RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 47, NUMBER 13 1 APRIL 1993-I

Normal-state anisotropic resistivity of $Nd_{2-x}Ce_xCuO_{4-y}$: Evidence for scattering by anisotropic spin fluctuations

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(Received 29 December 1992)

We report measurements of the anisotropic resistivity of the electron-doped copper oxide compound $Nd_{2-x}Ce_xCuO_{4-y}$ using the Montgomery method. The full temperature dependence of both resistivity components is well described using a simple Boltzmann transport analysis. Deduced values of the Fermi wave vector, $k_F \approx 0.2/a$, an effective interplanar coupling, $J_\perp \approx 39$ K, the effective mass ratio, $m_c/m_a \approx 600$, and the relevant electron-magnon matrix element, $|g_0'| \approx 180$ meV, are reported.

Much attention has recently focused on the anomalous normal-state properties of the metallic copper oxides; some of the less exotic proposals to explain the novel behavior have included Fermi-liquid-based approaches.[1-3] Although a variety of scattering mechanisms have been invoked in an attempt to explain the linear in-plane resistivity of the hole-doped copper oxides,[1-4] the nearly quadratic in-plane resistivity observed[5-8] for the electron-doped $Nd_{2-x}Ce_xCuO_{4-y}$ system has generally been termed "conventional,"[2,7] i.e., attributed to the well-established T^2 Fermi-liquid behavior due to electron-electron scattering.[9] However, the temperature coefficient of the T^2 term is anomalously large, 10^5-10^6 times greater than in conventional metals. Furthermore, no consensus exists concerning the departure from T^2 behavior at high T: both a correction due to two-dimensional (2D) scattering[6] and an electron-phonon contribution[7] have been suggested. Even more confusing are the reports of anisotropy in $Nd_{2-x}Ce_xCuO_{4-y}$: the perpendicular resistivity has been reported both to exhibit a T^2 behavior[10] and to display logarithmic corrections;[8] reported values of the resistive anisotropy vary by as much as a factor of 30.[8,10,11]

A dominant difference between hole and electron doping in the copper oxides is manifested in their magnetic behavior: the magnetic frustration that results from hole doping leads to a rapid decrease in spin correlations and a corresponding loss of long-range antiferromagnetic (AFM) order at very low concentrations, $x \approx 0.02$.[12] The primary effect of electron doping, however, is only to dilute the spin system, so that long-range spin correlations are maintained to very high concentrations;[13] indeed, long-range order with $T_N = 160$ K for $x = 0.15$ has been observed.[14] The presence of local moments correlated on length scales long compared to the electron mean free path provides a framework for a simple understanding of anisotropic transport in the electron-doped materials.

Here we present the first detailed study of the temperature dependence of the anisotropic resistivity of $Nd_{2-x}Ce_xCuO_{4-y}$ using the Montgomery method.[15] Previous measurements, using four-in-a-line,[6,7,16] inappropriate,[8] or unspecified[10] contact configurations, apparently provide limited or misleading results, including a mixing of the resistivity tensor components and a large underestimation of the anisotropy. We show that the full anisotropic resistivity can be understood in the simple framework of a nearly cylindrical Fermi surface with scattering by well-understood anisotropic spin fluctuations. Many salient features of the anisotropic transport in this system become transparent in this framework: the Fermi wave vector k_F scales inversely with impurity scattering, presumably due to localization effects; deviations from a nearly perfect T^2 in-plane behavior increase for smaller k_F due to Fermi-surface effects; the more nearly $T^{3/2}$ perpendicular resistivity arises naturally from spin anisotropy. We also obtain estimates of the effective-mass ratio, $m_c/m_a \approx 600$, and the electron-magnon scattering matrix element, $|g_0'| \approx 180$ meV.

Crystals ($\sim 2\times 1\times 0.1$ mm^3) were grown from high purity oxides (99.99% Nd_2O_3 and CeO_2, 99.999% CuO) in CuO-rich flux, similar to techniques described previously;[17] their single crystal nature was verified with x-ray Laue diffraction. Reduction anneals (750–950 °C) were performed to improve conductivity or induce superconductivity. Cerium concentrations, determined by energy dispersive spectroscopy, are accurate and spatially homogeneous to $\Delta x \approx 0.01$. Oxygen contents are only estimated ($\Delta y \approx 0.02$) by comparison to similar polycrystalline anneals. We stress that careful Montgomery method[15] analysis is critical for distinction between resistance and resistivity in highly anisotropic materials.

Typical anisotropic resistivity data for several crystals are shown in Figs. 1 and 2 as functions of the square of the absolute temperature over the temperature range 5–300 K. The upper (lower) inset shows typical raw resistance data for current applied along the c (a) direction. Experimental errors are smaller than the size of the data symbols. In the lower panels, the in-plane resistivity ($\rho_a \equiv \rho_\parallel$) is seen to vary markedly from sample to sample, with notable features: the temperature-dependent part varies very nearly as T^2, as noted previously,[5-8,10] and the slope of the T^2 term increases with the residual ($T=0$) resistivity. In the upper panels of Figs. 1 and 2, much different behavior is evident for the out-of-plane resistivity ($\rho_c \equiv \rho_\perp$); these data indicate that the temperature-dependent component of ρ_c both varies

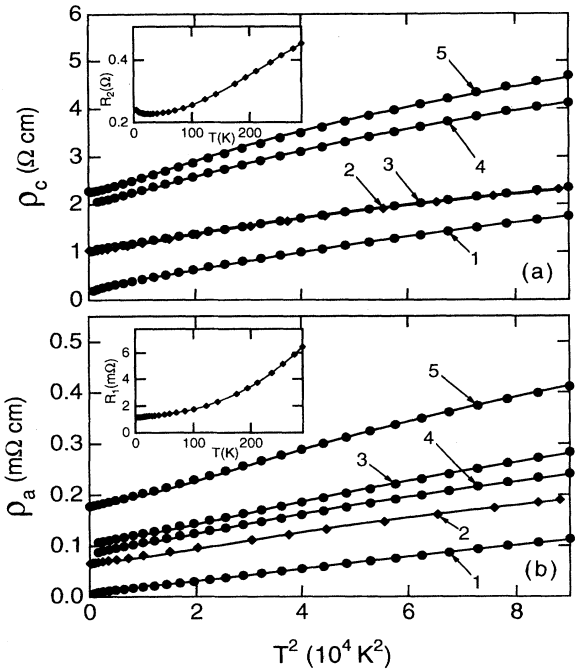

FIG. 1. Normal-state anisotropic resistivity of relatively low resistivity specimens of $Nd_{2-x}Ce_xCuO_{4-y}$ as a function of the square of the absolute temperature. (a) Out-of-plane resistivity ($\rho_c=\rho_\perp$); (b) in-plane resistivity ($\rho_a=\rho_\parallel$). Lines are fits to a theory of scattering by anisotropic spin fluctuations. Samples are (1) $x=0.19$, $y\approx0.03$; (2) $x=0.20$, $y\approx0.01$; (3) $x=0.19$, $y\approx0.01$; (4) $x=0.19$, $y\approx0.02$; and (5) $x=0.18$, $y\approx0.02$. Insets: raw resistance data for sample 2, with dimensions $475\times110\times18.6\ \mu m^3$.

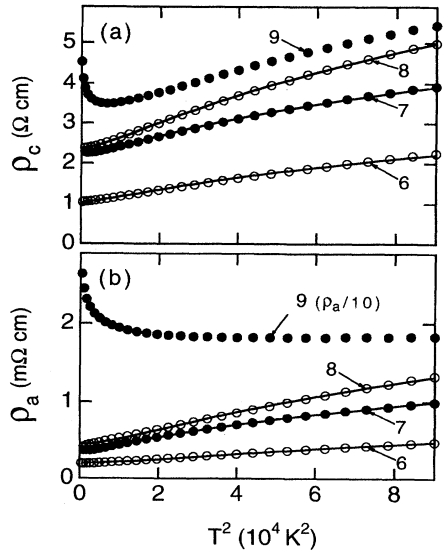

FIG. 2. Normal-state anisotropic resistivity of relatively high-resistivity specimens of $Nd_{2-x}Ce_xCuO_{4-y}$ as a function of the square of the absolute temperature, similar to Fig. 1. Samples are (6) $x=0.18$, $y\approx0.02$; (7) $x=0.20$, $y\approx0.00$; (8) $x=0.16$, $y\approx0.02$; and (9) $x=0.19$, $y\approx-0.01$. Samples 6 and 8 (open symbols) are superconducting.

much more slowly than T^2 and is nearly independent of the impurity concentration. On a logarithmic plot, $\rho_c(T)$ varies very nearly as $T^{3/2}$. Note that the resistive anisotropies, $\rho_c/\rho_a\approx0.5-2\times10^4$, are significantly higher than in the hole-doped $La_{2-x}Sr_xCuO_{4-y}$ (~160) (Ref. 18) and $YBa_2Cu_3O_{7-\delta}$ (~100) (Ref. 19) compounds, but comparable to highly anisotropic $Bi_2Sr_2CaCu_2O_{8-x}$.[20,21] Also shown in Fig. 2 are data for an over-oxygenated sample (top curve), which typically exhibit localized behavior.

The notably strong anisotropy in temperature dependence and, to a lesser degree, the power-law behavior over such a broad temperature range are difficult to reconcile with electron-phonon scattering. Moreover, in previous work for $\rho_\parallel$,[7] realistic values of the electron-phonon spectral density $\alpha^2F(\omega)$ and the coupling constant λ could be stretched only far enough to explain a relatively small contribution from electron-phonon scattering; any more constrained estimates suggest that such contributions are nearly negligible. We feel the oft-promoted[2,6,7] electron-electron contribution in its simple form is an even more unlikely explanation, as the magnitude observed here can be 10^5-10^6 times that ordinarily expected.[9] Theories incorporating itinerant magnetism provide a plausible mechanism for a large enhancement of electron-electron scattering,[22,23] but such a model[4] permits some five free parameters for the in-plane scattering alone; such complexity is hardly justifiable in light of the simple behavior observed here.

Instead, we opt for a simple anisotropic transport model in terms of scattering of electrons on a nearly cylindrical Fermi surface by anisotropic spin fluctuations. Evidence for nearly cylindrical Fermi surfaces in other copper oxide compounds is strong;[24–26] band-structure calculations for $Nd_{2-x}Ce_xCuO_{4-y}$ (Ref. 27) indicate a single free-electron-like antibonding band at the Fermi level. Similarly, Hall coefficient measurements[11] indicate that a weakly correlated electron-like band dominates the transport. That such electrons could be scattered by magnetic fluctuations is quite apparent: Raman and other spectroscopies[28–30] have shown that, even at high doping in the metallic state, the local moment character of the Cu^{2+} ions is maintained, and the nearest-neighbor exchange coupling $J_\parallel\approx980$ K persists. Also, neutron-scattering experiments[30–32] indicate that AFM spin correlation lengths are quite long even well above the Néel temperature.

The standard variational expression for the resistivity is[33]

$$\rho(T)=\frac{\int d^3k\int d^3k'|\Phi_{\mathbf{k}}-\Phi_{\mathbf{k}'}|^2\mathcal{P}_{\mathbf{k}\mathbf{k}'}}{k_BT|\int d^3k e\mathbf{v}_{\mathbf{k}}\Phi_{\mathbf{k}}(\partial f^0_{\mathbf{k}}/\partial\varepsilon_{\mathbf{k}})|^2}\ , \tag{1}$$

where $\Phi_{\mathbf{k}}$ is the trial variational function, $\mathbf{v}_{\mathbf{k}}$ and $\varepsilon_{\mathbf{k}}$ are the velocity and energy of an electron with wave vector $\mathbf{k}$, and $f^0_{\mathbf{k}}$ is the equilibrium Fermi-distribution function, $f^0_{\mathbf{k}}=(e^{(\varepsilon_{\mathbf{k}}-\mu)/k_BT}+1)^{-1}$. The scattering probability per unit time per unit $\mathbf{k}$-space volume is

$$\mathcal{P}_{\mathbf{k}\mathbf{k}'}=\frac{2\pi}{\hbar}\frac{V_0}{(2\pi)^3}|G_{\mathbf{k}\mathbf{k}'}|^2 f^0_{\mathbf{k}}(1-f^0_{\mathbf{k}'})n_{\mathbf{q}}\delta(\varepsilon_{\mathbf{k}}+\hbar\omega_{\mathbf{q}}-\varepsilon_{\mathbf{k}'}) , \tag{2}$$

where $\mathbf{q}=\mathbf{k}-\mathbf{k}'$, $V_0=a^2c$ is the unit cell volume, and $n_{\mathbf{q}}$ is the Bose-Einstein distribution function, $n_{\mathbf{q}}=(e^{\hbar\omega_{\mathbf{q}}/k_BT}-1)^{-1}$. The dispersion of $\omega_{\mathbf{q}}$ is discussed below. For the scattering matrix element, $|G_{\mathbf{k}\mathbf{k}'}|$, a dominant feature can be factored out: as a result of a Bogoliubov transformation, the antiferromagnetic magnon operators from spin-wave theory have a $1/\sqrt{\omega}$ dependence ($\sim\sqrt{2J_\parallel/\hbar\omega}$).[34] Thus, it is appropriate to define

$$|G_{\mathbf{k}\mathbf{k}'}|^2=\frac{1}{\omega_{\mathbf{q}}}|g_{\mathbf{k}\mathbf{k}'}|^2 . \tag{3}$$

We assume a simple effective-mass approximation, $\varepsilon_{\mathbf{k}}=\hbar^2(k_x^2+k_y^2)/2m_a+\hbar^2k_z^2/2m_c$ and define k_F by $\varepsilon_F\equiv\hbar^2k_F^2/2m_a$, where ε_F is the Fermi energy. With a nearly cylindrical Fermi-surface cutoff at the Brillouin zone boundary, $k_z=\pm\pi/c$, it is straightforward to calculate the resistivity components from Eqs. (1)–(3).[35] For $m_c\gg m_a$, one finds

$$\rho_\alpha=\rho_{0\alpha}+\frac{a^2c^3C_\alpha}{4\pi^5k_BTe^2\hbar^2}\int_0^{2k_F}dq_\parallel\int_0^{2\pi/c}dq_\perp\frac{[(2k_F)^2-q_\parallel^2]^{-1/2}[(2\pi/c)-q_\perp]q_\alpha^2|g_{\mathbf{k}\mathbf{k}'}|^2}{[\exp(\hbar\omega_{\mathbf{q}}/k_BT)-1][1-\exp(-\hbar\omega_{\mathbf{q}}/k_BT)]} , \tag{4}$$

where $\alpha=\parallel$ or $\perp$. Here, the $\rho_{0\alpha}$'s are the residual resistivity components; for an isotropic impurity relaxation time, one finds $\rho_{0\perp}=(m_c/m_a)(C_\perp/2C_\parallel)^{1/2}\rho_{0\parallel}$. The coefficients C_α are $C_\parallel=m_a^2/k_F^4$ and $C_\perp=9m_c^2/2(\pi/c)^4$.

Equation (4) is valid for scattering by any boson satisfying Eq. (3). However, we find that the data of Figs. 1 and 2 can only be fit if one considers an *extremely* anisotropic dispersion for $\omega_{\mathbf{q}}$. We thus consider an antiferromagnetic magnon dispersion, for which a great deal of information is available. Numerous theoretical[36–39] and experimental[30–32,39–42] studies indicate that the spin-$\frac{1}{2}$ Heisenberg Hamiltonian captures the basic physics of the magnetism of the insulating copper oxide parent compounds. Above the Néel temperature, the dominant in-plane exchange coupling $J_\parallel$ produces correlated 2D regions; the long-range 3D transition is driven by much weaker interplanar coupling $J_\perp$ or in-plane anisotropy J_{xy}.[36–38] For simplicity, we consider only the interplanar coupling $J_\perp$, and thus consider the two magnon branches (defining $\alpha_\perp\equiv J_\perp/J_\parallel$)

$$\omega_{\mathbf{q}\pm}=\omega_\parallel[(1+\tfrac{1}{2}\alpha_\perp)^2-(\gamma_\parallel\pm\tfrac{1}{2}\alpha_\perp\gamma_\perp)^2]^{1/2} , \tag{5}$$

where $\gamma_\parallel=\frac{1}{2}[\cos(q_xa)+\cos(q_ya)]$ and $\gamma_\perp=\cos(q_zc)$, similar to Ref. 42 (an erroneous factor of $\frac{1}{2}$ in the definition of $\gamma_\perp$ in Ref. 42 has been omitted).

For $\omega_\parallel$, one can correct the bare relation $\omega_\parallel=2J_\parallel/\hbar$ in two ways. First, corrections for quantum fluctuations of approximately 16% have been calculated by several techniques,[36,43] so one should set $\omega_\parallel\approx2.32J_\parallel/\hbar$ for the parent compounds. One expects a reduction of the spin-wave stiffness constant ρ_s due to dilution; e.g., from the calculations of Ref. 44, $\rho_s/\rho_{s0}=0.49$ for $x=0.18$. However, such a consideration does not change the quality of the fits; since Raman data indicate that dilution broadens the spectrum without noticeable softening, we simply set $\omega_\parallel=2.32J_\parallel/\hbar$ with $J_\parallel=980$ K. Also note that for short-range and small-q interactions (dominant, due to population effects), $|g_{\mathbf{k}\mathbf{k}'}|^2$ approaches an isotropic constant; we thus approximate $|g_{\mathbf{k}\mathbf{k}'}|^2\approx|g_0|^2$ in Eq. (4).

The solid lines in Figs. 1 and 2 are the least-squares fits to Eq. (4) using the dispersion relation of Eq. (5). For each sample, both components ρ_a and ρ_c were fit *simultaneously*. With $J_\parallel$ fixed, the only adjustable parameters were ρ_{0a}, m_c/m_a, k_F, $J_\perp$, and the product $m_a|g_0|$. Since the temperature-independent impurity terms essentially determine two parameters, the *fits to the full temperature dependence of both components depend only on the remaining three parameters.* As such, the quality of the fits to the simple physical model is evidently both remarkable and quite appealing.

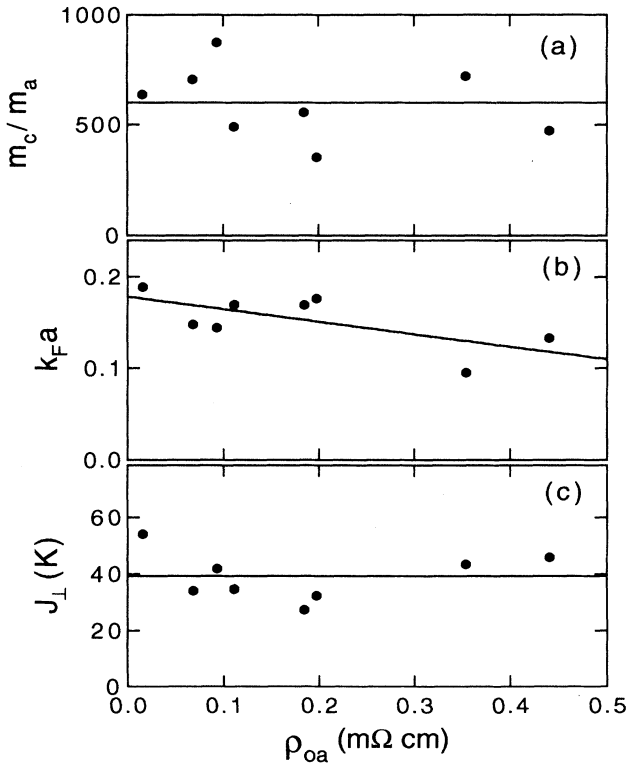

FIG. 3. Extracted values of the effective-mass ratio, m_c/m_a, the Fermi wave vector, k_F, and the effective interplanar exchange constant, $J_\perp$, as functions of the in-plane residual (impurity) resistivity. Solid lines are guides to the eye.

In Figs. 3(a)–3(c), we have plotted the extracted values of the parameters m_c/m_a, $k_F a$, and $J_\perp$ as functions of the residual in-plane resistivity, ρ_{0a}. The effective-mass ratio in Fig. 3(a) ranges between $\sim 400-800$, with appreciable scatter. The effective Fermi wave vector, $k_F \approx 0.2/a$, is quite small; k_F is seen to decrease slightly with increasing impurity concentration; such behavior is plausible if one considers that defects, particularly oxygen interstitials, are believed to trap electrons and decrease the conduction electron density. In Fig. 3(c), the average value of the effective interplanar coupling is $J_\perp = 39$ K. We note that this is much larger than estimated values for the parent compound, where the condition for the 3D transition, $J_\perp (M/M_0)^2 (\xi_{2D}/a)^2 \approx k_B T_N$,[36] has often been used to estimate $J_{\perp(x=0)} \approx 0.1$ K. The large values of $J_\perp$ extracted here may indicate that the doped carriers increase the interplanar coupling significantly, or may be an artifact of the simplified model (e.g., neglecting J_{xy} or other dilution effects[45] on ω_q). Upon Ce doping, the c-axis lattice constant shrinks significantly, a feature which may augment the increased interplanar coupling in the metallic state (in a pressure-dependent study on La_2CuO_4,[46] the calculated $J_\perp$ increased a factor of 20 with an $\sim 1\%$ decrease in the c-axis lattice constant). Finally, recalling the form of Eq. (3), we report the average value of $|g_0'| \equiv \sqrt{\hbar/2J_\parallel}(m_a/m)|g_0| = 180$ meV; we know of no calculation for comparison.

In summary, we have presented measurements and analysis of the normal-state anisotropic resistivity of $Nd_{2-x}Ce_xCuO_{4-y}$ single crystals; we invoked a simple model of scattering by anisotropic spin fluctuations. The model simultaneously describes in-plane and out-of-plane components, as well as the departure from T^2 behavior for high resistivity specimens. The full temperature dependence is describable by very few parameters; a variation in the small $k_F \approx 0.2/a$ with impurity level was noted, as were typical values of $m_c/m_a \approx 600$, $J_\perp \approx 39$ K, and $|g_0'| \approx 180$ meV. This work may lend support to models of magnetic scattering in hole-doped compounds, although there the spin-correlation lengths are so short that the data do not lend themselves to such simple analysis.

This work was supported by the National Science Foundation under Grant No. DMR-9158089 and the Robert A. Welch Foundation under Grant No. F-1191. We gratefully acknowledge useful conversations with Q. Niu and M. Marder.

[1] P. B. Allen, W. E. Pickett, and H. Krakauer, Phys. Rev. B **37**, 7482 (1988).
[2] A. Virosztek and J. Ruvalds, Phys. Rev. B **42**, 4064 (1990).
[3] Q. Si and K. Levin, Phys. Rev. B **44**, 4727 (1991).
[4] T. Moriya, Y. Takahashi, and K. Ueda, J. Phys. Soc. Jpn. **59**, 2905 (1990).
[5] Y. Hidaka and M. Suzuki, Nature (London) **338**, 635 (1989).
[6] C. C. Tsuei, A. Gupta, and G. Koren, Physica C **161**, 415 (1989).
[7] N. Tralshawala *et al.*, Phys. Rev. B **44**, 12 102 (1991).
[8] M. A. Crusellas *et al.*, Physica C **180**, 313 (1991).
[9] For a review, see M. Kaveh and N. Weber, Adv. Phys. **33**, 257 (1984).
[10] C. C. Almasan and M. B. Maple, in *Chemistry of High Temperature Superconductors,* edited by C. N. R. Rao (World Scientific, Singapore, 1991), pp. 205–242.
[11] Z. Z. Wang *et al.*, Phys. Rev. B **43**, 3020 (1991).
[12] See, e.g., R. J. Birgeneau and G. Shirane, in *Physical Properties of High Temperature Superconductors I,* edited by D. M. Ginsberg (World Scientific, Singapore, 1989), pp. 150–211.
[13] G. M. Luke *et al.*, Phys. Rev. B **42**, 7981 (1990).
[14] M. Matsuda *et al.*, Phys. Rev. B **45**, 12 548 (1992).
[15] H. C. Montgomery, J. Appl. Phys. **42**, 2971 (1971).
[16] S. J. Hagen *et al.*, Phys. Rev. B **43** 13 606 (1991).
[17] S. Piñol *et al.*, Physica C **165**, 265 (1990).
[18] T. Kimura *et al.*, Physica C **192**, 247 (1992).
[19] T. A. Friedman *et al.*, Phys. Rev. B **42**, 6217 (1990).
[20] S. Martin *et al.*, Phys. Rev. B **41**, 846 (1990).
[21] X.-D. Xiang *et al.*, Phys. Rev. Lett. **68**, 530 (1992).
[22] T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, Berlin, 1985).
[23] A. J. Schindler and M. J. Rice, Phys. Rev. **164**, 759 (1967).
[24] C. G. Olson *et al.*, Phys. Rev. B **42**, 381 (1990).
[25] H. Haghighi *et al.*, Phys. Rev. Lett. **67**, 382 (1991).
[26] C. M. Fowler *et al.*, Phys. Rev. Lett. **68**, 534 (1992).
[27] S. Massida *et al.*, Physica C **157**, 571 (1989).
[28] I. Tomeno *et al.*, Phys. Rev. B **43**, 3009 (1991).
[29] S. Sugai and Y. Hidaka, Phys. Rev. B **44**, 809 (1991).
[30] R. J. Birgeneau *et al.*, Phys. Rev. B **38**, 6614 (1988).
[31] M. Matsuda *et al.*, Phys. Rev. B **42**, 10 098 (1990).
[32] T. R. Thurston *et al.*, Phys. Rev. Lett. **65**, 263 (1990).
[33] See, e.g., J. M. Ziman, *Electrons and Phonons* (Oxford University Press, London, 1964), Chap. IX.
[34] See e.g., D. C. Mattis, *The Theory of Magnetism I* (Springer-Verlag, Berlin, 1981), Chap. 5.
[35] B.-H. O, Q. Niu, and J. T. Markert (unpublished).
[36] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **39**, 2344 (1989).
[37] C. M. Soukoulis, S. Datta, and Y. H. Lee, Phys. Rev. B **44**, 446 (1991).
[38] H.-Q. Ding, Phys. Rev. Lett. **68**, 1927 (1992).
[39] S. M. Hayden *et al.*, Phys. Rev. B **42**, 10 220 (1990).
[40] S. M. Hayden *et al.*, Phys. Rev. Lett. **67**, 3622 (1991).
[41] Y. Endoh *et al.*, Phys. Rev. B **37**, 7443 (1988).
[42] B. Keimer *et al.*, Phys. Rev. B **45**, 7430 (1992).
[43] A. Auerbach and D. P. Arovas, Phys. Rev. Lett. **61**, 617 (1988).
[44] A. B. Harris and S. Kirkpatrick, Phys. Rev. B **16**, 542 (1977).
[45] W. J. L. Buyers, D. E. Pepper, and R. J. Elliot, J. Phys. C **6**, 1933 (1973).
[46] M. C. Aronson *et al.*, Phys. Rev. B **44**, 4657 (1991).