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Fluctuations in the Hall conductivity and dynamics of the order parameter in $YBa_2Cu_3O_{7-\delta}$ superconductors

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Fluctuation contributions to the Hall and longitudinal conductivities in $YBa_2Cu_3O_{7-\delta}$ epitaxial films have been investigated. Two remarkable features were observed: (1) the fluctuation contribution to the Hall conductivity, $\sigma_{xy}^{\mathrm{fl}}$, changes sign at temperatures $T^*(H) > T_c(H=0)$ and (2) the ratio $\sigma_{xy}^{\mathrm{fl}}/\sigma_{xx}^{\mathrm{fl}}$, where $\sigma_{xx}^{\mathrm{fl}}$ is the fluctuation longitudinal conductivity, shows a sharp minimum near $T_c(H)$. These features may reflect an unusual behavior of the imaginary part of the relaxation time of the order parameter in the vicinity of the transition temperature.

Thermodynamic fluctuations of the order parameter are known to affect different physical properties of the high-T_c superconductors: the longitudinal resistivity,[1-3] magnetization,[1-4] Ettinghausen coefficient,[5] heat capacity.[6] Data on the Hall effect in the fluctuation region are scarce. Separation of the fluctuation contribution to the Hall conductivity is difficult because the Hall effect is strongly temperature dependent in the normal state.[7] Assuming a linear temperature dependence of R_H^{-1} (where R_H is the Hall coefficient) in the normal state, Iye *et al.*[8] reported that the fluctuation Hall conductivity in $EuBa_2Cu_3O_{7-\delta}$ films has a sign opposite to the sign of the normal state Hall conductivity. Hagen *et al.*[9] found that fluctuations reduced the Hall resistivity ρ_{xy} from its normal state value and reported a sign change of ρ_{xy} near the upper critical field H_{c2}. The sign change of ρ_{xy} has been attributed[2,10,11] to the negative value of the particle-hole asymmetry parameter λ_0^{-1} which is equal to the imaginary part of the relaxation time of the order parameter γ: $\lambda_0^{-1}=\mathrm{Im}\gamma$. Aronov and Rapoport[12] have recently shown that in the region of Gaussian fluctuations the sign of λ_0^{-1} for $YBa_2Cu_3O_{7-\delta}$ should be positive.

In this paper I report on very careful measurements of the Hall resistivity, ρ_{xy}, and the longitudinal resistivity, ρ_{xx}, in $YBa_2Cu_3O_{7-\delta}$ epitaxial films. The purpose of this study is to obtain the Hall conductivity, σ_{xy}, as well as the longitudinal conductivity, σ_{xx}, and to separate the fluctuation contributions to these quantities, $\sigma_{xy}^{\mathrm{fl}}$ and $\sigma_{xx}^{\mathrm{fl}}$, respectively. The fluctuation Hall conductivity $\sigma_{xy}^{\mathrm{fl}}$ turns out to change sign, from a positive value corresponding to the sign of the normal state Hall conductivity σ_{xy}^{n}, to a negative value, which corresponds to the sign of σ_{xy} in the mixed state. This occurs at temperatures above the zero-field transition temperature. The ratio $\sigma_{xy}^{\mathrm{fl}}(T)/\sigma_{xx}^{\mathrm{fl}}(T)$ at a fixed magnetic field has a minimum near the mean-field transition temperature $T_c(H)$ as determined from the scaling of the fluctuation conductivity $\sigma_{xx}^{\mathrm{fl}}$. This investigation can provide information about anomalous behavior of the particle-hole asymmetry parameter in the vicinity of the transition temperature.

The epitaxial $YBa_2Cu_3O_{7-\delta}$ films have been prepared at the Institute of Applied Physics (Nizhniy Novgorod, Russia) by laser deposition onto $SrTiO_3$ substrates. Two films with thicknesses of 1000 Å referred to below as film 1 and film 2 have been used in this study and have shown similar results. Data presented are those for film 1. The midpoint zero-field transition temperature is approximately 90 K; the transition width is less than 1 K. The c axis is perpendicular to the substrate, the applied magnetic field $\mathbf{H}\parallel c$. The Hall resistivity has been measured by commutating the voltage and current probes as previously used by us for the investigation of $Bi_2Sr_2CaCu_2O_x$ single crystals.[13] The voltage has been checked to be linear in the direct current for the current strength 100–1000 μA within the whole range of magnetic fields (0.2 T$< H <$4 T) and temperatures (80 K$< T <$185 K). Most of the data have been obtained at a current of 600 μA which corresponds to a current density $j \approx$1200 A/cm^2. The presence of eight probes on each film allows three independent measurements of ρ_{xy} and two independent measurements of ρ_{xx}. The variations of ρ_{xy} and ρ_{xx} obtained on different pairs of contacts on the same film do not exceed 3% and 1%, respectively. Within experimental error these variations are temperature independent, indicating high uniformity of the samples.

The temperature dependence of the Hall conductivity is shown in the inset in Fig. 1. Because $\rho_{xy}^2 \ll \rho_{xx}^2$, the Hall conductivity has been calculated as $\sigma_{xy} = \rho_{xy}/\rho_{xx}^2$. In the normal state, σ_{xy} is positive, proportional to the magnetic field strength, and strongly increases with decreasing temperature. As the temperature is lowered to the superconducting region, the Hall conductivity becomes negative and increases rapidly in magnitude, in agreement with the observation by Luo *et al.*[14] that the decrease of $\mid \rho_{xy} \mid$ is slower than ρ_{xx}^2 in $YBa_2Cu_3O_{7-\delta}$ in the mixed state. In agreement with single-crystalline $YBa_2Cu_3O_{7-\delta}$ data by Chien *et al.*,[15] the Hall conductivity in epitaxial films at $T >$110 K closely follows the relation $\sigma_{xy} \sim H/T^3$, where H is the magnetic field strength. This is demonstrated in Fig. 1 where the temperature dependences of $\sigma_{xy}T^3$ are plotted, although one can see slight deviations from the above relation with increasing temperature, in agreement with Ref. 15.

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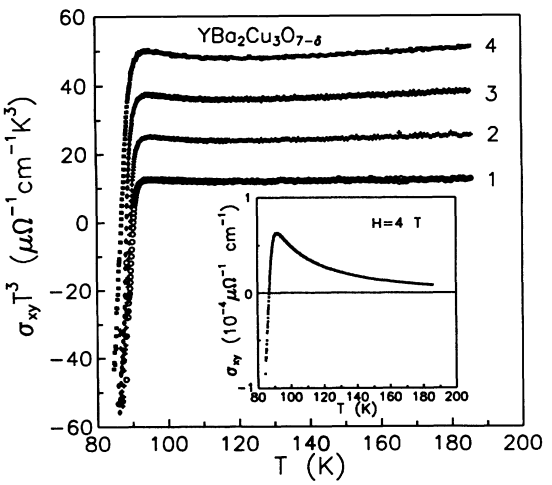

FIG. 1. Temperature dependence of $\sigma_{xy}T^3$ in various magnetic fields as indicated. Inset: Temperature dependence of σ_{xy} in a magnetic field of 4 T.

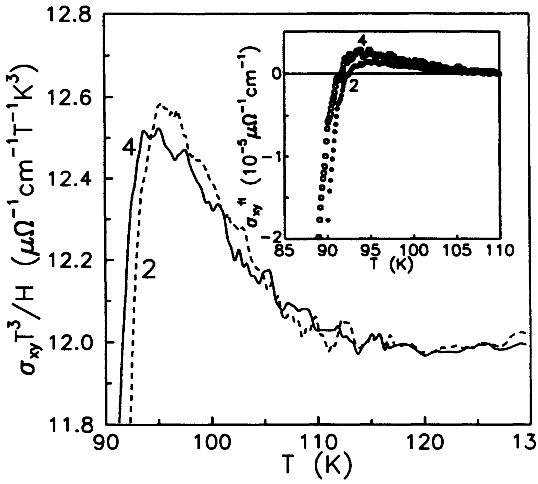

FIG. 2. Temperature dependence of $\sigma_{xy}T^3/H$ near T_c in magnetic fields of 2 T and 4 T. Inset: Temperature dependence of $\sigma_{xy}^{\mathrm{fl}}$ in magnetic fields of 2 T and 4 T.

Such a striking power law of the Hall conductivity can be explained within the theory by Anderson[16] in terms of spinon-spinon scattering, resulting in a T^2 law for the cotangent of the Hall angle ($\cot\theta = \rho_{xx}/\rho_{xy}$), and in terms of holon-spinon scattering, resulting in ρ_{xx} which is linear in T, if scattering by impurities is not very strong. Another insight into the Hall effect in $YBa_2Cu_3O_{7-\delta}$ proposed by Carrington *et al.*[17] is based on Fermi-surface calculations by Pickett *et al.*[18] This model involves a square Fermi surface with rounded corners. The unusual behavior of the Hall conductivity in the normal state may arise if the mean free paths on the curved and flat parts have different T dependences, but no concrete prediction for these dependences has been provided. Nonetheless, the relation $\sigma_{xy} = aH/T^3$ is approximately valid over a wide temperature range (up to 185 K, as one can see from Fig. 1, and up to 360 K as seen in Ref. 15). We find a=12 $(\mu\Omega\ \mathrm{cm\ T})^{-1}\mathrm{K}^3$ and 10.8 $(\mu\Omega\ \mathrm{cm\ T})^{-1}\mathrm{K}^3$ for film 1 and film 2, respectively. The deviation $\delta\sigma_{xy}/H$ of the Hall conductivity from the relation $\sigma_{xy} = aH/T^3$ between 110 K and 185 K and 1 T and 4 T is less than $1.5\times10^{-7}(\mu\Omega\ \mathrm{cm\ T})^{-1}$.

The temperature dependence of $\sigma_{xy}T^3$ near T_c is presented in Fig. 2. To show data for different magnetic fields, the Hall conductivity is divided by the magnetic field strength. To obtain higher resolution, data of Fig. 1 have been smoothed over a window of seven points (0.6 K). On cooling below 110 K–105 K, a pronounced deviation from the relation $\sigma_{xy} \sim 1/T^3$ occurs. As the temperature is lowered 10 K (from 105 K to 95 K), this deviation reaches a value of $\Delta\sigma_{xy}/H \approx 5.5\times10^{-7}$ $(\mu\Omega\ \mathrm{cm\ T})^{-1}$. For the same temperature range fluctuations are known to contribute significantly to different physical properties of $YBa_2Cu_3O_7$: the longitudinal resistivity, magnetization (see, e.g., Ref. 1), Ettinghausen coefficient,[5] specific heat.[7] It is natural to attribute the deviation from the relation $\sigma_{xy} \approx aH/T^3$ to a fluctuation effect also. The maximum in T dependence of $\sigma_{xy}T^3$ is shifted to lower temperatures and decreases in magnitude with increasing magnetic field. Measurements at a current of 3 mA have indicated that the latter result is not an artifact of the smoothing procedure.

In Fig. 2, inset, the temperature dependences of the fluctuation part of the Hall conductivity $\sigma_{xy}^{\mathrm{fl}} = \sigma_{xy} - \sigma_{xy}^{n}$, with σ_{xy}^{n}=12 H/T^3 $(\mu\Omega\ \mathrm{cm})^{-1}$, are shown for magnetic fields of 2 T and 4 T (data for σ_{xy} are not smoothed). At $T^* \approx$92 K, $\sigma_{xy}^{\mathrm{fl}}$ changes sign. This temperature is higher than the transition temperature in zero magnetic field. Chien *et al.*[15] have also observed an increase of $\sigma_{xy}T^3$ on cooling below 105 K–110 K which they have attributed to a fluctuation effect, but data at lower temperatures have not been presented.

In view of the fact that the temperature dependences of the Hall conductivity and/or of the Hall coefficient in the normal state are not completely understood, the question arises: To what extent does the calculated excess Hall conductivity depend on a fitting procedure for the normal state Hall conductivity? For instance, one might take into account a small deviation from the law $\sigma_{xy}T^3/H$ =const in the normal state, say, in the form $\sigma_{xy}T^3/H = \sum_n \alpha_n T^n$ (α_n are constants, n=0, 1, 2). This procedure would change the result for $\sigma_{xy}^{\mathrm{fl}}/H$ by a minor value $<2\times10^{-8}$ $(\mu\Omega\ \mathrm{cm\ T})^{-1}$ at 85 K< T <100 K. Iye *et al.*[8] used a linear temperature dependence for R_H^{-1} to calculate σ_{xy}^{n} in $EuBa_2Cu_3O_{7-\delta}$ films and reported a $\sigma_{xy}^{\mathrm{fl}}$ with negative sign. Although, as it has been pointed out by Chien *et al.*,[15] the Hall current rather than the Hall coefficient itself is the source of the anomalous Hall effect in the normal state and a linear temperature dependence does not provide an accurate description of data for R_H^{-1}, it follows from the data by Iye *et al.*[8] also that there is a temperature region where $\sigma_{xy}^{\mathrm{fl}}$ >0. Thus, at T=90 K (9 K higher than the zero resistance T_c of their film) their approximation for σ_{xy}^{n} at H=5 T gives 59.4 $(\Omega\ \mathrm{cm})^{-1}$, whereas the experimental value is approximately 62–63 $(\Omega\ \mathrm{cm})^{-1}$. So, independent of the approximation for σ_{xy}^{n}, both the analysis of data presented in this paper and data previously published[8,15] show that at temperatatures well above T_c the sign of $\sigma_{xy}^{\rm fl}$ is positive and corresponds to the sign of σ_{xy}^{n} and that on cooling the sign of $\sigma_{xy}^{\rm fl}$ changes and corresponds to the sign of σ_{xy} in the mixed state.

Two contributions to the fluctuation Hall conductivity are known to exist: One comes from currents carried by the superconducting fluctuations [the Aslamazov-Larkin (AL) process], while another comes from additional scattering by the fluctuations [the Maki-Thompson (MT) process] (see Ref. 19). The MT term for the Hall conductivity has the same sign as σ_{xy} in the normal state and therefore cannot explain the sign change of the fluctuation Hall conductivity.

Fukuyama *et al.*,[19] Abrahams *et al.*,[20] and recently Ullah and Dorsey[2] have discussed the fluctuation Hall conductivity in terms of dynamics of the order parameter, within the time-dependent Ginzburg-Landau (GL) equation. They have shown that the nonvanishing AL term for σ_{xy} appears if there is an imaginary part of the order parameter relaxation time ($\lambda_0^{-1} \neq 0$). A microscopic mechanism breaking particle-hole symmetry and resulting in a nonzero imaginary part of the order parameter relaxation time was considered in Ref. 19. λ_0^{-1} turned out to be proportional to the energy derivative of the density of states at the Fermi level. Within the Hartree approximation, Ullah and Dorsey[2] have calculated different transport properties of a superconductor near T_c in a magnetic field. They have found that in three-dimensional (3D) systems $\sigma_{xx}^{\rm fl} H^{1/3} T^{-2/3}$ should be a function of $[T - T_c(H)]/(TH)^{2/3}$ only, provided that Γ_0^{-1}=Reγ is a constant (see Refs. 10,19). The scaling functions for $\sigma_{xy}^{\rm fl}$ are the same as those for $\sigma_{xx}^{\rm fl}$, apart from the replacement of Γ_0^{-1} by λ_0^{-1}: $\sigma_{xy}^{\rm fl}/\sigma_{xx}^{\rm fl} = \lambda_0^{-1}/\Gamma_0^{-1}$.

Scaling of the longitudinal conductivity has been experimentally observed by Welp *et al.*[1] Following them, we show a 3D scaling plot for $\sigma_{xx}^{\rm fl}$ in the main frame of Fig. 3, while the inset depicts the data before scaling.

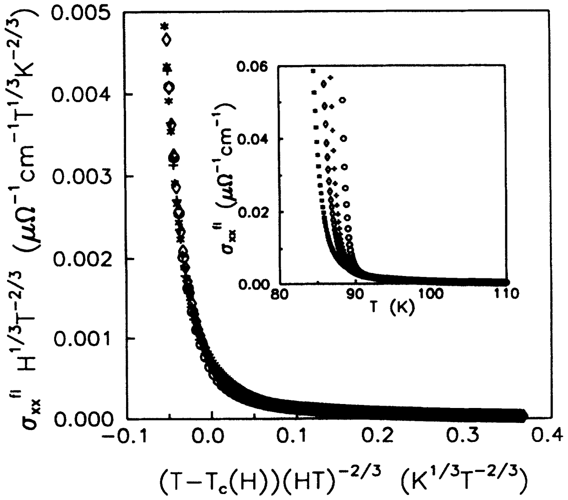

FIG. 3. 3D scaling of $\sigma_{xx}^{\rm fl}$. Inset: Temperature dependence of $\sigma_{xx}^{\rm fl}$ at H=1 T, 2 T, 3 T, and 4 T (from right to left).

We obtained $\sigma_{xx}^{\rm fl}$ by subtracting the normal state contribution $\sigma_{xx}^{n} = 1/\rho_{xx}^{n}$ from the measured data, where the normal state resistivity ρ_{xx}^{n} is assumed to be linear in temperature. This linear behavior is found between 130 K and 300 K, namely, ρ_{xx}^{n}=$1.24T$+7.3 $\mu\Omega$cm for film 1 and ρ_{xx}^{n}=$1.33T$+11.1 $\mu\Omega$cm for film 2. Below 130 K $d\rho_{xx}/dT$ deviates significantly. From the scaling procedure we obtain the values of $T_c(H)$ as fit parameters depicted by the closed circles in the inset in Fig. 4. They lie on a straight line with slope 1.4 T/K, in good agreement with Ref. 1.

Next we consider the ratio of $\sigma_{xy}^{\rm fl}$ to $\sigma_{xx}^{\rm fl}$ plotted in Fig. 4. According to theory,[12,19] the AL term in the Hall conductivity is more singular than in the longitudinal conductivity, while for the MT process the divergence of $\sigma_{xy}^{\rm fl}$ is the same as that of $\sigma_{xx}^{\rm fl}$. Aronov and Rapoport[12] have considered the effect of the Gaussian fluctuations on the Hall conductivity and have shown that for $\rm YBa_2Cu_3O_{7-\delta}$ the AL term should have the same sign as σ_{xy} in the normal state. So concentrating on the high-temperature results in Fig. 4, from the fact that $\sigma_{xy}^{\rm fl}$ is positive and that $\sigma_{xy}^{\rm fl}/\sigma_{xx}^{\rm fl}$ increases with decreasing temperature, we conclude that in the region of Gaussian fluctuations there is a noticeable positive contribution to σ_{xy} due to the AL process. On further cooling the ratio $\sigma_{xy}^{\rm fl}/\sigma_{xx}^{\rm fl}$ starts to decrease and changes sign because of the sign reversal of $\sigma_{xy}^{\rm fl}$. It only remains for us to assume that on cooling, the AL term changes sign from positive to negative.

As has been noticed above, the imaginary part of the order parameter relaxation time is positive and thus cannot explain the sign change of $\sigma_{xy}^{\rm fl}$ in superconducting

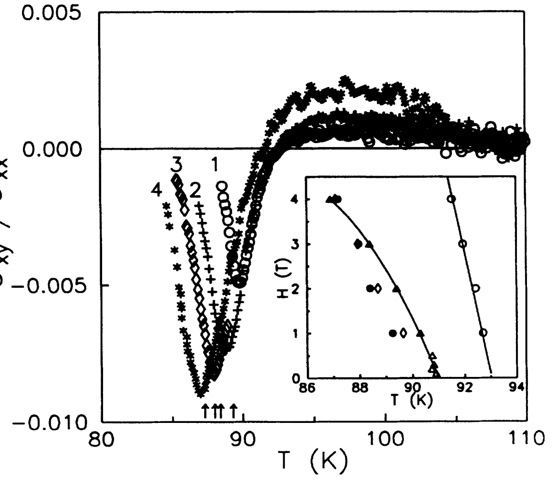

FIG. 4. Temperature dependence of $\sigma_{xy}^{\rm fl}/\sigma_{xx}^{\rm fl}$ in various magnetic fields as indicated. Data for H=3 T and 4 T are not shown above 89 K and 105 K, respectively. Arrows mark the positions of $T_c(H)$ determined from 3D scaling of $\sigma_{xx}^{\rm fl}$. Inset: The H-T diagram for the Hall effect in $\rm YBa_2Cu_3O_{7-\delta}$. Open circles and triangles indicate loci for the sign reversal of $\sigma_{xy}^{\rm fl}$ and σ_{xy}, respectively (solid lines are guides for the eye); diamonds indicate loci for the minima in the temperature dependences of $\sigma_{xy}^{\rm fl}/\sigma_{xx}^{\rm fl}$; solid circles: $H_{c2}(T)$ as determined from scaling of $\sigma_{xx}^{\rm fl}$.

$YBa_2Cu_3O_{7-\delta}$ (Ref. 12) if only Gaussian fluctuations are considered. We should point out a reason why the Aronov-Rapoport theory[12] breaks down. One can assume that the sign of the particle-hole asymmetry parameter in the critical region is opposite to that in the region where the fluctuations do not interact. The critical region in high-T_c superconductors is strongly broadened in comparison with the "conventional" superconductors due to large anisotropy, a large Ginzburg-Landau parameter κ, and a high transition temperature (see Refs. 1, 21). It is quite possible that interactions between the fluctuations become important at temperatures a few K above T_c (Ref. 21) and result in the sign change of λ_0^{-1}.

As has already been mentioned, within the Hartree approximation the temperature dependences of $\sigma_{xy}^{\text{fl}}/\sigma_{xx}^{\text{fl}}$ (Fig. 4) are those of λ_0^{-1}.[2] In the nearest vicinity of $T_c(H)$ marked by arrows in Fig. 4 there is a remarkable minimum in the temperature dependences of $\sigma_{xy}^{\text{fl}}/\sigma_{xx}^{\text{fl}}$ in a fixed magnetic field. This observation probably reflects anomalous behavior of the imaginary part of γ and thus a drastic change in the dynamics of the order parameter.

In addition to the ordinary AL term which comes from the nonzero λ_0^{-1}, the AL diagram with skew scattering gives also a nonvanishing contribution to the Hall conductivity if this diagram is considered to the next order, i.e., if the four-point skew-scattering vertex is involved.[22] The fluctuating part of σ_{xy} in this case has been shown to have a minus sign due to the next order of the GL Hamiltonian. The skew-scattering rate is proportional to the impurity concentration and spin of the spin-orbit interaction producing the skew scattering, but this spin has not been assigned in Ref. 22. This consideration is beyond the Hartree approximation used in Ref. 2. It would be desirable to have a realistic theoretical estimate of the skew-scattering rate in high-temperature superconductors. Usually, however, vertex corrections are small; see, e.g., Ref. 23.

The inset in Fig. 4 shows the H-T diagram for the Hall effect in $YBa_2Cu_3O_{7-\delta}$. The temperatures $T^*(H)$ (open circles) at which the superconducting fluctuations give rise to the sign reversal of σ_{xy}^{fl} are significantly higher than the mean-field transition temperatures given by the solid circles. On cooling below $T^*(H)$ the strong negative increase of σ_{xy}^{fl} eventually leads to the sign reversal of the Hall conductivity at a temperature denoted by the open triangles in the inset in Fig. 4. Also this temperature is seen to be larger than $T_c(H)$ which represents the mean-field transition temperature. This proves that the sign anomaly of the Hall effect needs an explanation in terms of general properties of the dynamics of the order parameter, both in the fluctuation regime and in the mixed state. This phenomenon occurs at relatively high temperatures [above $T_c(H)$] and therefore is not consistent with a suggestion by Wang and Ting[24] who have associated the sign anomaly with pinning.

To conclude, we have shown that the fluctuation Hall conductivity changes sign at $T > T_c(H = 0)$ in $YBa_2Cu_3O_{7-\delta}$. Within the theory by Ullah and Dorsey,[2] the data provide evidence that the imaginary part of the order parameter relaxation time λ_0^{-1} changes sign at $T > T_c(H = 0)$ and has a minimum at $T \approx T_c(H)$. It is probable that one should also take into account corrections due to the skew-scattering mechanism as proposed by Aronov and Hikami.[22] It is clear that to explain the data presented in this paper phenomenological approaches are not appropriate but a microscopic theory extended to the critical region is required.

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[1] U. Welp *et al.*, Phys. Rev. Lett. **67**, 3180 (1991).

[2] S. Ullah and A. T. Dorsey, Phys. Rev. B **44**, 262 (1991).

[3] K. Semba, T. Ishii, and A. Matsuda, Phys. Rev. Lett. **67**, 769 (1991); see also Ref. 5 in Ref. 1.

[4] W. C. Lee, R. A. Klemm, and D. C. Johnston, Phys. Rev. Lett. **63**, 1012 (1989).

[5] T. T. M. Palstra *et al.*, Phys. Rev. Lett. **64**, 3090 (1990).

[6] S. E. Inderhees *et al.*, Phys. Rev. Lett. **66**, 232 (1991).

[7] For a review, see N. P. Ong, in *Physical Properties of High-Temperature Superconductors*, edited by D. M. Ginsberg (World Scientific, Singapore, 1990), Vol. 2, p. 459.

[8] Y. Iye, S. Nakamura, and T. Tamegai, Physica (Amsterdam) **159C**, 616 (1989).

[9] S. J. Hagen *et al.*, Phys. Rev. B **41**, 11 630 (1990).

[10] A. T. Dorsey, Phys. Rev. B **46**, 8376 (1992).

[11] N. B. Kopnin, B. I. Ivlev, and V. A. Kalatsky, Pis'ma Zh. Eksp. Teor. Fiz. **55**, 717 (1992) [JETP Lett. **55**, 751 (1992)].

[12] A. G. Aronov and A. B. Rapoport, Mod. Phys. Lett. B **6**, 1083 (1992).

[13] N. V. Zavaritsky, A. V. Samoilov, and A. A. Yurgens, Physica (Amsterdam) **180C**, 417 (1991); A. V. Samoilov, Phys. Rev. Lett. **71**, 617 (1993).

[14] J. Luo *et al.*, Phys. Rev. Lett. **68**, 690 (1992).

[15] T. R. Chien *et al.*, Phys. Rev. B **43**, 6242 (1991).

[16] P. W. Anderson, Phys. Rev. Lett. **67**, 2992 (1991).

[17] A. Carrington *et al.*, Phys. Rev. Lett. **69**, 2855 (1992).

[18] W. E. Pickett *et al.*, Science **255**, 46 (1992).

[19] H. Fukuyama, H. Ebisawa, and T. Tsuzuki, Prog. Theor. Phys. **46**, 1028 (1971).

[20] E. Abrahams, R. E. Prange, and M. J. Stephen, Physica (Amsterdam) **55**, 230 (1971).

[21] An estimate of the width of the critical region ϵ_g based on the Ginzburg criterion predicts a value of the order of 0.1 K for $YBa_2Cu_3O_7$ [C. J. Lobb, Phys. Rev. B **36**, 3930 (1987)]. D. S. Fisher, M. P. Fisher, and D. A. Huse [Phys. Rev. B **43**, 130 (1991)] argue that the Ginzburg criterion underestimates ϵ_g by a factor as large as 25.

[22] A. G. Aronov and S. Hikami, Phys. Rev. B **41**, 9548 (1990).

[23] R. Ikeda, T. Ohmi, and T. Tsuneto, J. Phys. Soc. Jpn. **58**, 1377 (1989).

[24] Z. I. Wang and C. S. Ting, Phys. Rev. Lett. **67**, 3618 (1991).