Vanishingly small Maki-Thompson superconducting fluctuation in the magnetoresistance of high- T_c superconductors

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The magnetoresistance (MR) of high- T_c cuprate superconductors (HTSC's) is studied. We have shown that the Zeeman term of the Maki-Thompson (MT) process could not be detected in the longitudinal MR of 90 K YBCO crystals. We show here that the MT process is thought to be also negligible in the transverse MR of 90 K and Zn-doped YBCO crystals. The MR, which has been assigned previously to a MT-orbital process, is more naturally understood as normal-state orbital MR that is proportional to the square of the Hall angle. We conclude that the MT-fluctuation is vanishingly small, at least above T_c , in these HTSC's. In addition to the Hall measurements, MR measurements also support the hypothesis of two distinct relaxation times in HTSC's. [S0163-1829(97)03817-4]

Normal-state magnetoresistance (MR) of a superconductor has two different origins. One is caused from suppression of superconducting fluctuation, by the applied magnetic field, and the other is the normal-state MR (in the sense of the Boltzmann transport theory).

Let us consider the fluctuational MR first. Just above the transition temperature, resonant scattering occurs between electrons of opposite spin and momentum over the Fermi surface, leading to the Cooper instability. Normal electrons begin to form bound pairs of a finite lifetime. The virtual pair state causes excess conductivity, this is the Aslamazov-Larkin (AL)-type fluctuational conductivity. The pair decays into a long-wavelength diffusive mode of the normal electrons with small total momentum. This mode also decays into single quasiparticles by phase-breaking scattering, or back into the virtual pair state again. This diffusive mode causes excess conductivity, this is the Maki-Thompson (MT)-type fluctuational conductivity.^{4–6} The AL-fluctuation arises from a "superfluid" of the virtual resonant pair state, whereas the MT fluctuation arises from a "normal fluid" of the diffusive mode. The fluctuational MR comes from suppression of AL and MT fluctuation under a magnetic field. Near the transition, MR from the AL fluctuation dominates. As temperature increases, a crossover from AL to MT fluctuation takes place.⁷ The first theory of the fluctuational MR that could be applied to high- T_c cuprate superconductors (HTSC's) was proposed by Aronov, Hikami, and Larkin (AHL).⁸ In the theory the Zeeman effect on paraconductivity was considered and it has been successfully observed in the HTSC.^{9,1} It has also demonstrated the singlet nature of the Cooper pair in HTSC's. In the conventional superconductors, the depairing by spin-splitting Zeeman energy is negligible compared to the orbital depairing effect i.e., AL-orbital (ALO) and MT-orbital (MTO) terms. In the HTSC's, however, the Zeeman effect is indispensable because of the short coherence length. Moreover, in the longitudinal MR, where $B \| I \perp c$, the Zeeman terms, AL-Zeeman (ALZ) and MT-Zeeman (MTZ), are dominant.

On the other hand, in the metallic state, the ordinal orbital magnetoresistance, in the Boltzmann sense, generally exists.

It comes from the bending effect of an electron trajectory by the applied magnetic field in the mean free time between scatterings. Hereafter, we call this MR the normal-state orbital MR (NOMR).

Treatment of the MT fluctuation in HTSC's is still controversial. Rice *et al.*¹⁰ reported that the MT process is surprisingly dominant in the fluctuation Hall conductivity of untwinned YBCO crystals. According to Rice, the MT process is larger than the AL process from T_c to 180 K. On the other hand, we have reported¹ that the MTZ contribution could not be detected in the longitudinal MR of 90-K YBCO twinned crystals. Sekirnjak *et al.*¹¹ proposed that the failure of detecting the MTZ term implies that the MTO contribution, too, is absent or at least rather small. Recently, Lang *et al.*¹² reported that the MT fluctuation is vanishingly small from T_c to 130 K from the measurement of fluctuation Hall conductivity in (Bi,Pd)₂Sr₂Ca₂Cu₃O_x.

Recently, MR of HTSC's have been reconsidered including the normal-state orbital MR, for $YBa_2Cu_3O_y$,^{3,11} $La_{2-x}Sr_xCuO_4$,^{3,13} $Bi_{2.1}Sr_{1.9}Ca_{1.0}Cu_2O_{8+\delta}$,¹⁴ and thin film $Bi_2Sr_2Ca_2Cu_3O_x$.¹² In particular, Harris *et al.*³ pointed out that the MR which had been assigned to the MTO process, may be the normal-state orbital MR (NOMR).

For a fourfold two-dimensional Fermi surface (FS), if there is no contribution from the fluctuational MR, the orbital MR ($\Delta \rho / \rho$) can be written

$$\frac{\Delta\rho}{\rho} = \langle \theta(s)^2 \rangle - \langle \theta(s) \rangle^2, \tag{1}$$

where $\langle \cdots \rangle$ means the average over the FS, and $\theta(s)$ is the local Hall angle on the arc length ds along the FS. From the observed Hall angle $(\theta_H = \langle \theta(s) \rangle)$ and MR $(\Delta \rho / \rho)$, they showed that for 90-K and 60-K YBCO, all three terms in Eq. (1) share the same temperature dependence, i.e., T^{-4} . Kohler's rule $[\Delta \rho / \rho = F(H/\rho) \propto T^{-2}]$ (Ref. 15) is strongly violated, which means breakdown of a single-relaxation-time approximation. This indicates that $\theta(s)$ changes uniformly with *T* around the FS and excludes the proposed models of the Hall angles which assumes the FS effect on the relaxation time.^{16,17}

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FIG. 1. MR data of a YBa₂Cu₃O_{7- δ} crystal in the transverse configuration. Note that the data are plotted against B^2 . At each temperature, ten data points were taken, five while increasing the magnetic field and the other while decreasing. We subtracted the small systematic drift $\Delta \rho / \rho \sim 0.01\%$ supposing that amount of drift is proportional to the time interval. We obtained representative MR at 1 T as a slope of the least-squares fit, shown as solid lines.

Here, however, a natural question arises. Does the observed MR mainly come from the normal-state MR? Is the paraconductivity contribution really negligible? Therefore, we measured the effect of Zn doping on the MR, because it affects the MT magnetoconductivity and the normal-state MR in different ways. We can determine which is dominant in the observed MR.

The crystals used in this study are the same, 90-K YBCO in Ref. 1 and Zn-doped YBCO in Ref. 18, that we used in the earlier works. Samples were grown by the Ba-Cu rich flux method¹⁹ and annealed at 500°C for 100 h in ambient pressure oxygen. For each crystal, Zn concentration was determined by the electron-probe microanalysis after the magnetoresistance measurement. Contamination by the crucible material (Pt) is found to be less than $2 \sim 6 \times 10^{-3}$ per Cu.

Data were taken by a commercial 12-T superconducting magnet with the He flow controlled by a needle valve. Two PID-controlled heaters are used to stabilize sample temperature. One is placed just behind a rotatable copper block, the sample stage, and is controlled by a carbon-glass-resister (CGR) sensor (H=0) and a capacitance sensor ($H\neq 0$) embedded in the sample stage. Another one, is placed at the shield block which surrounds the sample stage and acts as a heat bath. For every destination temperature, we first used a CGR sensor to stabilize sample temperature within 10 mK. Then, before applying a magnetic field we switched control from the CGR sensor to the capacitance sensor (Lake Shore CS-401). After the sample temperature was well stabilized under capacitance sensor control, the magnetic field was applied. Figure 1 shows the raw MR data of a YBa₂Cu₃O_{7- δ} crystal in the transverse configuration. As shown in Fig. 1, for all samples in the longitudinal and the transverse configuration we measured MR in a weak-field regime where $\Delta \rho / \rho \propto B^2$ holds. Here, $\Delta \rho = \rho(B) - \rho(0)$. We obtained the normalized MR at 1 T, as a slope of the leastsquares fit. In the following discussions, we use this normalized MR as the representative MR. Magnetoconductance $-\Delta\sigma$ was calculated as a change in the inverse resistivity $\Delta \sigma = \rho^{-1}(B) - \rho^{-1}(0)$. For every crystal, we have compared the following two models.



FIG. 2. Magnetoconductance $-\Delta\sigma = -[\rho^{-1}(B) - \rho^{-1}(0)]/B^2$ of a fully oxygenated YBa₂Cu₃O_{7- δ} crystal is plotted against reduced temperature $\epsilon = \ln(T/T_c)$ in a log-log scale. The *C* factor is chosen to be 1.05. (a) shows the optimal fit by the model (i), with $T_c = 92.5$ K, $\xi_{ab} = 14.0$ Å, $\xi_c = 4.6$ Å, $\hbar \tau_{\phi}^{-1}(k_BT)^{-1} = 2.0$, and a mean free path: $\ell'(T_c) = 117$ Å (Ref. 22). (b) shows the optimal fit by the model (ii), with $T_c = 92.5$ K, $\xi_{ab} = 13.2$ Å, $\xi_c = 2.9$ Å, b/m = 0.0217, and c = 0. NOMR of the form $\Delta\rho/\rho B^2$ $= [m/(bT^2 + c)]^2$ (Ref. 3) is used.

Model (i): The observed MR is the fluctuational MR of the AL and MT types. Longitudinal MR is fitted by contributions from ALZ and MTZ. Transverse MR is fitted by contributions from ALO, ALZ, MTO, and MTZ.

Model (ii): The observed MR is composed of AL-type fluctuational MR and the normal-state orbital MR (NOMR) which is proportional to the squared Hall angle. Longitudinal MR is fitted only by the ALZ contribution. Transverse MR is fitted by contributions from ALO, ALZ, and NOMR.

We use the Thompson-corrected AHL theory²⁰ of fluctuational MR in the clean limit as pointed out by Bieri and Maki,²¹ and also by Ref. 1. Here, we mention the *C* factor, an adjustable parameter which comes from sample inhomogeneity. We estimated it from the observed $d\rho/dT$ of the sample. We take $d\rho_a/dT$ of the untwinned sample *A* reported in Ref. 10 as the ideal $d\rho/dT$ of the CuO₂ plane. Since our samples are densely twinned in a submicron scale, CuO-chain conductivity is thought to be negligible. We used $(d\rho_{ab}/dT)_{\text{sample}}/(d\rho_a/dT)_{\text{untwin}}$ as the standard *C* factor of the sample. Since Zn doping makes the resistivity curve shift parallel upward with its slope unchanged, we also use the same method to estimate *C* factors of Zn-doped samples.

In Figs. 2(a) and 2(b), the MR of a $YBa_2Cu_3O_{7-\delta}$ twinned crystal is shown with theoretical fits by model (i)



FIG. 3. Magnetoconductance $-\Delta \sigma = -[\rho^{-1}(B) - \rho^{-1}(0)]/B^2$ of a fully oxygenated YBa₂(Cu_{0.972}Zn_{0.028})₃O₇₋₈ crystal is plotted against reduced temperature $\epsilon = \ln(T/T_c)$ in a log-log scale. The *C* factor is chosen to be 1.5. (a) shows the optimal fit by the model (i), with $T_c = 57.8$ K, $\xi_{ab} = 17.0$ Å, $\xi_c = 12.7$ Å, $\hbar \tau_{\phi}^{-1}(k_B T)^{-1} = 0.93$, and a mean free path: $\ell(T_c) = 50$ Å (Ref. 22). (b) shows the optimal fit by the model (ii), with $T_c = 57.8$ K, $\xi_{ab} = 18.4$ Å, $\xi_c = 7.0$ Å, b/m = 0.023, and c/m = 384. The NOMR of the form $\Delta \rho/\rho B^2 = [m/(bT^2 + c)]^2$ (Ref. 3) is used.

and (ii), respectively. The *C* factor is chosen to be 1.05. If we take model (i), NOMR and Anderson localization are assumed to be negligible. Here we tried a full fit with the four components: ALO, ALZ, MTO, and MTZ. As we have mentioned earlier,¹ if we include the MTZ term, we could not obtain reasonable parameters. In particular, a large out-of-plane coherence length, ξ_c , and a large pair breaking parameter, $\hbar \tau_{\phi}^{-1} (k_B T)^{-1}$, generally tend to be obtained.

Obtained parameters are $T_c = 92.5$ K, $\xi_{ab} = 14.0 \pm 0.3$ Å, $\xi_c = 4.6 \pm 0.3$ Å, $\hbar \tau_{\phi}^{-1}/k_B T = 2.0 \pm 0.2$, and a mean free path $\ell(Tc) = 117$ Å.²² The obtained $\xi_c \approx 4.6$ Å is inconsistently large compared with other measurements.^{23,24,10} The obtained pair breaking $\hbar \tau_{\phi}^{-1} \approx 2k_B T$ exceeds the thermal energy, and is also rather difficult to consider. If we take model (ii), the MR is assumed to be a sum of ALO, ALZ, and NOMR of $\Delta \rho / \rho B^2 = CT^{-4}$ form. This is a special form of $\Delta \rho / \rho B^2 = [m/(bT^2 + c)]^2$ with c = 0. Contributions from the MT process and Anderson localization are both assumed to be negligible. In Fig. 2(b), an optimal fit with T_c = 92.5 K, $\xi_{ab} = 13.2 \pm 0.3$ Å, $\xi_c = 2.9 \pm 0.1$ Å, and $C = (m/b)^2 = 2128$ K⁴ is shown.

In Figs. 3(a) and 3(b), the MR of a 2.8% Zn-doped YBCO crystal is shown with the theoretical fits by models (i) and (ii), respectively. The C factor is chosen to be 1.5. If we take

model (i), the NOMR and Anderson localization are both assumed to be negligible. Here, we also tried a full fit with the four components: ALO, ALZ, MTO, and MTZ. We also obtain a large ξ_c . Obtained parameters are $T_c = 57.8$ K, $\xi_{ab} = 17.0 \pm 1.5$ Å, $\xi_c = 12.7 \pm 0.3$ Å, $\hbar \tau_{\phi}^{-1} / k_B T = 0.9 \pm 0.2$, and a mean free path: $\ell(T_c) = 50$ Å.²² Below 120 K, the mean free path is estimated to be longer than 40 Å and is still larger than the obtained in-plane coherence length ξ_{ab} (=17 Å), so the clean limit analysis is appropriate. However, the obtained $\xi_c = 12.7$ Å is comparable to the obtained in-plane coherence length $\xi_{ab} = 17.0$ Å, and these values are clearly out of the limit of the layered-structure model. It is inconsistent with other experiments. If we take model (ii), the MR is assumed to be the sum of ALO, ALZ, and NOMR of the form $\Delta \rho / \rho B^2 = [m/(bT^2 + c)]^2$.³ The contributions from the MT process and Anderson localization are both assumed to be negligible. In Fig. 3(b), the optimal fit with $T_c = 57.8$ K, $\xi_{ab} = 18.4 \pm 0.3$ Å, $\xi_c = 7.0 \pm 0.1$ Å, $b/m = 0.023 \pm 0.002$, and $c/m = 384 \pm 26$ is shown.

From the orbital MR of the 0.7% Zn-doped YBCO crystal,¹⁸ we obtained an optimal fit with model (ii) with a *C* factor of 1.3, $T_c = 84.0$ K, $\xi_{ab} = 14.5$ Å, $\xi_c = 3.0$ Å, b/m = 0.022, and c/m = 100.

From the above analyses, rather different coherence lengths were obtained compared with our previous analysis of resistive transitions.¹⁸ In these, $\rho_0(T)$, the resitivity without superconducting fluctuation, was estimated as a linear extrapolation of resistivity over the higher temperature. A C factor was treated as a fitting parameter. On the other hand, here, the C factor is determined by the observed $d\rho/dT$ of the sample. Though the obtained coherence-length values are different, the tendency, reduction of anisotropy by Zn doping, does not change. Recently, Panagopoulos et $al.^2$ reported a systematic decrease in the anisotropy ratio $\gamma = \lambda_c(0) / \lambda_{ab}(0)$ with Zn doping from ac-susceptibility measurements of the grain aligned Zn-doped-YBCO powder, which is consistent with our results, whereas Axnäs et al.²⁶ reported an increase of the anisotropy ratio ξ_{ab}/ξ_c with Zn doping. We do not understand this discrepancy.

From the fit by model (ii), we can only determine the ratio b/m and c/m in the NOMR formula. In order to determine *m* and *c*, we use b=0.041,²⁷ then we obtain m=1.9, c=0



FIG. 4. The impurity contribution C(x) in NOMR is plotted against Zn concentration x. The solid line is a guide to the eyes. The C(x) obtained from the MR fit (filled circles) coincides with those from Hall measurements by Chien *et al.* (Ref. 2) (open circles, converted into the data under B = 1 T).

for 90-K YBCO, m = 1.9, c = 186 for Zn 0.7%-doped YBCO, and m = 1.8, $c = 684 \pm 46$ for Zn 2.8%-doped YBCO.

It is well known that the Hall angle for impurity-doped YBCO behaves like $\cot \theta_H = b_H T^2 + c_H$, here c_H changes in proportion to the impurity concentration² and b_H reflects the carrier density.¹⁶ The coefficient *m*, an adjustable parameter, is necessary to express the right-hand side of Eq. (1) by the Hall angle $\langle \theta(s) \rangle$, assuming that all three quantities in Eq. (1) share the same temperature dependence: $(b_H T^2 + c_H)^{-2}$. The obtained *m* value means that $\langle \theta(s)^2 \rangle \approx 5 \langle \theta(s) \rangle^2$. The observed MR $(\Delta \rho / \rho)$ is comparable to $\langle \theta(s)^2 \rangle$ and is larger than $\langle \theta(s) \rangle^2$ over the measured temperature range. This is just the opposite case, where the observed value is a small remnant produced by the subtraction of large values. In that case, special attention to both measurement and analysis is required.

Even for the Zn-doped YBCO, data of the higher temperature region are well explained only by the normal-state MR of the form $\{m/[bT^2+C(x)]\}^2$, where x is the Zn concentration. Moreover, as shown in Fig. 4, the C(x) obtained from our MR fit coincides well with that of the Hall measurement by Chien *et al.*² There is scarcely a portion of the

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MR which should be assigned to the MT process, although a rather small contribution cannot be ruled out from the data. Similar coincidence of the Hall measurement and the MR fit is reported for a single crystal $La_{1.83}Sr_{0.17}CuO_4$.³

From the above analysis, we must conclude that at least above T_c , in the optimally doped 90-K and Zn-doped YBCO, the Maki-Thompson fluctuation seems to be vanishingly small. However, there is increasing evidence of dwave, and in general, gapless superconductivity in YBCO.^{28–30} The MT paraconductivity originates from the phase-correlated diffusive mode of the "normal fluid," so the gaplessness might be insufficient for the MT process to vanish.³¹ Rather, it may be originated from the unconventional normal-state property of HTSC's. The MR, which has been assigned to the MT-orbital process,^{1,9,18} should be assigned to the genuine normal-state MR which is proportional to a square of the Hall angle. In addition to the Hall effect, MR also supports the hypothesis of two relaxation times in HTSC's.^{2,3,32,33} It might be a consequence of the non-Fermiliquid nature of HTSC's as described by the resonating valence bond theory^{32,34} or the theory of spin-charge-separated quantum liquids.35

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