arbitrary scattering time and with magnetic fields. Here we use the result of Ref. 1 that includes effects of higher order terms which is necessary in quantitative analysis. In order to treat field dependence below as well as above T_c , the result is extended to be applicable for cases with nonzero magnetic fields and to include renormalization effects on the mass term. We note that the theory neglects the vertex corrections due to the impurity scattering and applies to the clean case.

Following Ref. 1, the conductivity of normal electrons with fluctuation effects is given by

0163-1829/96/53(2)/892(4)/\$06.00 53 892 © 1996 The American Physical Society

$$
\sigma_c = \sigma_0 \times F[D(B, T), \lambda],
$$

\n
$$
F[D(B, T), \lambda] = \frac{\lambda}{8} \int_0^\infty dx [\cosh^{-2}(x/4)]
$$

\n
$$
\times \left(1 + \frac{\lambda^2 + x^2}{|D(B, T) + (\lambda + ix)^2|}\right)
$$

\n
$$
\times \frac{1}{\text{Re}\sqrt{D(B, T) + (\lambda + ix)^2}},
$$
 (1)

where σ_0 is the normal state conductivity without fluctuation effects, and $\lambda = \hbar / \tau k_B T$ (τ is the scattering time and *T* is the temperature). The parameter $D(B,T)$ represents strength of fluctuation under magnetic fields *B* which is given by

$$
D(B,T) = D_0 D_1,
$$

\n
$$
D_0 = 1/\pi N(0) \xi_{ab}^2 k_B T_c,
$$

\n
$$
D_1 = \beta \sum_{m=0}^{1/\beta} \left[(\varepsilon_R + m\beta)(\varepsilon_R + m\beta + r) \right]^{-1/2},
$$
 (2)

where $N(0) = m_{ab}/2\pi\hbar^2$ is the two-dimensional density of state for a single spin $(m_{ab}$ is the effective mass), β is equal to $2B/B_{c2}$ (B_{c2} is the mean field upper critical field), ε_R is the renormalized mass, and r is the anisotropic parameter defined later in Eq. (3) . We determine the renormalized mass using the Hartree approximation which provides results close to the mean field ones in the flux flow regime. $9,10$ The relevant Ginzburg-Landau (GL) Hamiltonian is given by

$$
H = \sum_{i} \int \int dx dy \Bigg[N(0) \Bigg\{ \ln \Big(\frac{T}{T_{c0}} \Big) |\Psi_{i}|^{2} + \xi_{ab}^{2} \Big| \Bigg(i \nabla + \frac{2\pi}{\Phi_{0}} A \Big) \Psi_{i} \Big|^{2} + \Big(\frac{r}{4} \Big) |\Psi_{i+1} - \Psi_{i}|^{2} \Bigg\} + \Big(\frac{b}{2} \Big) |\Psi_{i}|^{4} \Bigg],
$$

$$
b = \frac{7 \zeta(3) N(0)}{8 \pi^{2} (k_{B} T_{C})^{2}},
$$
 (3)

where $A = Bxe_y$, Φ_0 is the flux quantum, and $\zeta(x)$ is the Riemann's zeta function. Then the Hartree approximation leads to

$$
\varepsilon_R = \varepsilon_0 + \frac{7\,\zeta(3)}{32\,\pi^2} \, D(B,T),
$$

$$
\varepsilon_0 = \ln(T/T_{c0}) + B/B_{c2}.
$$
 (4)

In numerical calculations of D_1 in Eq. (4) the cutoff for summation by m is done by the following replacement:¹¹

$$
D_1 = [1/7\zeta(3)]\beta \sum_{m=0}^{\infty} [(\varepsilon_R + m\beta)(\varepsilon_R + m\beta + r)]^{-1/2} C_m(\beta),
$$

$$
C_m(\beta) = \sum_{n=0}^{\infty} \frac{1}{[0.5 + n + (2/\pi^2)\beta(m + 0.5)]^2(0.5 + n)}.
$$
 (5)

FIG. 1. Calculated magnetic field dependence of ΔD [Eq. (9)] that corresponds to the first order perturbation results of the conductivity is shown for several temperatures. The dashed and dotdashed lines represent the limiting results given by Eqs. (6) and (7) , respectively (the former result is independent of temperature).

The same procedure is also employed in the calculation of D_1 of Eq. (2) as an approximation.

The $D(B,T)$ contains ε_R so that Eq. (4) should be solved self-consistently. However, approximate solutions are obtained in limiting cases. One case is $\varepsilon_0 \le 0$, where ε_R is small and can be neglected compared to $|\varepsilon_0|$. In this case we have

$$
D(B,T) = [32\pi^2/7\zeta(3)]\left|\varepsilon_0\right|.\tag{6}
$$

In the other limit $\varepsilon_0 \ge 0$, the second term on the right-hand side of Eq. (4) is neglected compared to ε_0 , so that the Gaussian approximation holds as follows:

$$
\varepsilon_R = \varepsilon_0. \tag{7}
$$

The result of the intermediate case depends on the value of D_0 . From the relation⁹

$$
D_0 = \frac{64\pi^3 k_B T_c \mu_0 \xi_{ab}^2 \kappa^2}{7 \zeta(3) \Phi_{0}^2 s},
$$
\n(8)

where μ_0 is the permeability, *s* is the lattice spacing along the c axis, and κ is the GL parameter, and using typical values of T_c =80 K, ξ_{ab} =12 Å, s =15 Å, and κ =100, we obtain D_0 of about 0.7. We show in Fig. 1 the magnetic field dependence of $D(B,T)$ defined by

$$
\Delta D = |D(B,T) - D(0,T)|,\tag{9}
$$

for several values of T/T_{c0} from 0.7 to 1.3 in the case of *D*₀=0.7. In Eq. (9) *D*(0,*T*) for T/T_{c0} < 1 is determined by an extrapolation. Such a procedure is also employed for similar quantities defined later. It is seen that ΔD increases with decreasing temperatures. The curves for $T/T_{c0} > 1$ are concave, while those for T/T_{c0} =0.7 and 0.8 show linear dependences. The results of T/T_{c0} =0.7 and 1.3 are close to the limiting values given by Eqs. (6) and (7) , respectively, which are also shown in the figure. The curve of T/T_{c0} =0.9, on the other hand, is convex reflecting a saturation of the steep increase in the low field region. These features represent re-

FIG. 2. Calculated magnetic field dependence of ΔF [Eq. (10)] including higher order effects on the conductivity is shown for several temperatures. FIG. 3. Experimental results on the *c*-axis conductivity are

sults of the first order perturbation in the fluctuation parameter $D(B,T)$. Figure 2 shows the quantity

$$
\Delta F = F[D(B, T), \lambda] - F[D(0, T), \lambda], \quad (10)
$$

which includes higher order effects in $D(B,T)$. The ΔF depends on λ which is considered to be around 1.¹² Here we choose a rather small value of 0.8 from a fit to experimental results. Compared to the first order results, higher order effects make the magnetic field dependence weaker for lower temperatures. The difference between high and low temperatures becomes smaller and the magnitudes of T/T_{c0} =0.7 and 0.8 are reversed. On the other hand, the shapes of the curves are not changed except that a small tendency of concavity appears in the curve of T/T_{c0} =0.7. In the next section these theoretical results are compared with experiments.

III. COMPARISON WITH EXPERIMENTAL RESULTS

The Bi 2212 single crystal was prepared by the floating zone method.¹³ The T_c of the sample used for the measurements was 79 K and the size was 0.25 mm² in area and 17 μ m in thickness. The two terminal measurement was performed using top and bottom electrodes covering almost all area. The contact resistance of 1.8 Ω which is much smaller than that of the sample is subtracted from the data. The applied current density was 8.4 mA/cm². The pulsed magnetic fields are generated and applied along the *c* axis of the sample, the details of which are given in Ref. 7.

Figure 3 shows the conductivity $\sigma_{\exp}(B,T)$ as a function of magnetic fields obtained at several temperatures below as well as above T_c . Above T_c increase in the conductivity with increasing magnetic fields, namely negative magnetoresistance, is clearly seen from the zero field. Below T_c the conductivity first decreases sharply with increasing magnetic fields. This comes from the AL conductivity. In the higher field region, however, the conductivity gradually increases with increasing magnetic fields. Figure 4 shows the magnetic field dependence of the conductivity defined by

$$
\Delta \sigma_{\text{exp}} = \sigma_{\text{exp}}(B, T) - \sigma_{\text{exp}}(0, T). \tag{11}
$$

We see that $\Delta \sigma_{\text{exp}}$ increases with decreasing temperatures. It then takes a maximum between $T=77.8$ and 74.4 K, and

shown as a function of magnetic fields up to 30 T for several temperatures.

decreases gradually with lowering temperatures. The curves for high temperatures are concave, while those of low temperatures are linear. These features coincide with the calculated results shown in Fig. 2.

In order to fit the theoretical results, Eq. (10) , to the experiments by varying T_{c0} , D_0 , σ_0 , and B_{c2} (λ is fixed to 0.8), we first note that the experimental curve of $T=85.1$ K is relatively close to those of lower temperatures. This leads us to choose rather high T_{c0} above 85 K. We optimized D_0 by comparing calculated and experimental results by selecting several values of T_{c0} . The σ_0 is estimated from the relation $\sigma_{\exp}(0,T) > \sigma_0 F(0,T)$ which should hold at all temperature. The B_{c2} is then determined by comparing $\Delta \sigma_{exp}$ and $\sigma_0 \Delta F$ for $T=74.4$ K. The results thus obtained with $T_{c0}=94$ K $(T_c/T_{c0} = 0.84)$, $D_0 = 0.82$ ($m_{ab} = 3.6$), $\sigma_0 = 0.022/\Omega$ cm, and B_{c2} =130 T (ξ_{ab} =16 Å) are marked in Fig. 4. The σ_0 is less than 0.022/ Ω cm which gives B_{c2} less than 130 T. We see that the agreement below T_c is satisfactory. Above T_c , on the

FIG. 4. Experimental results on the magnetic field dependence of the conductivity $[Eq. (11)]$ are shown. The markers are theoretical results obtained from a fit by the DS conductivity $(+: 113.9)$ K; \bullet : 96.8 K; *: 89.8 K; \bowtie : 85.1 K; \odot : 82.6 K; \bullet : 77.8 K; **▲**: 74.4 K; \diamond : 71.3 K; \triangle : 68.4 K).

other hand, the fit in the low field region below 10 T is good, but in the higher field region the magnetic field dependence of the experimental results is stronger than that of the theoretical results. Choosing higher T_{c0} and/or smaller λ reduces this discrepancy only slightly. We have not yet identified the reason of the discrepancy, but one possibility may be due to an approximation of neglecting fluctuation propagator with nonzero frequencies in the calculation of $D(B,T)$. Those corrections are more important for cases with higher temperatures and higher magnetic fields. Nevertheless overall agreement between theoretical and experimental results are reasonably well, and this provides a strong evidence to indicate importance of fluctuation effects due to the DS term in understanding physical properties of high- T_c superconductors.

In order to check whether the deduced parameters, T_{c0} , D_0 , and B_{c2} , are reasonable or not, we calculate the AL conductivity below T_c under magnetic fields according $to^{3,9,10}$

$$
\sigma_{\rm AL} = \frac{e^2}{128\hbar} \frac{s}{\xi_{ab}^2} r^2 \beta \sum_{0}^{\infty} \left[(\varepsilon_R + m\beta)(\varepsilon_R + m\beta + r) \right]^{-3/2}.
$$
\n(12)

The experimental normal state conductivity, $\sigma_{N \exp}$, is determined from $\sigma_{\text{exp}}(0,T)$ by approximating the resistivity, $1/\sigma_{\text{exp}}(0,T)$, to the second order polynomial of temperature. The resistivity in the low field region is then given by

$$
R = 1/(\sigma_{\text{AL}} + \sigma_{N \text{ exp}}). \tag{13}
$$

Figure 5 compares the experimental results and calculated results obtained with T_{c0} =94 K, D_0 =0.82, B_{c2} =130 T, and $r=1.2\times10^{-5}$. The agreement is excellent, indicating validity of the deduced parameters.

We note that estimated σ_0 is below 0.022/ Ω cm which corresponds to the experimental conductivity only at 85 K in Fig. 3. Thus the semiconductorlike temperature dependence above 85 K is not explained by the fluctuation effect due to the DS term alone. This suggests that there is another conduction mechanism along the *c* axis in the present sample with a high resistivity which is of thermally activated type and insensitive to magnetic fields.

- *Present address: Research and Development Group, Oki Electric Industry Co., Ltd., 550-5, Higashiasakawa-cho, Hachioji-shi, Tokyo 193, Japan.
- ¹L. B. Ioffe, A. I. Larkin, V. V. Varlamov, and L. Yu, Phys. Rev. B 47, 8936 (1993).
- 2 G. Balestrino, M. Marinelli, E. Milani, V. V. Varlamov, and L. Yu, Phys. Rev. B 47, 6037 (1993).
- $3V$. V. Dorin, R. A. Klemm, V. V. Varlamov, A. I. Buzdin, and D. V. Livanov, Phys. Rev. B 48, 12 951 (1993).
- 4 L. G. Aslamazov and A. I. Larkin, Phys. Lett. **26A**, 238 (1968).
- 5 K. E. Grey and D. H. Kim, Phys. Rev. Lett. **70**, 1693 (1993).
- 6 A. F. Volkov, Solid State Commun. 88, 715 (1993).
- 7 K. Nakao, K. Hashimoto, K. Katamuku, N. Koshizuka, and S. Tanaka, in *Advances in Superconductivity IV* (Springer-Verlag,

FIG. 5. Experimental results on the resistivity obtained in the low field region below T_c and a theoretical fit (markers) by the AL conductivity using Eq. (13) are shown.

IV. CONCLUSION

We analyzed the *c*-axis negative magnetoresistance of Bi 2212 single crystals based on the fluctuation conductivity arising from the DS term. We found reasonable agreement between theoretical and experimental results on the magnetic field dependence below as well as above T_c using a single set of parameters. These parameters are shown to give also good agreement between the AL conductivity and experimental results obtained in the low field region below T_c . These results provide the first strong evidence to indicate importance of fluctuation effects due to the DS term in understanding physical properties of high- T_c superconductors. There is, however, some discrepancy between theoretical results and experiments in the high temperature and high field region. The reason of this discrepancy should be clarified in future.

ACKNOWLEDGMENT

This work was partially supported by NEDO for the R&D of Industrial Science and Technology Frontier Program.

Tokyo, 1994), p. 205; K. Nakao, K. Katamuku, K. Hashimoto, N. Koshizuka, and S. Tanaka, Physica B 201, 262 (1994).

- 8H. Kado, K. Nakao, K. Hashimoto, T. Egi, and N. Koshizuka, Advances in Superconductivity VII (Springer-Verlag, Tokyo, 1995), p. 85.
- 9R. Ikeda, T. Ohmi, and T. Tsuneto, J. Phys. Soc. Jpn. **60**, 1051 $(1991).$
- 10 S. Ullah and A. T. Dorsey, Phys. Rev. B 44, 262 (1991).
- 11 J. P. Hurault and K. Maki, Phys. Rev. B 2, 2560 (1970).
- 12For example, Y. Matsuda, T. Hirai, S. Komiyama, T. Terashima, Y. Bando, K. Iijima, K. Yamamoto, and K. Hirata, Phys. Rev. B 40, 5176 (1989).
- ¹³K. Takamuku, K. Ikeda, T. Takata, T. Miyatake, I. Tomeno, S. Gotoh, N. Koshizuka, and S. Tanaka, Physica C **185-189**, 451 $(1991).$