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Theory of NMR as a local probe for the electronic structure in the mixed state of the high-T_c cuprates

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We argue that nuclear-magnetic-resonance experiments are a site-sensitive probe for the electronic spectrum in the mixed state of the high-T_c cuprates. Within a spin-fermion model, we show that the Doppler-shifted electronic spectrum arising from the circulating supercurrent changes the low-frequency behavior of the imaginary part of the spin susceptibility. For a hexagonal vortex lattice, we predict that these changes lead to (i) a unique dependence of the ^{63}Cu spin lattice relaxation rate, $1/T_1$, on resonance frequency, and (ii) a temperature dependence of T_1 which varies with frequency. We propose a nuclear quadrupole experiment to study the effects of a uniform supercurrent on the electronic structure and predict that T_1 varies with the direction of the supercurrent.

The form of the fermionic excitation spectrum in and around a vortex in the mixed state of high-T_c cuprates has been the subject of intense research in the last few years.[1,2] It has been known for a long time that the supercurrent circulating a vortex gives rise to a Doppler-shift in the fermionic excitation spectrum.[3] For a *d*-wave order parameter, Volovik pointed out[1] that this shift leads to the scaling relation $D(\epsilon_F)\sim\sqrt{H}$, where $D(\epsilon_F)$ is the density of states (DOS) at the Fermi energy, and H is the applied field. The main contribution to the DOS comes from regions close to the nodes, but in real space far from the vortex cores. Specific-heat (SH) measurements by Moler *et al.*[4] on $YBa_2Cu_3O_{6.95}$ confirmed this scaling behavior. However, these experiments only provide information on the sample averaged DOS.

Curro[5] pointed out that the spin-lattice relaxation time, T_1, measured in nuclear-magnetic-resonance (NMR) experiments is a more direct probe of these effects since it can give site specific information on the electronic structure in the vicinity of the vortex. They recognized that (i) the dependence of the resonance frequency on the local field allows one to measure T_1 as a function of the nucleus position in the vortex lattice, and (ii) T_1 should depend on the distance from the vortex core. Therefore, T_1 measurements which are a purely electronic probe, in contrast to SH experiments, which are dominated by phononic contributions, permit a detailed test of any theoretical scenario for the electronic structure in the superconducting state. Experimentally, Curro found for the ^{17}O resonance in $YBa_2Cu_3O_7$ that $1/T_1$ increases with decreasing distance from the vortex core,[5] i.e., with increasing frequency. Preliminary results of Milling and Slichter[7] show a qualitatively similar behavior for the ^{63}Cu relaxation rate.

In this paper we use the spin-fermion model[6] to show that the experimentally observed frequency dependence of T_1 can be qualitatively explained by the spatial variation of the supercurrent, the resulting Doppler shift in the electronic spectrum, and the presence of antiferromagnetic spin fluctuations. We show that the Doppler-shifted electronic spectrum significantly changes the low-frequency behavior of the imaginary part of the spin susceptibility, $\chi''(\mathbf{q},\omega)$. Thus $\chi''(\mathbf{q},\omega)$ changes with the spatially varying supercurrent, and hence $1/T_1$, which is a weighted momentum average of χ'' in the zero-frequency limit, acquires a frequency, i.e., spatial dependence. Within our scenario we predict a temperature dependence of $1/T_1$ which changes uniquely with frequency. Our results thus provide insight into the *local* structure of electronic and magnetic excitations in the mixed state.

In what follows we consider for simplicity a single-layer system; an extension of our results to the case of a double-layer system, i.e., $YBa_2Cu_3O_{6+x}$, is straightforward. The ^{63}Cu spin-lattice relaxation rate, $1/T_1$, for an applied field parallel to the c axis is given by

$$\frac{1}{T_1T}=\frac{k_B}{2\hbar}(\hbar^2\gamma_n\gamma_e)^2\frac{1}{N}\sum_{\mathbf{q}} F_c(\mathbf{q})\lim_{\omega\to 0}\frac{\chi''(\mathbf{q},\omega)}{\omega}, \qquad (1)$$

where

$$F_c(q)=[A_{ab}+2B(\cos(q_x)+\cos(q_y))]^2, \qquad (2)$$

and A_{ab} and B are the on-site and transferred hyperfine coupling constants, respectively.[8,9] In order to describe the effects of a supercurrent on χ'' we use the spin-fermion model[6] in which the damping of antiferromagnetic spin fluctuations is determined by their coupling to planar quasiparticles. Within this model, the spin propagator, χ, is given by

$$\chi^{-1}=\chi_0^{-1}-\Pi, \qquad (3)$$

where χ_0 is the bare propagator, and Π is the bosonic self-energy given by the irreducible particle-hole bubble. It was earlier suggested[10] that Re χ^{-1} should possess a relaxational form such that for $\omega\to 0$

$$\mathrm{Re}\ \chi^{-1}=\chi_0^{-1}-\mathrm{Re}\,\Pi=\frac{\xi_{AF}^{-2}+(\mathbf{q}-\mathbf{Q})^2}{\alpha}, \qquad (4)$$

where ξ_{AF} is the magnetic correlation length, α is a temperature-independent overall constant, and $\mathbf{Q}=(\pi,\pi)$ is the position of the magnetic peak in momentum space which we assume to be commensurate.[11] The description of Re χ^{-1} in Eq. (4) as a relaxational, nondispersing spin mode at low frequencies is in agreement with the analysis of NMR and inelastic neutron-scattering experiments on several $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ compounds.[9,10,12]

We are thus left with the calculation of the imaginary part of Π which describes the spin damping brought about by the decay of a spin excitation into a particle-hole pair. In the superconducting state we find to lowest order in the spin-fermion coupling g[6]

$$-\Pi(\mathbf{q},i\omega_n)=g^2T\sum_{\mathbf{k},m}\{G(\mathbf{k},i\Omega_m)G(\mathbf{k}+\mathbf{q},i\Omega_m+i\omega_n)+F(\mathbf{k},i\Omega_m)F(\mathbf{k}+\mathbf{q},i\Omega_m+i\omega_n)\},\tag{5}$$

where G and F are the normal and anomalous Green's functions. Assuming that the supercurrent momentum, $\mathbf{p}_s$, possesses only a weak spatial dependence, we find in semiclassical approximation[3]

$$G(\mathbf{k},i\Omega_m)=\frac{v_{\mathbf{k}}^2}{i\Omega_m-E_{\mathbf{k}}}+\frac{u_{\mathbf{k}}^2}{i\Omega_m+E_{-\mathbf{k}}};$$

$$F(\mathbf{k},i\Omega_m)=-u_{\mathbf{k}}v_{\mathbf{k}}\left\{\frac{1}{i\Omega_m-E_{\mathbf{k}}}-\frac{1}{i\Omega_m+E_{-\mathbf{k}}}\right\}.\tag{6}$$

Here, $E_{\mathbf{k}}$ is the dispersion of the fermionic quasiparticles (bogoliubons), which up to linear order in p_s is given by[3]

$$E_{\mathbf{k}}=\sqrt{\epsilon_{\mathbf{k}}^2+|\Delta_{\mathbf{k}}|^2}+\mathbf{v}_F(\mathbf{k})\cdot\mathbf{p}_s,\tag{7}$$

where $\epsilon_{\mathbf{k}}$ is the electronic normal-state dispersion and $\mathbf{v}_F(\mathbf{k})=\partial\epsilon_{\mathbf{k}}/\partial\mathbf{k}$. Any changes in the d-wave gap $\Delta_{\mathbf{k}}=\Delta_0[\cos(k_x)-\cos(k_y)]/2$ and the BCS coherence factors $u_{\mathbf{k}},v_{\mathbf{k}}$ due to a supercurrent appear only to order p_s^2 and can thus be neglected.

Since the dominant contribution to Im Π for $\omega\to 0$ comes from quasiparticle excitations in the vicinity of the nodes, we expand $E_{\mathbf{k}}$ around the nodes to linear order in momentum and perform the momentum and frequency integrations in Eq. (5) analytically. Combining the results with Eqs. (1), (3), and (4) we find that the dependence of $1/T_1$ on temperature T and on $\mathbf{p}_s$ is determined by the set $\{D_m/T\}$, where $D_m=\mathbf{v}_F^{(m)}\cdot\mathbf{p}_s$ and $\mathbf{v}_F^{(m)}$ is the Fermi velocity at the node in the mth quadrant of the Brillouin zone. The full expression for $1/T_1$ for arbitrary $\{D_m/T\}$ is too cumbersome to be presented here, however, in the low-temperature limit where $|D_m/T|\gg 1$ for $m=1\cdots 4$, it simplifies and we obtain up to order $(T/D_m)^2$

$$\frac{1}{T_1T}=\frac{\mathcal{C}}{N}\left(\frac{1}{v_Fv_\Delta}\right)^2\sum_{n,m}\left\{\mathcal{F}(\mathbf{q}_{n,m})\left(D_mD_n+\frac{\pi^2T^2}{3}\right)+\mathcal{F}(\mathbf{q}_{n,m+2})\left(D_mD_n-\frac{\pi^2T^2}{3}\right)\right\}+O(e^{D_m/T}),\tag{8}$$

where the sum runs only over those nodes with $D_m,D_n<0$, $\mathcal{C}=(\alpha g)^2k_B(\hbar^2\gamma_n\gamma_e)^2/(2\hbar)$, $v_\Delta=|\partial\Delta_{\mathbf{k}}/\partial\mathbf{k}|$ at the nodes, and

$$\mathcal{F}(\mathbf{q}_{n,m})=\frac{F_c(\mathbf{q}_{n,m})}{(\xi_{AF}^{-2}+(\mathbf{q}_{n,m}-\mathbf{Q})^2)^2}.\tag{9}$$

Here, $\mathbf{q}_{n,m}$ is the momentum connecting nodes n and m. The opposite signs of the T^2 terms on the right-hand side of Eq. (8) arise from the combination of Fermi functions involved in different relaxation processes. While the first term describes a process in which a bogoliubon is simply scattered at the spin-fermion vertex, the second one involves processes in which two bogoliubons are simultaneously created or destroyed at the vertex. Since the dominant contribution to the relaxation rate in the low-temperature regime comes from the second term in Eq. (8), we find the unexpected result that in this limit $1/T_1T$ actually decreases with increasing temperature. Note, that for $T\to 0$ it follows from Eq. (8) that $1/T_1T\sim p_s^2$.

In the high-temperature limit ($|D_m/T|\ll 1$ for all m), we obtain to leading order in $(D_m/T)^2$

$$\frac{1}{T_1T}=\frac{\pi\mathcal{C}}{3N}\left(\frac{T}{v_Fv_\Delta}\right)^2\sum_{n,m}\mathcal{F}(\mathbf{q}_{n,m}),\tag{10}$$

where the sum runs over all nodes. For $T\gg|D_m|$ we thus recover as expected the temperature dependence of the relaxation rate in the absence of a supercurrent.

We now turn to NMR experiments in the mixed state of the high-T_c cuprates which possess a hexagonal vortex lattice. We consider for definiteness $YBa_2Cu_3O_7$, where $v_F\approx 0.4$ eV, $v_\Delta\approx 20$ meV, $A_{ab}/B=0.7$, and $\xi_{AF}\approx 2$ is temperature independent in the superconducting state.[9] Each nucleus in the sample is characterized by a resonance frequency $\Delta\nu(\mathbf{r})=\gamma_n\hbar H_z(\mathbf{r})$ and a supercurrent momentum $\mathbf{p}_s(\mathbf{r})$. Here $H_z(\mathbf{r})$ is the local magnetic field and γ_n is the nuclear gyromagnetic ratio. Since the electronic Zeeman splitting for typical applied fields is of the order 10^{-1} meV, and thus smaller than the Doppler shift for most of the nuclei, it can be neglected.

Our scenario for the calculation of T_1 is only applicable to a given nucleus, if in its vicinity $\Delta(\mathbf{r})$ is uniform and $\mathbf{p}_s(\mathbf{r})$ varies sufficiently slowly. In what follows we consider therefore only those nuclei which satisfy these criteria, i.e., nuclei which are $R>2-3\xi_{ab}$ from the center of the vortex core,[14] where ξ_{ab} is the superconducting in-plane coherence length. The local field, $H_z(\mathbf{r})$, is given by[3]

$$H_z(\mathbf{r})=B\sum_{\mathbf{q}}\frac{\exp(i\mathbf{qr})}{1+\lambda^2q^2},\tag{11}$$

where λ is the magnetic penetration depth, B is the average magnetic field, and $\mathbf{q}$ runs over the reciprocal lattice of the two-dimensional (2D) vortex array. Since nonlocal as well as nonlinear effects can effectively be accounted for by a renormalization of λ,[13] we neglect them in the following. We can then calculate the supercurrent momentum via $\nabla\times\mathbf{H}(\mathbf{r})\sim\mathbf{p}_s(\mathbf{r})$, and thus obtain a spatial relation between $H_z(\mathbf{r})$ and $\mathbf{p}_s(\mathbf{r})$. In Fig. 1 we plot the ^{63}Cu spectrum resulting from Eq. (11) (solid line) and $1/T_1T$ (open squares) as a function of

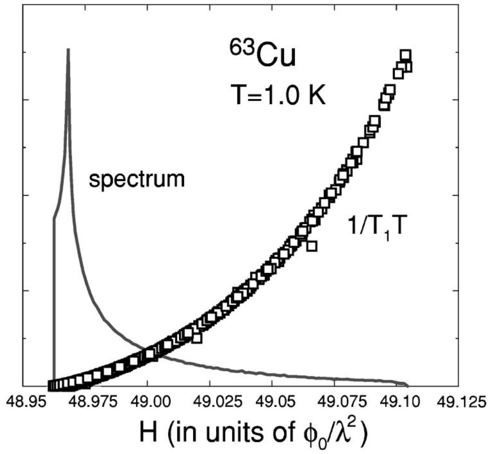

FIG. 1. The theoretical ^{63}Cu spectrum [solid line, Eq. (11)] and $1/T_1T$ (open squares) as a function of the local field for $T=1$ K. Both curves are truncated at a distance $2\xi_{ab}$ from the vortex core.

the local field H_z at $T=1$ K. Nuclei at the highest frequencies are located at a distance $2\xi_{ab}$ from the center of the vortex, nuclei at the lowest frequencies are in the center of a vortex triangle, those at the maximum of the spectrum are at the midpoint between two vortices. For $T=1$ K, the relaxation rate for practically all nuclei is determined by the low-temperature expression, Eq. (8). In this case, $1/T_1T \sim p_s^2$ and hence reflects the frequency dependence of p_s which increases with decreasing distance from the vortex core. The Doppler-shifted fermionic spectrum thus gives rise to a unique dependence of $1/T_1T$ on frequency within the vortex-lattice spectrum of ^{63}Cu. This result is in qualitative agreement with preliminary measurements by Milling and Slichter[7] who find that $1/T_1T$ increases with increasing local field over the whole ^{63}Cu spectrum.

Another unique signature of Doppler-shifted excitations appears in the temperature dependence of the relaxation rate. In Fig. 2 we present $1/T_1T$ as a function of field for three different temperatures (the 30 and 60 K curves are horizontally offset for clarity). For nuclei at the low-field end of the curve, p_s is small, $|D_m/T| \ll 1$ already at low T, and hence $(T_1T)^{-1} \sim T^2$ which increases with increasing temperature. On the other hand, for nuclei closer to the vortex core, and

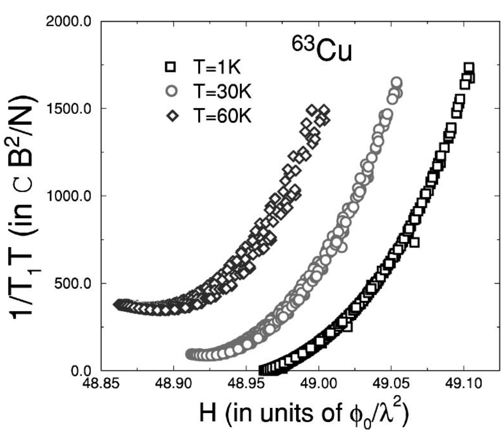

FIG. 2. $(1/T_1T)$ as a function of the local field for three different temperatures. The 30 and 60 K curves are horizontally offset for clarity.

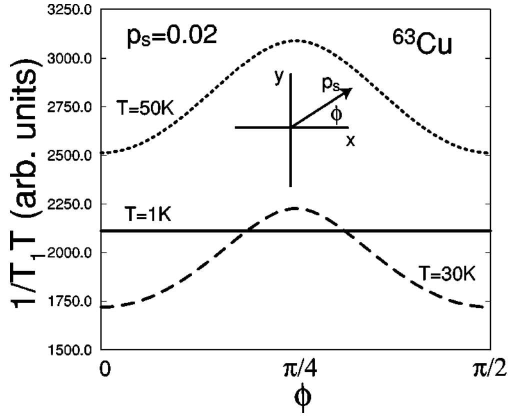

FIG. 3. $1/T_1T$ for $p_s=0.02$ and three different temperatures as a function of the angle ϕ between $\mathbf{p}_s$ and the crystal x axis (see inset).

particularly for those at the high-field end of the curve, p_s is large and $|D_m/T| \gg 1$ even for $T=60$ K (which is still below the lattice melting temperature at $T_m=70$ K). The temperature dependence of $1/T_1T$ is therefore described by Eq. (8), and thus decreases with increasing temperature. Between the low- and high-field end of the spectrum there exists a crossover region, characterized by a minimum in the relaxation rate. This nonmonotonic behavior of $1/T_1T$ as a function of local field arises from two competing contributions: while quasiparticle excitations involving nodes with $D_m>0$ become exponentially suppressed with increasing H_z, the contributions from the remaining (unsuppressed) excitations scale as $\sim p_s^2$ and thus increase. The crossover occurs approximately at those nuclei for which $\max|D_m| \sim O(T)$; the minimum thus shifts towards higher fields, i.e., larger values of $|D_m|$, with increasing temperature, as shown in Fig. 2.

While the absolute scale of our results in Figs. 1 and 2 depends on a number of parameters, it is practically independent of ξ_{AF} for $\xi_{AF}>3$. In contrast, the predicted temperature and frequency dependence of T_1 depends only on $\{D_m/T\}$ and thus presents a generic feature of a Doppler-shifted electronic spectrum due to the distribution of supercurrents in a hexagonal vortex lattice.

An alternative experiment which could observe the effects of a Doppler-shifted fermionic dispersion is a nuclear quadrupole resonance (NQR) measurement in which a uniform supercurrent is applied to the sample. Since in the low-temperature limit, Eq. (8), $1/T_1T$ depends on $D_m = \mathbf{v}_F^{(m)} \cdot \mathbf{p}_s$, with $\mathbf{v}_F^{(m)}$ being fixed by the underlying lattice, one would expect that the relaxation rate varies with the angle between the supercurrent momentum, $\mathbf{p}_s$, and the crystal axes. In Fig. 3 we plot $1/T_1T$, for $|\mathbf{p}_s|=0.02$ as a function of the angle ϕ between $\mathbf{p}_s$ and the crystal x axis (see inset). For $\phi=0$, i.e., for $\mathbf{p}_s$ parallel to the x axis, we find $D_{2,3}=-v_F q_s/\sqrt{2}$ while $D_{1,4}=+v_F q_s/\sqrt{2}$. $1/T_1T$ thus decreases with T for $|D_m/T| \gg 1$, and increases as $\sim T^2$ for $|D_m/T| \ll 1$, in agreement with the results in Fig. 3. On the other hand, $\phi=\pi/4$, i.e., $\mathbf{p}_s$ along the lattice diagonal, is a special case since $D_{1,3} = \pm v_F q_s$ while $D_{2,4}=0$. While the contribution to $1/T_1T$ from quasiparticle excitations involving only nodes 2 and 4 scales as T^2 for all temperatures, the contributions involving only nodes 1 and 3 decreases with increasing T. As a result, $1/T_1T$ increases for all temperatures, though only weakly at low T. For $T \gg v_F q_s$, the relaxation rate for all ϕ scales as T^2 and thus becomes angle independent. Note, that the strong angular dependence of the relaxation rate for intermediate temperatures explains the spread in values of $1/T_1T$ at a given local field which we found in Fig. 2, particularly in the $T=60$ K curve.

Spin diffusion and vortex vibrations[15] can potentially contribute to the spin-lattice relaxation and thus smear out the effects described above. However, spin diffusion is strongly suppressed by the inhomogeneity of the magnetic field in a vortex lattice.[16] Moreover, calculations on the effect of vortex vibrations in $YBa_2Cu_3O_{6+x}$[16] as well as an experimental comparison of ^{17}O and ^{63}Cu relaxation rates[5] conclude that vortex vibrations are irrelevant in the relaxation of ^{63}Cu but may play an important role in that of ^{17}O.

Recently, several groups have calculated T_1 in the superconducting state starting from a Fermi gas (FG) description of the cuprates, i.e., neglecting antiferromagnetic spin fluctuations.[16–18] We extended our calculations to the FG limit by using $\chi=\Pi/g^2$ and found two distinct differences between the relaxation rate for a FG and that in the presence of strong spin fluctuations. First, $1/T_1T$ for a FG always increases with temperature as long as A_{ab} and B possess the same sign, in contrast to our results in Eq. (8). Second, in the FG limit $1/T_1T$ increases monotonically with increasing local field in a vortex lattice, i.e., it does not exhibit the local minimum we found in Fig. 2. We propose that these differences in the predicted relaxation rates enable NMR experiments to determine which of the two limits applies to the superconducting state of the high-T_c cuprates.

Finally, we found above that the behavior of the relaxation rate in a vortex lattice reflects the presence of a supercurrent, magnetic fluctuations and nodes in the superconducting gap. We are currently studying[19] whether similar effects also occur in Sr_2RuO_4 for which strong indications of ferromagnetic fluctuations and a p-wave order parameter exist.[20]

In summary, we have demonstrated that NMR is a site-specific probe for the electronic structure in the mixed state of the high-T_c cuprates. We have shown that in a hexagonal vortex lattice, the Doppler-shifted electronic spectrum gives rise to a characteristic temperature and field dependence of the ^{63}Cu relaxation rate. We propose an NQR experiment in which the direction of a uniform supercurrent with respect to the crystal lattice is varied, and predict a unique angular dependence of T_1. Finally, we argue that our strong-coupling results are qualitatively different from those predicted for a FG.

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[1] G.E. Volovik, Pis'ma Zh. Éksp. Teor. Fiz. **58**, 457 (1993) [JETP Lett. **58**, 469 (1993)].

[2] Y. Wang and A.H. MacDonald, Phys. Rev. B **52**, R3876 (1995); D. Xu, S.K. Yip, and J.A. Sauls, *ibid.* **51**, 16 233 (1995); M. Franz and Z. Tesanovic, Phys. Rev. Lett. **80**, 4763 (1998).

[3] M. Tinkham, *Introduction to Superconductivity* (Krieger Publishing, Boca Raton, FL, 1980).

[4] K. Moler *et al.*, Phys. Rev. Lett. **73**, 2744 (1994).

[5] N. Curro, C. Milling, J. Haase, and C. P. Slichter (unpublished); N. Curro, Ph.D. thesis, University of Illinois at Urbana-Champaign (1998).

[6] P. Monthoux and D. Pines, Phys. Rev. B **47**, 6069 (1993); D.K. Morr and D. Pines, cond-mat/9807214 (unpublished).

[7] C. Milling and C. P. Slichter (private communication).

[8] Y. Zha, V. Barzykin, and D. Pines, Phys. Rev. B **54**, 7561 (1996).

[9] D. K. Morr, J. Schmalian, and D. Pines (unpublished).

[10] A.J. Millis, H. Monien, and D. Pines, Phys. Rev. B **42**, 167 (1990).

[11] Note that our results for $(T_1T)^{-1}$ are practically independent on whether **Q** is commensurate or incommensurate.

[12] T. Mason *et al.*, Phys. Rev. Lett. **77**, 1604 (1996); B. Lake *et al.* Nature (London) **400**, 43 (1999); P. Dai *et al.*, Phys. Rev. Lett. **80**, 1738 (1998).

[13] M.H.S. Amin, I. Affleck, and M. Franz, Phys. Rev. B **58**, 5848 (1998).

[14] P.I. Soininen *et al.*, Phys. Rev. B **50**, 13 883 (1994); M. Ichioka *et al.*, *ibid.* **59**, 8902 (1999).

[15] V.V. Demidov, Physica C **234**, 285 (1994).

[16] R. Wortis, Ph.D. thesis, University of Illinois at Urbana-Champaign, 1997; R. Wortis *et al.* cond-mat/9907093 (unpublished).

[17] I. Vekhter, J.P. Carbotte, and E.J. Nocol, Phys. Rev. B **59**, 1417 (1999).

[18] M. Takigawa, M. Ichioka, and K. Machida, Phys. Rev. Lett. **83**, 3057 (1999).

[19] D. K. Morr and D. Agterberg (private communication).

[20] T.M. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).