Toward a Unified Magnetic Phase Diagram of the Cuprate Superconductors

Alexander Sokol[1,2,3] and David Pines[1]
[1] *Department of Physics, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801-3080*
[2] *Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801-3080*
[3] *L.D. Landau Institute for Theoretical Physics, Moscow, Russia*
(Received 4 June 1993)

We propose a unified magnetic phase diagram of the cuprate superconductors. A new feature of this phase diagram is a broad intermediate doping region of quantum-critical, $z=1$, behavior, characterized by temperature independent T_1T/T_{2G} and linear T_1T, where the high energy spin waves are not overdamped. The spin gap in the moderately doped materials is related to the suppression of the spectral weight for frequencies smaller than $\Delta_\xi = c/\xi$ in the quantum disordered, $z=1$, regime. The crossover to the $z=2$ regime, where $T_1T/T_{2G}^2 \simeq \text{const}$, occurs only in the fully doped materials.

PACS numbers: 74.20.Mn, 75.10.Jm, 75.40.Gb, 75.50.Ee

Recent measurements [1–4] of the spin-echo decay rate, $1/T_{2G}$, for a number of cuprate oxides, taken together with earlier measurements of spin-lattice relaxation rate, $1/T_1$, provide considerable insight into their low frequency spin dynamics. In this Letter we show how these measurements may be combined with straightforward scaling arguments to demonstrate the remarkable universality of the low frequency magnetic behavior at high temperatures. This enables us to obtain a unified magnetic phase diagram for the Y- and La-based systems. In the presence of strong antiferromagnetic correlations at either a commensurate or incommensurate wave vector $\mathbf{Q}$, the main contribution to both T_{2G}^{-1} [5] and T_1^{-1} (see [6]) for copper comes from small $\widetilde{\mathbf{q}} = \mathbf{q} - \mathbf{Q}$, so that one may write

$$\frac{1}{T_1T} \sim \int d^2\widetilde{\mathbf{q}} \lim_{\omega\to 0} \frac{\chi''(\widetilde{\mathbf{q}},\omega)}{\omega}, \qquad \frac{1}{T_{2G}} \sim \left[\int d^2\widetilde{\mathbf{q}}\, \chi^2(\widetilde{\mathbf{q}},0)\right]^{1/2}, \tag{1}$$

where $\chi(\widetilde{\mathbf{q}},\omega)$ is the electronic spin susceptibility near $\mathbf{Q}$. On making use of quite straightforward scaling arguments, when applicable, one may substitute $\chi(\mathbf{q},\omega) = \xi^{2-\eta}\hat{\chi}(\widetilde{q}\xi,\omega/\bar{\omega})$ into Eq. (1) ($-\eta$ is the scaling dimension of the real space spin correlator) and obtain

$$\frac{1}{T_1} \sim T\xi^{-\eta}\bar{\omega}^{-1}, \quad \frac{1}{T_{2G}} \sim \xi^{1-\eta}, \quad \frac{T_1T}{T_{2G}} \sim \xi\bar{\omega}, \tag{2}$$

where ξ is the correlation length and $\bar{\omega}$ an appropriate energy scale.

Applicable scaling regimes.—We consider first an antiferromagnetic insulator described by the $S=1/2$ Heisenberg model with the exchange coupling J. Because the spin stiffness, $\rho_s \simeq 0.18J$ [7], is small compared to J, the *quantum-critical* (QC) scaling regime [8], where the only energy scale is set by temperature, $\bar{\omega} \sim T$, exists over a substantial temperature range above $T \simeq 2\rho_s$ [9,10]. The dynamical exponent, z, which relates the characteristic energy and length scales according to $\bar{\omega} \sim \xi^{-z}$, is $z=1$ as a consequence of Lorentz invariance at $T=0$, reflected in the linear dispersion relation of the spin waves. In this case, $T_1T/T_{2G} \sim \bar{\omega}\xi \sim \xi^{1-z} \simeq \text{const}$; when unaffected by the lattice corrections, the magnitude of T_1T/T_{2G} is proportional to the spin wave velocity, c. Since $\xi \sim \bar{\omega}^{-1/z} \sim T^{-1/z} \sim T^{-1}$, one further obtains $1/T_1 \sim T\xi^{z-\eta} \sim T^{\eta}$ [10] and $1/T_{2G} \sim \xi^{1-\eta} \sim T^{\eta-1}$; because the critical exponent η is negligible, $1/T_1 \simeq \text{const}$ (see a related discussion in Ref. [6]), while $1/T_{2G} \sim T^{-1}$. For the purpose of experimental comparisons, the next to leading terms, which depend on ρ_s/T [10,11], can be treated as nonzero intercept in both T_1T and T_{2G}. The numerical results of Ref. [12] show, however, that temperature dependent corrections to the ratio T_1T/T_{2G} are anomalously small.

A second regime of interest is the two-dimensional *renormalized classical* (RC) regime, $T_N \lesssim T \lesssim 2\rho_s$, which is characterized by an exponential increase of the correlation length and relaxation rates [8]. In this case, $1/T_1 \sim T^{3/2}\exp(2\pi\rho_s/T)$ [6] and $1/T_{2G} \sim T\exp(2\pi\rho_s/T)$. The prefactors arising from the $\ln\xi$ corrections [8] lead to a power-law temperature dependence of the ratio $T_1T/T_{2G} \sim T^{1/2}$, while $z=1$ leads to the cancellation of the leading (exponential) terms.

According to numerical calculations for the insulator in the 2D $S=1/2$ Heisenberg model [12,13], as long as $T \lesssim J$, the damping, γ_q, of the high energy ($\omega_q \gtrsim c/\xi$) spin wave excitations, is small throughout the Brillouin zone; hence, for both RC and QC regimes those can be treated as good eigenstates of the model. The dynamical susceptibility can then be well approximated as

$$\chi(\mathbf{q},\omega) = \phi_q\left(\frac{1}{\omega+\omega_q+i\gamma_q} - \frac{1}{\omega-\omega_q+i\gamma_q}\right), \tag{3}$$

except near the origin, where the dynamics is diffusive as a consequence of total spin conservation, and near the

0031-9007/93/71(17)/2813(4)$06.00

Néel ordering vector, $(\pi/a, \pi/a)$, where it is relaxational; for $\xi^{-1} \lesssim \tilde{q} \lesssim a^{-1}$, the expression (3) is valid in both QC and RC regimes, where $\phi_q \sim 1/\tilde{q}$ and $\omega_q \simeq c\tilde{q}$. According to Ref. [14], the one-magnon neutron scattering intensity in the insulator is indeed well described by Eq. (3) with $\gamma_q \ll \omega_q$.

It has been conjectured in Refs. [10] that the small doping as well as randomness related to it are not likely to affect the scaling behavior at high temperatures. Quite generally, one would expect a departure from $z=1$ behavior only when spin waves become overdamped [15]. Since this would require a substantial increase in spin wave damping, to $\gamma_q > \omega_q$, compared to its insulator value, $\gamma_q \ll \omega_q$, there may be an intermediate regime in which the spin waves coexist with the electron-hole continuum, even if their damping is increased compared to the insulator. To the extent this occurs, the system can remain in the QC regime with $z=1$ in a wide range of doping and temperatures due to the Lorentz invariant terms in the action.

In the insulator, the zero temperature energy gap $\Delta = \hbar c/\xi$ for the spin-1 excitations in a quantum disordered (QD) regime is, again, related to the Lorentz invariance at $T=0$ [8] (note that QD regime is not related to quenched disorder). Hence, as long as the Lorentz invariant terms in the action are still important and the correlation length saturates, the low frequency ($\omega < \Delta$) spectral weight could be suppressed even in a metal. We suggest that the *spin gap* phenomenon in $YBa_2Cu_3O_{6.63}$ and $YBa_2Cu_4O_8$, characterized by a sharp increase in T_1T and decrease of the bulk susceptibility, is related to this suppression, and this phase corresponds to a QD, $z=1$, regime.

In the opposite limit of large doping, the spin waves are replaced by relaxational modes. In this *overdamped* (OD) regime, the self-consistent renormalization approach developed by Moriya *et al.* [16] (see also Ref. [15]) and the phenomenological theory of Millis *et al.* [17], for $YBa_2Cu_3O_7$ may be expected to apply. The dynamical exponent $z=2$, while $\eta=0$; in contrast with the previous case, the mean field exponent $z=2$ is not fixed by a symmetry, but rather follows from the scaling analysis of Refs. [15,16]. One obtains $T_1T/T_{2G}^2 \sim \bar{\omega}\xi^2 \sim \xi^{2-z} \simeq \text{const}$, while $T_1T/T_{2G} \sim \xi^{-1}$. At high temperatures, the energy scale $\bar{\omega} \sim T$, so that $\xi \sim \bar{\omega}^{-1/z} \sim T^{-1/z} \sim T^{-1/2}$, in which case $T_1T/T_{2G} \sim T^{1/2}$, and, separately, $1/T_1 \sim T\xi^z \simeq$ const, $1/T_{2G} \sim \xi \sim T^{-1/2}$. We emphasize that $1/T_1 \simeq$ const at high temperatures is predicted for both $z=1$ and $z=2$ regimes, while the predictions for $1/T_{2G}$ are different. Finally, at still larger hole densities, the short range antiferromagnetic correlations between spins will tend to disappear; in this limit, $\xi \lesssim a$ is independent of temperature, and one recovers the Korringa law, $1/T_1 \sim T$, while $1/T_{2G} \simeq$ const. This regime corresponds to a normal metal, in which any remaining antiferromagnetic correlations can be described by a temperature independent $F^a(\mathbf{p}, \mathbf{p}')$.

$La_{2-x}Sr_xCuO_4$.—Above T_N, the insulating parent compound, La_2CuO_4, is well described by the 2D Heisenberg model with the nearest-neighbor exchange coupling $J \simeq 1500$ K [18]. A nearly temperature independent $1/T_1$ is observed in the insulating La_2CuO_4 above 650 K [2], as expected in the QC, $z=1$, regime [6,10]. The absolute value of $1/T_1 \simeq 2700$ sec^{-1} at high temperatures [2] is in very good agreement with both $1/N$ expansion [10] and finite cluster [12] calculations for the Heisenberg model. Further, the ratio T_1T/T_{2G} measured in the insulator [2,4] is nearly temperature independent in the broad range $450\,\text{K} < T < 900\,\text{K}$ (Fig. 1), as is expected in the QC, $z=1$, regime.

A finite cluster calculation in the $S=1/2$ 2D Heisenberg model [12] with no adjustable parameters used (hyperfine and exchange couplings were determined from other experiments, see Ref. [12]) indeed yields a nearly temperature independent $T_1T/T_{2G} \simeq 4.3 \times 10^3$ K^{-1} for $T > J/2 \simeq 750$ K (Fig. 1), in excellent agreement with the experimental result, 4.5×10^3 K [2].

An especially striking feature of the $1/T_1$ data [2] is the nearly doping independent absolute value of $1/T_1$ in the high temperature limit (Fig. 1) for $x=0-0.15$, i.e., up to the *optimal* concentration for the superconductivity. Since it would be rather unrealistic to assume that exactly the same value can be obtained in different pictures of magnetism for low- and high-doped La-based materials, we suggest that the high temperature magnetic behavior over the doping range in which metallic behavior is found above T_c, $7.5\% \lesssim x \lesssim 15\%$, has the same physical origin as that found for the insulating state, which im-

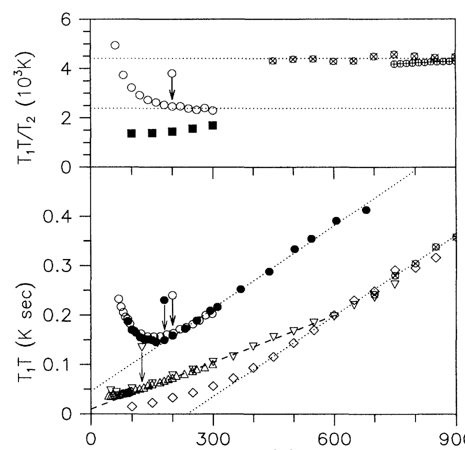

FIG. 1. Experimental data on T_1T and T_1T/T_{2G}: ⊗, La_2CuO_4 [2,4]; ◇, $La_{1.96}Sr_{0.04}CuO_4$ [2]; ∇, $La_{1.85}Sr_{0.15}CuO_4$ [2]; Δ, $La_{1.85}Sr_{0.15}CuO_4$ [26]; •, $YBa_2Cu_4O_8$ [27]; ∘, $YBa_2Cu_3O_{6.63}$ [3,20]; ■, $YBa_2Cu_3O_{6.9}$ [1]. For La_2CuO_4, only the data in the QC regime are shown. Also shown (⊕) are the results of numerical calculation of $1/T_1$ [12] and T_1T/T_{2G} ([12] and this work) for the insulator. The arrows indicate our proposed values for the crossover temperature from the QC to QD regimes.

plies $z=1$. In this case, a nearly doping independent and linear in temperature T_1T follows [10] directly from the Josephson scaling (Fig. 1). While the neutron scattering experiments [19] show incommensurate peak positions in $\chi''(\mathbf{q},\omega)$, moderate incommensurability is not likely to influence the high energy spin wave spectrum and thus the nuclear relaxation at $T\sim 900$ K. We thus conclude that at high temperatures $La_{1.85}Sr_{0.15}CuO_4$ is in the QC, $z=1$, regime.

For lower temperatures, T_1T in $La_{2-x}Sr_xCuO_4$ begins to deviate from its doping independent linear behavior (Fig. 1) with a crossover temperature which is roughly proportional to $x^{1/2}$. We conjecture that only below this crossover, quenched disorder, or a modification of the spin wave spectrum due to incommensurability, become important. As it is evident from Fig. 1, for $T \lesssim 125$ K, T_1T in $La_{1.85}Sr_{0.15}CuO_4$ begins to depart from linear temperature dependence observed for 125 K$< T <$ 600 K, exhibiting an upturn for $T \approx$60–70 K. We attribute this effect to a crossover to the quantum disordered regime, in which the low frequency spectral weight for the spin waves is suppressed.

$YBa_2Cu_3O_{6.63}$ and $YBa_2Cu_4O_8$ have quite similar properties. The product T_1T measured in $YBa_2Cu_3O_{6.63}$ [3,20] is linear in temperature for 160 K$\lesssim T \lesssim$ 300 K, while it exhibits similar behavior in $YBa_2Cu_4O_8$ for 170 K$\lesssim T \lesssim$800 K. Since a linear T_1T is predicted in both quantum-critical ($z=1$) and overdamped ($z=2$) regimes at high temperatures, to distinguish between these regimes, we turn to the $1/T_{2G}$ data on $YBa_2Cu_3O_{6.63}$ [3], and plot T_1T/T_{2G} and T_1T/T_{2G} as a function of temperature (Fig. 2). In the range 200 K $< T <$ 300 K, T_1T/T_{2G} is nearly constant, while T_1T/T_{2G}^2 varies significantly, in agreement with the prediction for $z=1$. Were this material in the $z=2$ regime, T_1T/T_{2G} would increase as the temperature increases, while T_1T/T_{2G}^2 would be constant. We thus conclude that above 200 K, $YBa_2Cu_3O_{6.63}$, and the closely related $YBa_2Cu_4O_8$, are in the QC, $z=1$, regime.

For temperatures below 150 K, $1/T_1$ sharply drops down as the temperature decreases, while $1/T_{2G}$ [3,20] saturates. We argue that such a suppression of the low frequency spectral weight (*spin gap*) reflects a crossover to the quantum disordered, $z=1$, regime. Thus, unlike the scenario proposed by Millis and Monien [21], we argue that for all three materials the primary origin of the suppression of the low energy spin fluctuation spectral weight is the same. We also note that universal dependence of the neutron scattering intensity on ω/T observed in $YBa_2Cu_3O_{6.6}$ at high temperatures [22] is expected for the QC scaling.

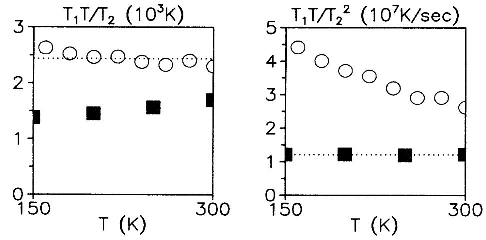

FIG. 2. Experimental data on T_1T/T_{2G} and T_1T/T_{2G}^2: ○, $YBa_2Cu_3O_{6.63}$ [3,20]; ■, $YBa_2Cu_3O_{6.9}$ [1].

$YBa_2Cu_3O_{6.9}$.—For nearly stoichiometric $YBa_2Cu_3O_{6.9}$, T_1T/T_{2G} [1] somewhat increases as the temperature increases, while T_1T/T_{2G}^2 remains constant (Fig. 2), in agreement with the one-loop scaling prediction for the *overdamped*, $z=2$, regime [16]. The departure from the Korringa law $1/T_1 \sim T$ and large copper-to-oxygen ratio of the relaxation rates shows that the antiferromagnetic enhancement is still quite substantial. In the overdamped regime, the spin wave branch is either destroyed, or due to the small correlation length, has appreciable spectral weight only for energies much larger than the maximal temperature at which this compound is chemically stable. Therefore, for experimentally accessible temperatures no departure from the overdamped regime is observed.

In conclusion, we have shown that the nuclear relaxation data in a broad range of doping levels, which includes both metallic and insulating materials, possesses features characteristic of the quantum critical regime of a clean antiferromagnetic insulator with the dynamical exponent $z=1$. This behavior suggests that even for substantial doping, such as that in $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_{6.63}$, and $YBa_2Cu_4O_8$, the spin waves are not overdamped. While such magnetic properties may arise in a one-component as well as a two-component *microscopic model*, we call attention to an explicit example which leads to this kind of dynamics directly, namely, a model of spins and fermions with both spin-spin (J) and weak spin-fermion ($\tilde{J}$) exchange interaction [23]. On the other hand, recent numerical results [24] indicate the presence of a broad region of QC, $z=1$, behavior in a one band description given by the moderately doped t-J model.

On the basis of the above analysis, we suggest the unified magnetic phase diagram for the cuprate superconductors shown in Fig. 3; the proposed boundary between the QC and QD regimes is determined from the nuclear relaxation data shown in Fig. 1. We propose that as the hole doping increases, the transition from the insulating to the overdoped regime occurs in two stages. First, the system becomes metallic; the damping of spin waves increases somewhat compared to its value in the insulator, but since the spin waves remain propagating modes, the dynamical exponent is $z=1$, and the quantum critical regime persists over a wide range of temperatures and doping levels. Then, at substantially higher doping, the spin fluctuations acquire purely relaxational character and the dynamical exponent crosses over to $z=2$. Thus, in the underdoped materials, there exist quasiparticle excitations (the charge degrees of freedom) coupled to spin waves. At high temperatures, the spin fluctua-

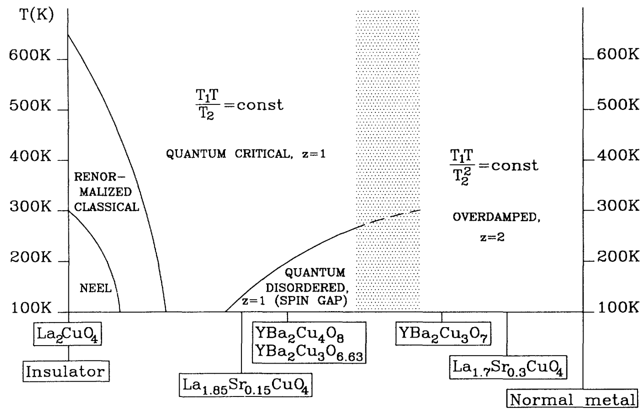

FIG. 3. The proposed magnetic phase diagram for the cuprate superconductors above 100 K.

tions of the coupled system can be described by the nonlinear sigma model with renormalized parameters which depend on doping. The charge response of the system is determined by the quasiparticles, whose properties (resistance, etc.) reflect their coupling to the spin fluctuations.

We further argue that the spin gap phenomenon observed in the underdoped materials reflects the same physics as the formation of the gap for spin waves in the the quantum disordered, $z=1$, phase of a nearly critical clean antiferromagnetic insulator. This scenario suggests that in compounds where the spin gap is observed, at high temperatures both the bulk susceptibility (see Ref. [10]) and the resistivity should be linear in temperature and T_1T/T_{2G} should be temperature independent. We show, in a subsequent paper [25], that our scenario leads in a natural way to a unified description of the results of nuclear relaxation, magnetic susceptibility, and neutron scattering experiments.

We are grateful to M. Takigawa, T. Imai, and C.P. Slichter for communicating their experimental data to us in advance of publication. We would like to thank G. Aeppli, V. Barzykin, G. Blumberg, S. Chakravarty, A.V. Chubukov, S.L. Cooper, E. Dagotto, D. Frenkel, L.P. Gor'kov, M.V. Klein, A.J. Millis, R.R.P. Singh, and D. Thelen for valuable discussions, and T. Imai and C.P. Slichter for numerous stimulating conversations on nuclear resonance phenomena. This work has been supported by the NSF Grants No. DMR89-20538 through the Materials Research Laboratory and No. DMR91-20000 through the Science and Technology Center for Superconductivity.

[1] T. Imai *et al.*, Phys. Rev. B **47**, 9158 (1993).
[2] T. Imai *et al.*, Phys. Rev. Lett. **70**, 1002 (1993).
[3] M. Takigawa (private communication).
[4] T. Imai *et al.*, Phys. Rev. Lett. **71**, 1254 (1993).
[5] C.H. Pennington and C.P. Slichter, Phys. Rev. Lett. **66**, 381 (1991); see also D. Thelen and D. Pines (to be published).
[6] S. Chakravarty and R. Orbach, Phys. Rev. Lett. **64**, 224 (1990).
[7] R.R.P. Singh, Phys. Rev. B **39**, 9760 (1989).
[8] S. Chakravarty, B.I. Halperin, and D.R. Nelson, Phys. Rev. B **39**, 2344 (1989).
[9] S. Sachdev and J. Ye, Phys. Rev. Lett. **69**, 2411 (1992).
[10] A.V. Chubukov and S. Sachdev, Phys. Rev. Lett. **71**, 169 (1993); A.V. Chubukov, S. Sachdev, and J. Ye (unpublished).
[11] We thank S. Chakravarty for pointing out to us that in the asymptotic QC limit the correction terms can be expanded in powers of $(\rho_s/T)^{1/\nu}$, where the critical exponent $\nu \simeq 0.7$.
[12] A. Sokol, E. Gagliano, and S. Bacci, Phys. Rev. B **47**, 14 646 (1993); (to be published).
[13] M. Makivić and M. Jarrell, Phys. Rev. Lett. **68**, 1770 (1992).
[14] S.M. Hayden *et al.*, Phys. Rev. Lett. **67**, 3622 (1991).
[15] A.J. Millis (to be published).
[16] T. Moriya, Y. Takahashi, and K. Ueda, Phys. Rev. Lett. **59**, 2905 (1990), and references therein.
[17] A.J. Millis, H. Monien, and D. Pines, Phys. Rev. B **42**, 167 (1990).
[18] S. Chakravarty, in *Proceedings of High Temperature Superconductivity*, edited by K.S. Bedell *et al.* (Addison-Wesley, CA, 1990); E. Manousakis, Rev. Mod. Phys. **63**, 1 (1991).
[19] S.-W. Cheong *et al.*, Phys. Rev. Lett. **67**, 1791 (1991).
[20] M. Takigawa *et al.*, Phys. Rev. B **43**, 247 (1991).
[21] A.J. Millis and H. Monien [Phys. Rev. Lett. **70**, 2810 (1993)] have suggested that temperature dependent bulk susceptibility in $YBa_2Cu_3O_{6.63}$ is due to the gap for spin excitations induced by the interplanar coupling, while in $La_{1.85}Sr_{0.15}CuO_4$ it is caused by the SDW fluctuations, in contrast with our in-plane mechanism. The spin gap regime has been discussed in a number of publications; some of them are listed as Ref. [7] of the above paper.
[22] B.J. Sternlieb *et al.*, Phys. Rev. B **47**, 5320 (1993).
[23] J.R. Schrieffer, L. Lilly, and N.E. Bonesteel, in Proceedings of the APS 1993 March Meeting, Abstract No. G23-1 (unpublished); (private communication); see also L.P. Gor'kov and A. Sokol, Physica (Amsterdam) **159C**, 329 (1989).
[24] A. Sokol, R.L. Glenister, and R.R.P. Singh (unpublished).
[25] V. Barzykin *et al.* (unpublished).
[26] Y. Kitaoka *et al.*, Physica (Amsterdam) **189C**, 189 (1990).
[27] T. Machi *et al.*, Physica (Amsterdam) **173C**, 32 (1991).