PHYSICAL REVIEW B **78**, 140506(R) (2008)

Pseudogap ground state in high-temperature superconductors

J. G. Storey,* J. L. Tallon, and G. V. M. Williams
MacDiarmid Institute, Industrial Research Ltd., P.O. Box 31310, Lower Hutt, New Zealand
(Received 19 March 2008; revised manuscript received 29 July 2008; published 7 October 2008)

By re-examining recently published photoemission data we demonstrate that the apparent Fermi surface of the pseudogap ground state is a disconnected arc centered on the nodes. This scenario is also consistent with results from Raman spectroscopy and NMR. We also show that it is consistent with doping-dependent entropy data and discuss the origins of this unusual electronic "arc-liquid" state. We show that the pseudogap closes, and the arcs become connected, when the antiferromagnetic (AF) correlation length falls to the size of the lattice parameter. These results are consistent with a picture of Fermi-surface reconstruction due to short-range AF correlations.

DOI: 10.1103/PhysRevB.78.140506 PACS number(s): 74.25.Jb, 74.25.Bt, 74.25.Dw, 74.72.−h

In recent years, many studies, including heat capacity,[1–4] Andreev reflection,[5] Raman spectroscopy,[6] angle-resolved photoemission spectroscopy (ARPES),[7–10] and scanning tunneling microscopy,[11] have converged in support of the presence of two distinct gaps in the low-energy electronic structure of optimal- and underdoped high-T_c cuprates. These are the pseudogap which opens in the normal state (NS) below a critical hole doping of $p_{\mathrm{crit}} \approx 0.19$ hole/Cu (Refs. 2, 3, 12, and 13) and the superconducting (SC) gap which opens at T_c. In ARPES the pseudogap is apparent as a depletion of states near the $(\pi,0)$ Brillouin-zone boundary. As a result, the Fermi surface seems to consist of a set of disconnected "Fermi arcs."[14] Such a state would not be a Fermi liquid and it certainly heralds unconventional behavior.

Below T_c the SC gap opens on these arcs, shrouding their character at zero temperature from direct observation. In what has become an influential paper, Kanigel *et al.*[15] reported measurements of the Fermi-arc length (FAL) above T_c as a function of temperature for various doping levels. Their results were plotted as a function of reduced temperature $t = T/T^*$ where T^* is the temperature above which pseudogap effects are no longer observed in their analysis. When plotted in this fashion, the FAL seems to extrapolate linearly to zero at $t=0$. Thus, they suggest that the $T=0$ pseudogap state is a nodal liquid at all doping levels.

Such a conclusion would have fundamentally important implications for the two scenarios envisaged for the pseudogap. One views the pseudogap as a competing-order parameter while the alternative is to consider the pseudogap as a pairing gap arising from a phase incoherent pairing state or from real-space local pairs. Here is the problem: given that at low doping the pseudogap is usually observed to exceed the SC gap, a nodal ground-state pseudogap would remove all states available for superconductivity. The manifest persistence of superconductivity in underdoped cuprates effectively would eliminate the competing-order-parameter scenario. The normal-state-pairing scenario alone would survive because the ground-state pseudogap is identical to the BCS state. The proposed nodal pseudogap ground state thus provides an important arbiter of these models.

Recently however, we were able to show that Raman data paint a different picture.[16] Raman spectroscopy provides a useful tool for studying energy gaps because the selection of different polarizations allows different regions of the Brillouin zone to be probed. The Raman data of Le Tacon *et al.*[6] were used to test two scenarios for the pseudogap ground state: a nodal state or a Fermi-arc state. It was found that finite FALs at zero temperature were required to reproduce the trend observed in the zone-diagonal B_{2g} Raman response.[16]

Significantly, two other groups reached the same conclusion. First, Wen and Wen[4] determined the change in specific-heat coefficient at high magnetic fields in $La_{2-x}Sr_xCuO_4$ and inferred a nonzero value of the normal-state specific-heat coefficient at $T=0$. Second, Zheng *et al.*[17] carried out NMR measurements in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ at very high fields, sufficient to fully suppress superconductivity and expose the pseudogap ground state. They found nonzero $1/T_1T$ values at $T=0$ which are suppressed to zero as the field is reduced and superconductivity is re-established. Both of these indicate a finite normal-state residual density of states (DOS) at $T=0$ consistent with an arc-liquid state. Moreover, the residual DOS grows with increased doping consistent with growth of the $T=0$ FAL, as will be discussed below. In the following we seek to reconcile this apparent conflict between NMR and specific heat on the one hand and the ARPES results[15] on the other.

In Fig. 1 we replot the data of Kanigel *et al.*,[15] unscaled in terms of absolute temperature, from the raw data shown in Fig. 4(a) of their paper. We see that, with the exception of the most underdoped sample ($T^*=500$ K), the FALs extrapolate to nonzero values at $T=0$. The six data points for $T^* = 240/260$ K give a least-squares linear intercept of $36.3° \pm 1.2°$. Further, the $T=0$ FAL increases with increasing doping, consistent with our Raman modeling where a $T=0$ FAL roughly proportional to T_c was found to reproduce the data. This conclusion is also precisely consistent with the specific-heat and NMR data noted above. The scaling analysis of Kanigel *et al.*[15] conceals all these important details.

Here we independently determine the apparent FAL from the electronic entropy, S, as follows. We effectively subsume the unconventional strong-correlation physics into the pseudogap phenomenology and then use a Fermi-liquid approach to calculate the NS entropy from a rigid ARPES-derived dispersion. This may seem a peculiar juxtaposition (of the exotic and conventional) but it works, yielding the full p and T dependences of the entropy and superfluid density, with no adjustable parameters.[16,18] We presume that its

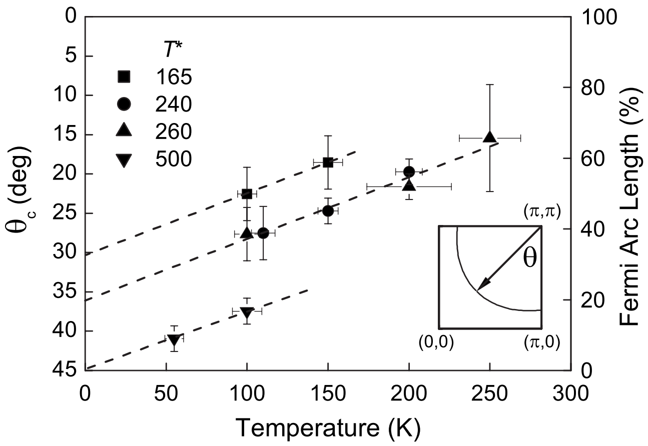

FIG. 1. Solid symbols: Fermi-arc length extracted from the raw data of Ref. 15 plotted vs temperature. With the exception of the most underdoped sample, the Fermi-arc lengths extrapolate to finite values at $T=0$. Inset: Fermi surface showing the angle θ.

success derives from the fact that, at low enough T, the entropy and superfluid density are dominated by the near-nodal regions where quasiparticle lifetimes are long. In further support, we note that across the entire p and T regimes one finds experimentally that $S/T=a_W\chi_0$ where χ_0 is the static susceptibility and a_W is the Wilson ratio for nearly free electrons.[13]

Previously,[18] we used a nodal pseudogap model [$\theta_{0,\max}=45°$ in Eq. (1)]. Although excellent fits to the normal-state entropy were obtained, we could not evaluate the SC gap, $\Delta(T)$, self-consistently using the BCS gap equation in the presence of the pseudogap. The fully nodal "non-states-conserving" pseudogap model simply removed too many states to provide a converging solution. As a result, the pseudogap was left out of the calculation of $\Delta(T)$, a somewhat artificial solution. We now note that a ground-state arc model allows the pseudogap to be included in the self-consistent calculation.

Shown in Fig. 2(a) are normal-state fits made to the entropy data of Loram *et al.*[13] using the approach described previously[18] but now with an extra parameter $\theta_{0,\max}$ that defines the angle to which the FAL extends at $T=0$. The T dependence of the FAL is given by

$$\theta_0(T) = \theta_{0,\max}\left[1-\tanh\left(\frac{T}{2T^*}\right)\right]. \qquad (1)$$

The parameters extracted from the fits are shown in Fig. 2(b) (solid symbols) along with experimental values from the data of Kanigel *et al.*[15] and other ARPES papers (open symbols). The inferred distance away from the van Hove singularity (vHS), E_F-E_{vHS}, is shown in units of millielectron volts in the inset of Fig. 2(b). This descends rapidly with doping and the vHS crossing occurs at $p\approx 0.22$ as before.[18,19] The general agreement of the fits is very good and, importantly, shows the $T=0$ Fermi-arc angle (arrowed) tending toward 45° as doping decreases. This is a key conclusion: the Fermi arcs collapse to point nodes at $p=0.05$ where superconductivity disappears. At this point there is no residual spectral weight available for superconductivity.

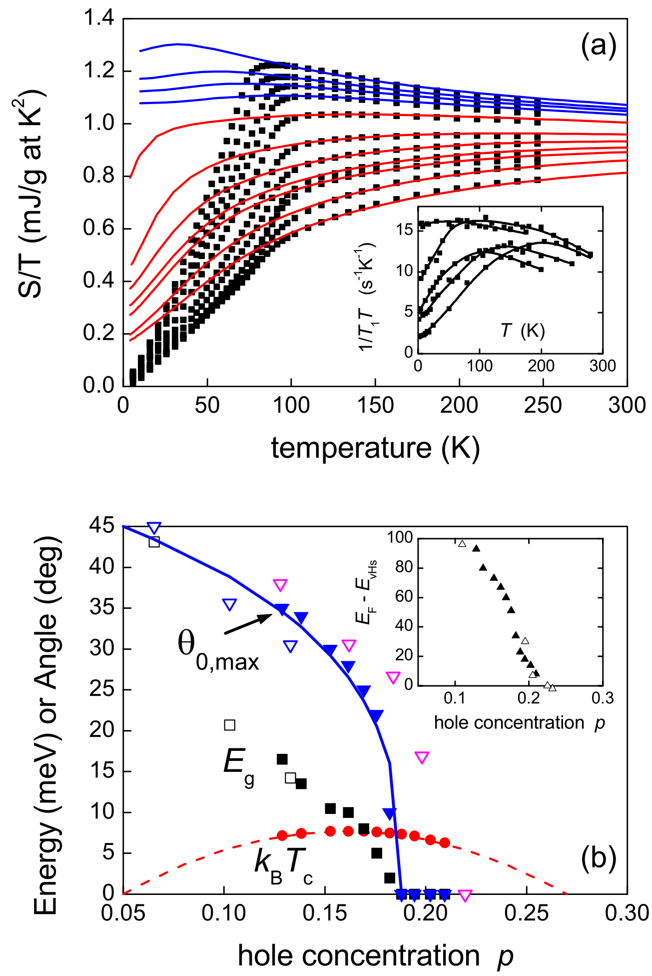

FIG. 2. (Color online) (a) Normal-state fits to the Bi-2212 entropy data of Loram *et al.* (Ref. 13) assuming a finite arc pseudogap model. For clarity every 20th data point only is shown. Inset: High-field $1/T_1T$ data from Ref. 17 showing finite states at $T=0$. (b) Fermi-arc angle, $\theta_{0,\max}$, at $T=0$ and pseudogap magnitude $E_g=k_BT^*$ extracted from the fits in (a). Open symbols are experimental values, $\theta_{0,\max}$ and E_g, which are from Ref. 15. Inset: Position of Fermi level E_F-E_{vHS} (in millielectron volts), from fits (solid symbols) and from Refs. 19–21 (open symbols).

This conclusion, namely, that the pseudogap ground state is an arc liquid and the arc smoothly contracts to the node as $p\rightarrow 0.05$, is not only consistent with the data of Kanigel *et al.*[15] [open blue down triangles in Fig. 2(b)] but it is supported in detail by the $1/T_1T$ data of Zheng *et al.*[17] As noted, they use high fields (28.5–43 T) sufficient to fully suppress superconductivity and expose the pseudogap ground state. Their data, reproduced in the inset of Fig. 2(a), show a residual nonzero value of $1/T_1T$ as $T\rightarrow 0$ which is progressively reduced to zero as doping is decreased. The correspondence with the entropy data shown in the same figure is apparent and our analysis of their data [open magenta down triangles in Fig. 2(b)] shows a similar doping dependence of $\theta_{0,\max}$, though this is a single-layer cuprate. A similar quantitative correspondence can be found with the residual specific-heat coefficient inferred by Wen and Wen.[4]

An exacting test of the $T=0$ FAL is also provided by the

T dependence of the superfluid density $\rho_s(T)$ which, if the Fermi arc continues to shrink toward the node, would exhibit a downturn at low temperature[18] that is not observed. In fact, the detailed T dependence of $\rho_s(T)$ should provide strong constraints on whether and by how much the Fermi arcs continue to shrink below T_c. The c-axis infrared conductivity $\sigma_c(\omega)$ measured by Yu *et al.*[22] also provides a guide as to the evolution of the Fermi arcs below T_c. On cooling below T^* these authors found a smooth loss of spectral weight below the pseudogap energy ω_{PG} which is transferred to higher energy above the gap. With the onset of superconductivity spectral weight below the SC gap is transferred to the $\omega=0$ superfluid delta function. In this way both gaps can be clearly identified. Importantly, below T_c no further weight is transferred above ω_{PG} and no further loss of spectral weight occurs in the frequency range between ω_{SC} and ω_{PG}. This places tight constraints on the degree to which Fermi arcs continue to shrink below T_c and certainly indicates that they do not collapse to the nodes.

This all seems rather persuasive except that more recently Kanigel *et al.*[23] reported a further study on Fermi arcs and the arc angles from this study appear to extrapolate approximately to the nodal points at $T=0$. This conflicts with their former data and we can only reserve judgment pending further studies. Certainly the former data are much more in keeping with the Raman, specific-heat, entropy, infrared, superfluid density, and NMR data we have discussed.

We consider some possible interpretations of this Fermi-arc phenomenology. (a) There may be some competing order which opens up a gap on the Fermi surface. This would account for the abrupt crossover from strong to weak superconductivity seen in the specific-heat jump, the condensation energy, critical currents, and superfluid density.[13] Certain recent experiments support such a scenario.[24,25] Such a gap may be nodal but broadening from strong scattering smears out the gap near the nodes giving the misleading impression of pristine Fermi arcs near the zone diagonal.[16] We have found that a simple model of a linear-in-T scattering rate which saturates at low T recovers the phenomenology shown by $\theta_{0,\max}(p)$ in Fig. 2(b). (b) Alternatively, the pseudogap phenomenology could derive from incoherent pairing above T_c.[26] But the continuing problem for this scenario is that the crossover from strong to weak superconductivity (below $p_{\mathrm{crit}}=0.19$) occurs even in the $T=0$ ground state where thermal fluctuations are no longer present.[12,16] The pseudogap persists at $T=0$ and this remains a crucial obstacle to fluctuation models.

We feel the most promising scenario is (c) that described, e.g., by Harrison *et al.*[27] where residual short-range antiferromagnetic (AF) order results in a folding of the Brillouin zone along the $(0,\pi)-(\pi,0)$ line, opening up a true gap near the zone boundary with the consequent formation of reconstructed Fermi pockets near the zone diagonal. Due to the finite AF correlation length, ξ_{AF}, the spectral weight of the Fermi pockets on the zone-center side remains strong while that on the (π,π) side is washed out. The effect is that the hole pockets at low doping evolve into disconnected arcs at higher doping which eventually extend out around the large Fermi surface (probably occurring at p_{crit} where the pseudogap closes). This effectively reproduces the full Fermi-arc phenomenology as described above. Here, the evolution of the Fermi arc is governed by ξ_{AF} and its rapid decrease with increasing doping. The closure of the pseudogap would then correspond to a critical length scale for ξ_{AF} and we suggest that this is probably $\xi_{\mathrm{AF}}\approx a$, where a is the lattice parameter.

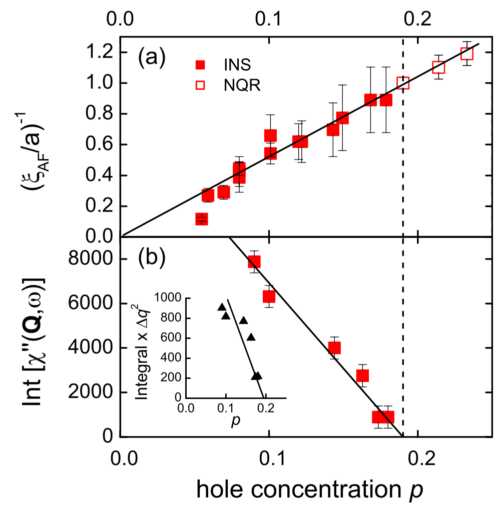

FIG. 3. (Color online) (a) The p dependence of ξ_{AF}^{-1} obtained from the q width of $\chi''(\mathbf{q}_{\mathbf{AF}},\omega)$ from Ref. 28 and from NQR data (Ref. 30). (b) The p dependence of $I=\int\chi''(\mathbf{q}_{\mathbf{AF}},\omega)d\omega$ (in arbitrary units) integrated from 0 to 50 meV. Inset: $I\times\Delta q^2$.

To show this we plot in Fig. 3(a) the doping dependence of the normal-state magnitude of ξ_{AF}^{-1} for $YBa_2Cu_3O_{7-\delta}$ determined from the q width of the $(\pi/2,\pi/2)$ peak in the imaginary part of the dynamical susceptibility, $\chi''(\mathbf{q},\omega)$.[28] Here we have converted oxygen deficiency, δ, to hole concentration in our usual way.[29] In addition we have included values of ξ_{AF} determined from nuclear quadrupole resonance (NQR) measurements[30] of $1/T_1T$ data for overdoped $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$. The value of ξ_{AF} from the NQR data is determined only within a multiplicative constant and so the absolute values are enforced by continuity with the neutron-scattering data. The figure shows that $\xi_{\mathrm{AF}}=a$ at $p\approx p_{\mathrm{crit}}=0.19$. When $\xi_{\mathrm{AF}}<a$ the concept of short-range AF correlations breaks down and, rather naturally, the Fermi surface should be fully recovered from zone-folding effects at this point. It is possible that, as the Fermi arc breaks through the zone boundary the change in topology is discontinuous, leading to the occurrence of a quantum critical point.[31]

Further, the AF weight as measured by $\int\chi''(\mathbf{q}_{\mathbf{AF}},\omega)d\omega$, where the integral is from 0 to 50 meV, is plotted in Fig. 3(b) and this falls rapidly, projecting to zero at $p=p_{\mathrm{crit}}$. Here $\mathbf{q}_{\mathbf{AF}}$ is the AF wave vector. These values have been reported by Bourges[32] and we have simply converted δ values to p values. This reveals an abrupt loss of AF weight at p_{crit}. More accurately, because of the increasing q width with doping, we should also integrate over q. We approximate this by calculating $\Delta q^2\times\int\chi''(\mathbf{q}_{\mathbf{AF}},\omega)d\omega$ and this is shown in the inset of Fig. 3(b). Again, the total weight descends rapidly and projects to zero at p_{crit}. We suggest that both the critical contraction of ξ_{AF} and the critical loss of AF weight are generic properties of the cuprates and, together, they allow the critical extension of the Fermi arcs to a fully connected Fermi surface. ($La_{2-x}Sr_xCuO_4$, with its magnetic incommensuration which extends to high doping is clearly an exception. Nevertheless, we presume this is added complexity on top of otherwise generic behavior.)

These relationships are nicely condensed into a single expression. From the present observations $\xi_{AF}=a[p_{crit}/p]$ while we have previously shown[13] that $E_g \approx J[1-p/p_{crit}]$. This leads to the result that $E_g \approx J[1-a/\xi_{AF}]$ which effectively defines both E_g and the pseudogap line T^* purely in terms of AF parameters.

In summary, we have shown that when plotted in terms of absolute temperature, recent ARPES measurements show the pseudogap ground state to exhibit finite disconnected Fermi arcs, not point nodes. We show that this scenario is consistent with results from Raman, specific-heat, NMR, and infrared studies and we therefore consider our conclusions to be robust. As a consequence, theories that attempt to reproduce a nodal scenario may be misdirected. We show that the pseudogap closes, and the arcs become connected, when the AF correlation length falls to the size of the lattice parameter. These results are consistent with a picture of Fermi-surface reconstruction due to short-range AF correlations.

*Present address: Cavendish Laboratory, Cambridge University, Cambridge CB3 0HE, UK.

[1] J. W. Loram, K. A. Mirza, J. R. Cooper, W. Y. Liang, and J. M. Wade, J. Supercond. **7**, 243 (1994).

[2] J. L. Tallon, G. V. M. Willams, M. P. Staines, and C. Bernhard, Physica C **235-240**, 1821 (1994).

[3] J. L. Tallon and J. W. Loram, Physica C **349**, 53 (2001).

[4] H. H. Wen and X. G. Wen, Physica C **460-462**, 28 (2007).

[5] G. Deutscher, Nature (London) **397**, 410 (1999).

[6] M. Le Tacon, A. Sacuto, A. Georges, G. Kotliar, Y. Gallais, D. Colson, and A. Forget, Nat. Phys. **2**, 537 (2006).

[7] K. Tanaka *et al.*, Science **314**, 1910 (2006).

[8] M. Hashimoto, K. Tanaka, T. Yoshida, A. Fujimori, M. Okusawa, S. Wakimoto, K. Yamada, T. Kakeshita, H. Eisaki, and S. Uchida, Physica C **460-462**, 884 (2007).

[9] W. S. Lee, I. M. Vishik, K. Tanaka, D. H. Lu, T. Sasagawa, N. Nagaosa, T. P. Devereaux, Z. Hussain, and Z. X. Shen, Nature (London) **450**, 81 (2007).

[10] T. Kondo, T. Takeuchi, A. Kaminski, S. Tsuda, and S. Shin, Phys. Rev. Lett. **98**, 267004 (2007).

[11] M. C. Boyer, W. D. Wise, K. Chatterjee, M. Yi, T. Kondo, T. Takeuchi, H. Ikuta, and E. W. Hudson, Nat. Phys. **3**, 802 (2007).

[12] C. Bernhard, J. L. Tallon, T. Blasius, A. Golnik, and C. Niedermayer, Phys. Rev. Lett. **86**, 1614 (2001).

[13] J. W. Loram, J. Luo, J. R. Cooper, W. Y. Liang, and J. L. Tallon, J. Phys. Chem. Solids **62**, 59 (2001).

[14] M. R. Norman *et al.*, Nature (London) **392**, 157 (1998).

[15] A. Kanigel *et al.*, Nat. Phys. **2**, 447 (2006).

[16] J. G. Storey, J. L. Tallon, G. V. M. Williams, and J. W. Loram, Phys. Rev. B **76**, 060502(R) (2007).

[17] G. Q. Zheng, P. L. Kuhns, A. P. Reyes, B. Liang, and C. T. Lin, Phys. Rev. Lett. **94**, 047006 (2005).

[18] J. G. Storey, J. L. Tallon, and G. V. M. Williams, Phys. Rev. B **77**, 052504 (2008).

[19] A. Kaminski, S. Rosenkranz, H. M. Fretwell, M. R. Norman, M. Randeria, J. C. Campuzano, J. M. Park, Z. Z. Li, and H. Raffy, Phys. Rev. B **73**, 174511 (2006).

[20] H. Ding *et al.*, Phys. Rev. Lett. **76**, 1533 (1996).

[21] A. A. Kordyuk, S. V. Borisenko, M. Knupfer, and J. Fink, Phys. Rev. B **67**, 064504 (2003).

[22] L. Yu, D. Munzar, A. V. Boris, P. Yordanov, J. Chaloupka, T. Wolf, C. T. Lin, B. Keimer, and C. Bernhard, Phys. Rev. Lett. **100**, 177004 (2008).

[23] A. Kanigel, U. Chatterjee, M. Randeria, M. R. Norman, S. Souma, M. Shi, Z. Z. Li, H. Raffy, and J. C. Campuzano, Phys. Rev. Lett. **99**, 157001 (2007).

[24] J. Xia *et al.*, Phys. Rev. Lett. **100**, 127002 (2008).

[25] B. Fauqué, Y. Sidis, V. Hinkov, S. Pailhès, C. T. Lin, X. Chaud, and P. Bourges, Phys. Rev. Lett. **96**, 197001 (2006).

[26] V. J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995).

[27] N. Harrison, R. D. McDonald, and J. Singleton, Phys. Rev. Lett. **99**, 206406 (2007).

[28] A. V. Balatsky and P. Bourges, Phys. Rev. Lett. **82**, 5337 (1999).

[29] J. L. Tallon, C. Bernhard, H. Shaked, R. L. Hitterman, and J. D. Jorgensen, Phys. Rev. B **51**, 12911 (1995).

[30] G. V. M. Williams, S. Krämer, and M. Mehring, Phys. Rev. B **63**, 104514 (2001).

[31] J. L. Tallon, J. W. Loram, and C. Panagopoulos, J. Low Temp. Phys. **131**, 387 (2003).

[32] P. Bourges, in *Neutron Scattering in Novel Materials*, Proceedings of the Eighth PSI Summer School on Neutron Scattering, Zuoz, Switzerland, edited by A. Furrer (World Scientific, Singapore, 2000).