

Fluctuation conductivity in the 110-K phase of Ni-doped (Bi,Pb)-Sr-Ca-Cu-O superconductors

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The fluctuation conductivity above T_c was measured in (Bi,Pb)-Sr-Ca-(Cu_{1-x},Ni_x)-O superconductors having the 110-K phase. The temperature dependence of the paraconductivity can be described by a power law following Aslamazov-Larkin theory. For nondoped ($x=0$) samples, the conductivity data show a two-dimensional fluctuation of the order parameter, whereas Ni-doped samples show lower-dimensional fluctuations and reduced widths of the resistive transition. The critical exponent of the power law and its amplitude are well correlated with T_c , which decreases monotonously with increasing x up to ~ 0.02 and then saturates around 90 K over the range $0.02 \leq x \leq 0.1$. The observed crossover from two- to one- (or lower-) dimensional behavior with increasing Ni dopant level may be related to a fractal nature for the system. The T_c reduction of the present system is discussed from the standpoint of pair-breaking effects induced by the Ni doping.

I. INTRODUCTION

Series of high- T_c oxide systems commonly show two-dimensional (2D) characteristics in both normal and superconducting properties, with large anisotropy between in-plane and out-of-plane directions.¹ It is well known that measurements of the fluctuation conductivity above T_c (the paraconductivity) afford direct information on the dimensionality of the order parameter. Such studies are very useful for the cuprate superconductors where fluctuation effects can readily be observed even in bulk samples because of the extremely short coherence length. Experimental results for several of the oxide systems support the 2D fluctuation of the superconducting order parameter,²⁻¹⁰ though other results suggest crossover of the dimensionality from 2D to 3D (Refs. 11-14) or even 3D behavior.¹⁵⁻¹⁷ Recent studies on high-quality samples have indicated that the paraconductivity is basically 3D in Y-Ba-Cu-O (YBCO) type compounds¹⁸ and 2D in Bi-Sr-Ca-Cu-O (BSCCO) or Tl-Ba-Ca-Cu-O related compounds with the crossover from 2D to 3D near T_c .^{19,20}

The paraconductivity is generally discussed on the basis of the Aslamazov-Larkin (AL) and Maki-Thompson (MT) theories.²¹ For a high- T_c superconductor, however, the latter contribution can be neglected because of large depairing effects due to strong phase breaking scattering inherent in these kinds of materials.^{22,23} On this assumption, we can analyze the paraconductivity σ' of high- T_c oxides in terms of a simple power law

$$\frac{\sigma'}{\sigma_0} = \frac{\sigma(T) - \sigma_n(T)}{\sigma_0} = \frac{C}{n} \eta^{-n}, \quad (1)$$

where $\sigma(T)$ is the temperature-dependent conductivity measured, $\sigma_n(T)$ is the normal-state conductivity to be deduced from $\sigma(T)$ data at higher temperatures, σ_0 is the conductivity at some high temperature without the fluctuation (300K), $\eta = \ln(T/T_c) [\sim (T - T_c)/T_c]$ is the reduced temperature, and C and n are treated as adjustable parameters. We have included the critical exponent n in the amplitude of the power law, which would make it possible to express parameter C in a unified way among systems with different fluctuation dimensionality. In Eq. (1) n is related to the dimensionality D of the order parameter, such that

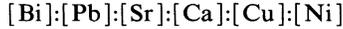
$$n = 2 - D/2. \quad (2)$$

In most cases the paraconductivity for high- T_c materials has been well described by the AL theory, or alternatively by the Lawrence-Doniach (LD) theory for layered superconductors in which only the AL process of the fluctuation is taken into account.²⁴

We have recently reported the effects of Ni substitution for Cu in the 110-K "2:2:2:3" phase of (Bi,Pb)-Sr-Ca-Cu-O compounds, and found that the T_c of the system reduces linearly for small Ni concentrations ($x < 0.03$) and then saturates around 90 K for higher dopant levels ($0.03 \leq x \leq 0.1$).²⁵ This behavior is qualitatively similar to that observed in Ni-doped BSCCO samples within the 80-K "2:2:1:2" phase.²⁶ Comparing the effects of Ni doping between the two phases, we have deduced that Ni in our present system substitutes in place of Cu not only in the pyramids but also the central Cu-O sheet. In this circumstance it is of interest to investigate the effects of the Ni substitution on the dimensionality of the order parameter. Experimental data were analyzed within the AL theory to determine the conduction dimensionality. The present results show a crossover of dimensionality with increasing Ni concentration.

II. EXPERIMENTS

The samples under study were prepared by the usual solid-state reaction with starting molar ratios



$$= 1.84:0.34:1.91:2.03:3.06(1-x):3.06x$$

Methods of sample preparation and characterization have been described previously.²⁵ Here we simply give a summary of our structural analyses. The 110-K phase was uniquely obtained up to $x=0.015$, beyond which the 80-K phase appears and grows with increasing x , the volume fraction between the two phases becoming comparable at $x=0.1$. For $x \geq 0.06$ some impurity phases were also detected. From x-ray photoemission studies, the Cu $2p$ peak position was found to shift towards the lower-energy side, while peaks for other atoms remained unchanged, indicating that Ni substituted for Cu only. The temperature dependence of the resistance in each sample was measured in detail using a fully automatic cryostat system equipped with a 10-K refrigerator. A platinum resistor was used as temperature sensor. The temperature scanning rate was slow enough (~ 1 K/min) to reproduce the resistance data with thermal cycling. The sample resistance was measured at every 0.15 K near T_c and 0.7 K for higher temperatures by means of a dc four-probe technique with a constant current of 1 mA, and then transformed into the resistivity by referring its value to that obtained by the van der Pauw method at room temperature.

III. RESULTS AND ANALYSES

In Fig. 1 we show the temperature dependence of the resistivity for our pure ($x=0$) and Ni-doped BSCCO compounds. All the samples measured here show linear temperature dependence of the resistivity above 150 K, as indicated by the solid lines expressed as $\rho_n(T) = AT + B$, where values of constants A and B are determined so as to fit the experimental data. The following method has been adopted to determine the values of these parameters. Choosing randomly one data point (T_1, ρ_1) between 180 and 230 K and another point (T_2, ρ_2) between 230 and 280 K we produce a set of values for A and B . This

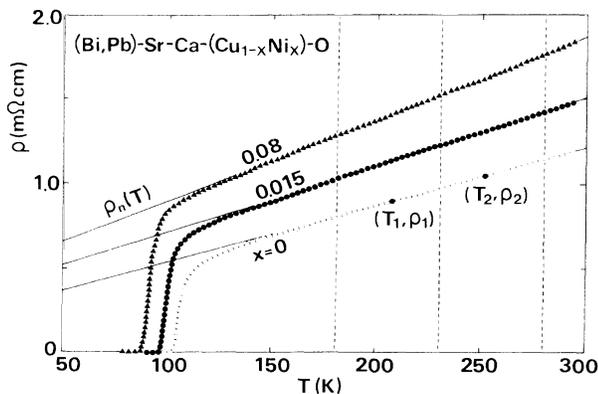


FIG. 1. Temperature dependence of the resistivity for (Bi,Pb)-Sr-Ca-(Cu_{1-x}Ni_x)-O samples with $x=0, 0.015$, and 0.08 . The solid lines represent the fitting form to the resistivity $\rho_n(T) = AT + B$. The broken lines define the two regions from which $\rho_n(T)$ is determined (see the text).

procedure is repeated a thousand times using a personal computer, and then values of A and B are obtained by taking the simple algebraic average. We have found that the present method gives more systematic results than a least-squares fitting. The deviation from the extrapolated linear dependence of $\rho_n(T)$ can be regarded as the excess conductivity due to the thermal fluctuation effect on the superconducting order parameter.

Determination of T_c is perhaps the most important problem in the analysis of the paraconductivity. The following three methods have been used for high- T_c superconductors: T_c is defined as a temperature (1) at which the resistivity of a sample vanishes or becomes a half of its normal-state value; (2) at which the temperature derivative of the resistivity $d\rho/dT$ has its maximum value or equivalently $d^2\rho/dT^2=0$; (3) T_c is determined so as to give the best fitting to the theory. Some authors have chosen a value of T_c by combining these methods. It is in fact possible to determine the dimensionality without knowing T_c by using the derivative method.^{5,7,20} Even with this technique, however, ambiguity still exists in the exponent of the conductivity behavior since data can be more scattered in derivative plots. While the last method commonly is used for conventional superconducting metallic films or alloys, it is not the case for our high- T_c oxides because of the intrinsic ambiguity in the conduction dimensionality. We have adopted the second method because of its well-defined nature, as indicated below. This method has been shown to provide a T_c value very close to the mean-field T_c from Eq. (1).⁵

Figure 2 shows the temperature dependence of the resistivity and its derivative $d\rho/dT$ near the superconducting transition for samples with $x=0$ and 0.08 . The arrows indicate the positions of T_c determined in the above-mentioned way. The paraconductivity can clearly be seen over a wide range of temperature, particularly in the case of the nondoped sample. The sharper structure of the $d\rho/dT$ vs T relation observed in Ni-doped samples indicates a narrower resistive transition width, thereby implying the suppression of the fluctuation.

In order to investigate the influence of the choice of T_c

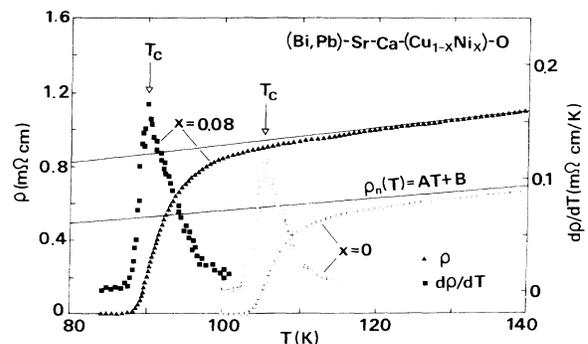


FIG. 2. Temperature dependence of the resistivity ρ and its derivative $d\rho/dT$ near T_c for (Bi,Pb)-Sr-Ca-(Cu_{1-x}Ni_x)-O samples with $x=0$ and 0.08 . The solid lines represent $\rho_n(T)$. T_c is defined from the criterion $d^2\rho/dT^2=0$, and is indicated by arrows.

TABLE I. Parameter values for the normal-state resistivity and the paraconductivity in (Bi,Pb)-Sr-Ca-(Cu_{1-x},Ni_x)-O systems. n , C , and T_c are plotted against x in Fig. 5.

x	$A = d\rho_n/dT$ ($\mu\Omega$ cm/K)	$B = \rho_n(0)$ (m Ω cm)	$\rho_n(300)$ (m Ω cm)	T_c (K)	n	C	σ'/σ_0 ($\eta=0.1$)
0	3.37	0.219	1.23	105.6	1.10	0.0210	0.239
0.005	3.56	0.331	1.40	101.3	1.20	0.0180	0.238
0.010	3.46	0.273	1.31	100.9	1.25	0.0163	0.231
0.015	3.86	0.351	1.51	98.8	1.40	0.0120	0.214
0.020	4.04	0.467	1.68	93.8	1.42	0.0128	0.237
0.040	4.47	0.530	1.87	92.5	1.60	0.0064	0.159
0.060	4.14	0.387	1.63	89.7	1.90	0.0044	0.285
0.080	4.73	0.442	1.86	90.1	1.80	0.0054	0.189
0.100	4.56	0.413	1.78	91.9	1.82	0.0047	0.172

on the conductivity analysis, we show in Fig. 3 log-log plots of the normalized conductivity σ'/σ_0 and the reduced temperature η , where σ_0 is a value of the conductivity at 300K obtained from the relation $\rho_n(T) = AT + B$. The results have been shown for a non-doped sample in the cases of three different T_c determinations. The criteria used are (a) $\rho = 0$, (b) $d^2\rho/dT^2 = 0$, and (c) $\rho = \rho_n/2$, giving T_c values 103.5, 105.6, and 106.6 K, respectively. The case for the criterion (b) gives the best adherence to the theoretical prediction Eq. (1) over the widest temperature range. Furthermore, the data for (b) closely corresponds to the case when the T_c is treated as a free parameter (the best-fitting value being $T_c = 105.8$ K) in the 2D AL theory portrayed by the solid line in Fig. 3. As has been pointed out,^{13,18} the results for $\ln \eta > -3$ are not so sensitive to the choice of T_c provided that values of T_c do not differ by much.

The effects of the Ni substitution upon the paraconductivity are shown in Fig. 4, where the data have been displayed in a similar manner to Fig. 3. The solid lines

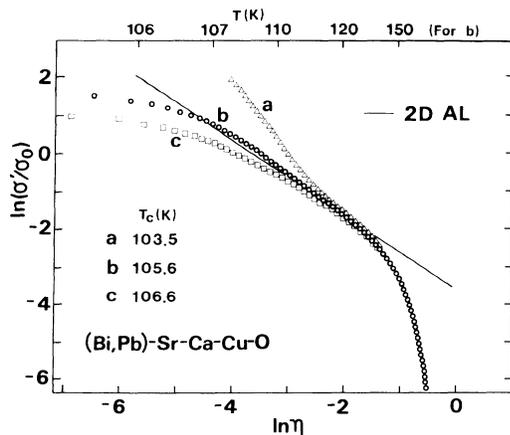


FIG. 3. Log-log plots for the reduced paraconductivity σ'/σ_0 vs the reduced temperature $\eta = \ln(T/T_c)$ for an undoped (Bi,Pb)-Sr-Ca-Cu-O sample, indicating influence of the choice of T_c on the data. σ_0 is the value of the conductivity at $T = 300$ K. The criteria to determine T_c are (a) $\rho = 0$, (b) $d^2\rho/dT^2 = 0$, and (c) $\rho = \rho_n/2$, giving T_c values indicated in the figure. The solid line represents the 2D AL theory Eq. (1) with $n = 1$ and $C = 0.024$.

have been obtained from Eq. (1) by choosing values of n and C so as to fit the data over the temperature range of $-3.5 \lesssim \ln \eta \lesssim -1.5$, where results do not depend too sensitively on the procedures to determine either T_c or $\rho_n(T)$. In this temperature region all the samples show conductivity data describable by Eq. (1). Values of T_c , n , and C in each sample are listed in Table I together with parameters for the normal-state resistivity. At temperatures outside the above range of $\ln \eta$ the data for all the samples fall below the theoretical lines, which might indicate a crossover of conduction dimensionality. However, it ought to be noticed that the simple power law of the AL theory can no longer hold in the region very close to T_c , nor again at much higher temperatures. For instance, in the former region interaction among Cooper pairs becomes important, while in the latter case terms other than Eq. (1) can modify the conductivity, neither of which circumstance is considered in simple AL theory.

As indicated in Fig. 4 the theoretical line for the sample with $x = 0$ has the slope $n = 1.1$, very close to the 2D fluctuation case ($n = 1$) for the order parameter. Summarizing experimental results for the paraconductivity so

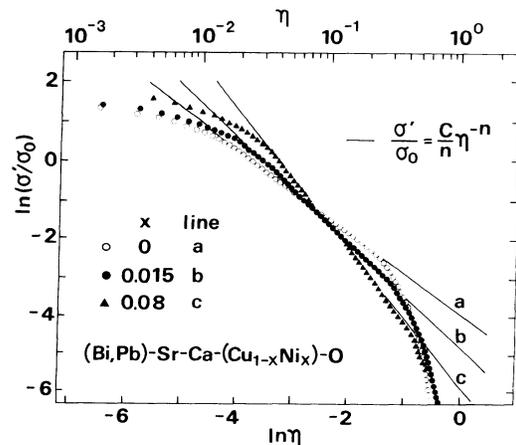


FIG. 4. Log-log plots for the reduced paraconductivity σ'/σ_0 vs the reduced temperature η for (Bi,Pb)-Sr-Ca-(Cu_{1-x},Ni_x)-O samples with $x = 0, 0.015$, and 0.08 . The solid lines represent the theoretical expression (1) fitted to the experimental data in the range $-3.5 \lesssim \ln \eta \lesssim -1.5$. Parameter values are listed in Table I.

far reported, YBCO compounds show 2D or 3D fluctuation depending on the nature of samples, while BSCCO or Tl-Ba-Ca-Cu-O materials (in both higher- and lower- T_c phases) have a 2D-like behavior even in polycrystalline samples. Clearly the latter materials having larger anisotropic character to their electronic properties show more marked 2D behavior. Our results for $x=0$ are therefore consistent with those observed by others. With regard to the Ni-doped samples it is obvious that the negative slope becomes steeper with x , indicating rapid development of lower-dimensional fluctuations. The theoretical lines for samples with different x tend to intersect at a point near $\eta=0.1$ where σ'/σ_0 has nearly the same values, as listed in Table I. This behavior is quite different from that in Fig. 3, and indicates that the changes in the slope cannot be a consequence of the choice of T_c nor $\rho_n(T)$ either. The values of C also decrease rapidly with increasing Ni concentration.

IV. DISCUSSION

The present analysis of the paraconductivity shows systematic changes in both the critical exponent and the amplitude for the Ni-doped BSCCO system. In Fig. 5 we display values of n and C as a function of x , and compare these with T_c variations for the system. There are close correlations among the parameters. Although the dependence of T_c on x is not quite the same as in our previous paper²⁵ in which T_c was determined from the zero-resistance criterion, its main features remain unchanged. With increasing x , T_c decreases monotonously in the region up to $x \sim 0.02$, beyond which T_c has a constant value of around 90 K. It is to be remembered that n is related to D via Eq. (2). In the AL theory values of D may be thought meaningful only for integers 1, 2, or 3, corresponding to $n = \frac{3}{2}$, 1, or $\frac{1}{2}$, respectively. (It might be possible to include the case of $D=0$ and $n=2$ for systems with fine particles.²⁷) However, it has been shown that

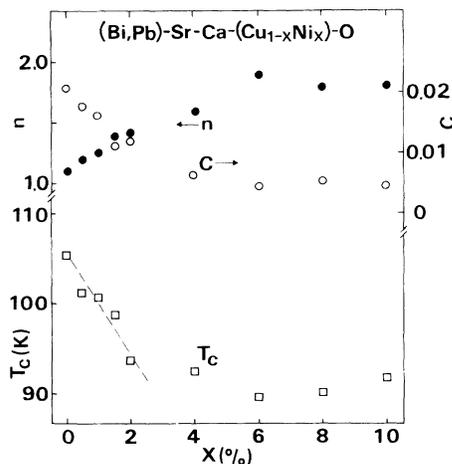


FIG. 5. Dependence of T_c , n , and C on the Ni concentration x obtained from the analyses of the paraconductivity for (Bi,Pb)-Sr-Ca-(Cu_{1-x}Ni_x)-O samples. The broken line indicates a linear reduction in T_c over the range $0 \leq x \leq 0.02$.

Eq. (2) can be generalized to embrace noninteger values of the dimensionality for inhomogeneous or granular superconductors.²⁸ There exists some experimental support for such a fractal description in the high- T_c oxides.^{27,29} In our case n increases with x , having a value around $\frac{3}{2}$ (1D behavior) at $x=0.02$, and tending to saturate between $\frac{3}{2}$ and 2 (possibly 0D) at higher dopant levels. Ying and Kwok³⁰ have reported that YBCO films can show 1D behavior in the paraconductivity in the region close to T_c , and that the results can be recognized as being due to the imperfection of the 2D character in the system. The conduction behavior with even lower dimensionality of order parameter observed in our samples probably is due to a percolative transport being induced by the Ni doping. This interpretation suggests that Ni must substitute for Cu ions not only in the pyramids (as in "2:2:1:2") but also in the central square planar Cu-O array.

In the AL theory the temperature-independent amplitude C depends on the characteristic size of sample dimensionality, so that the question might arise as to whether it is meaningful to compare the magnitude of C among samples having fluctuations of different dimensionality. However, it is to be noted that our results for the amplitude and the transition width are self-consistent. The magnitude of C can be regarded as a parameter which gives the resistive transition width associated with the fluctuation.⁵ Accordingly, the addition of Ni sharpens the resistive transition, as can be seen in Fig. 2, by suppressing the thermal fluctuation of the order parameter. This result is directly in line with the magnetic impurity effects in standard superconductivity, which can give rise to a significant depression in the paraconductivity as well as in T_c itself due to their pair-breaking efficacy. In a conventional 2D superconductor depairing effects have been introduced through the so-called pair-breaking parameter included in the MT term. It has been shown theoretically that magnetic impurities will affect the paraconductivity due mainly to suppression of the MT term.³¹ However, we do not expect a significant contribution from the MT term in our paraconductivity data, even in nondoped samples, because there is no indication for the $n=0$ behavior that provides direct evidence for a 2D MT contribution.

Some authors have discussed the magnitude of the amplitude of the power law, which in general depends on the sample quality. A possible explanation for this may be nonuniform current flow caused by sample inhomogeneity.^{11,5} Systematic changes in the amplitude and the exponent similar to the present results have been reported by Pureur *et al.*²⁹ in their study of the resistive transition for YBCO-type materials with inhomogeneities. They have shown such systems to have a critical exponent between 1 and $\frac{3}{2}$, and that the exponent becomes larger for the case of smaller amplitude, while T_c is independent of either of them. A similar correlation between the amplitude and the exponent may then be expected in other samples of fractal nature.

The present system includes some lower- T_c phase for $x \gtrsim 0.02$, and hence there is the possibility that this phase

may have some influence upon the dimensionality of the order parameter. In fact $\rho(T)$ behavior reported by Poddar *et al.*³² indicates that an undoped BSCCO sample mainly in the 110-K phase indeed shows paraconductivity over a wider range of temperature than that having both 80- and 110-K phases. From the following reasons, however, we conclude that our main results are not simply due to inclusion of the lower- T_c phase: (i) Values of n and C change significantly already in the region $0 \leq x \lesssim 0.02$ where the samples are thought to be single phase having sharp T_c values larger than or around 100 K; (ii) If T_c for the second phase is assumed to be 80 K, then the condition $\ln \eta \gtrsim -1.5$ under which the fluctuation effects are not important is operative for $T \gtrsim 100$ K; (iii) As can be seen in Fig. 3, the choice of a T_c value higher than that of the real T_c would cause a less steep negative slope and hence a higher-dimensional conduction, the opposite to what we have obtained in our system.

Next we would like to comment on the T_c reduction for our system from the standpoint of magnetic depairing. According to the Abrikosov-Gor'kov theory³³ for a BCS superconductor, T_c can be expressed as

$$\ln \left[\frac{T_{c0}}{T_c} \right] = \psi \left[\frac{1}{2} + \frac{\Gamma}{2\pi k_B T_c} \right] - \psi \left[\frac{1}{2} \right], \quad (3)$$

where T_{c0} is the value of T_c in the absence of magnetic impurities, ψ is the digamma function, and Γ is the pair-breaking parameter proportional to the magnetic impurity concentration. This formula gives the initial reduction in T_c as being linear in Γ : $T_c/T_{c0} \sim 1 - (\pi\Gamma/4k_B T_{c0})$. The results for T_c vs x shown in Fig. 5 and also those in our previous paper support such a linear reduction over the dopant range $0 \leq x \lesssim 0.02$. The saturation of T_c for higher x almost certainly results from the solubility limit x_c of Ni in the host material, which is roughly estimated as $x_c \sim 0.02$.²⁵ Beyond this value addition of Ni then is much less effective in suppressing superconductivity, both in 80- and 110-K phases. The above consideration is based on the magnetic impurity effect within standard BCS theory. A calculation of T_c in a system with magnetic impurities has recently been made on the basis of a negative- U model by one of the authors.³⁴ The results are qualitatively similar to those in the Abrikosov-Gor'kov theory except that the reduction in T_c is much weakened in a negative- U superconductor. Although a direct comparison with the calculation is difficult to make due to a lack of information on parameter values characteristic of the negative- U model, relative insensitivity to magnetic depairing would appear accordingly to be a general result for high- T_c materials with magnetic dopants.

From the above discussion, magnetic depairing effects due to Ni doping seem to give a satisfactory explanation of the present results for the reduction in both paraconductivity and T_c . However, it is necessary to point out that pair-breaking effects in the superconducting oxides

are not restricted to magnetic scattering. Ample evidence for phase-breaking scattering from nonmagnetic impurities and defects comes in systems under atomic substitution as an upturn in the behavior of $\rho(T)$ at low T . This may be attributed to electron localization driven by the random impurity potential.³⁵ The localization regime can quickly emerge for La-Sr-Cu-O and YBCO compounds having quite small amounts of dopants and can be seemingly independent of whether impurities are magnetic (e.g., Ni) or nonmagnetic (e.g., Zn).³⁵⁻³⁷ In consequence T_c in these systems is quickly reduced with increasing impurity concentration. On the other hand BSCCO-type compounds are more stable against localization effects, in the sense that the resistivity upturn behavior is observed only for systems having much higher dopant levels.²⁶ This fact allowed us to model the normal-state resistivity in the present system by a simple linear relation $\rho_n(T) = AT + B$, even though the effects of disorder are apparent in the fractal nature observed for the paraconductivity.

The T_c reduction in La-Sr-Cu-O and YBCO via the seemingly nonmagnetic impurities Zn or Ga can, it seems, be explained by a model in which these impurities also effectively behave as magnetic scatterers.³⁸ There are a number of experiments indicating that these perturbing ions induce magnetic moments in host high- T_c materials.³⁹⁻⁴¹ It is not clear at the present stage if the same circumstances will apply to BSCCO systems containing nonmagnetic impurities. Maeda *et al.*²⁶ have recently claimed that Zn-doped BSCCO in the 80-K phase (which has a T_c reduction similar to that for the case of Ni doping) shows no evidence of localized magnetic moments. Clearly further studies are required to clarify the role of nonmagnetic dopants in the BSCCO and Tl-Ba-Ca-Cu-O-type compounds.

V. CONCLUSION

We have investigated the effects of Ni substitution in a BSCCO system with the 110-K phase in the fluctuation regime. The paraconductivity has been analyzed within the context of the AL theory based on the assumption that the normal-state resistivity can be expressed as a simple linear relation and that the T_c is determined from the criterion $d^2\rho/dT^2=0$. The system shows a crossover of the order-parameter dimensionality from 2D to 1D or lower with increasing Ni concentration. The falling dimensionality could be the result of a fractal nature induced in the system by the impurity doping. Our conclusion on the dimensionality cannot be explained by the existence of the lower- T_c phase. The reduction in T_c for small dopant levels can be interpreted in terms of pair-breaking effects due to magnetic impurities (which are weaker in negative- U than in conventional superconductors). The width of the superconducting transition was found to be decreased by increasing Ni concentration,

which lends support to the existence of magnetic depairing in the present system. Nonetheless we anticipate for a local negative- U situation⁴² that comparable results will be obtained through substitution by nonmagnetic ions Zn^{2+} even in better metals like BSCCO, following a more generalized pair-breaking role. This we intend to pursue.

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