PHYSICAL REVIEW B **79**, 180505(R) (2009)

Thermopower across the stripe critical point of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$: Evidence for a quantum critical point in a hole-doped high-T_c superconductor

R. Daou,[1] Olivier Cyr-Choinière,[1] Francis Laliberté,[1] David LeBoeuf,[1] Nicolas Doiron-Leyraud,[1] J.-Q. Yan,[2] J.-S. Zhou,[2] J. B. Goodenough,[2] and Louis Taillefer[1,3,*]

[1]*Département de physique and RQMP, Université de Sherbrooke, Sherbrooke, Quebec, Canada, J1K 2R1*

[2]*Texas Materials Institute, University of Texas–Austin, Austin, Texas 78712, USA*

[3]*Canadian Institute for Advanced Research, Toronto, Ontario, Canada, M5G 1Z8*

(Received 29 March 2009; published 12 May 2009)

The thermopower S of the high-T_c superconductor $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ was measured as a function of temperature T near its quantum critical point, the critical hole doping $p^\star$ where all characteristic temperatures go to zero. Just above $p^\star$, S/T varies as $\ln(1/T)$ over a decade of temperature. Below $p^\star$, S/T undergoes a large increase at low temperature. As with the temperature dependence of the resistivity, which is linear just above $p^\star$ and undergoes a large upturn at low temperature, these are typical signatures of a quantum phase transition. This suggests that $p^\star$ is a quantum critical point below which some order sets in, causing a reconstruction of the Fermi surface, whose fluctuations are presumably responsible for the linear-T resistivity and logarithmic thermopower. All the evidence points to "stripe" order, a form of spin/charge modulation known to exist in this material.

DOI: 10.1103/PhysRevB.79.180505 PACS number(s): 74.72.Dn, 72.15.Jf, 74.25.Fy, 75.30.Kz

The nature of the pseudogap phase in high-T_c superconductors has yet to be elucidated. Quantum oscillation studies[1] have revealed that the large holelike Fermi surface characteristic of highly overdoped cuprates[2] is modified in the pseudogap phase, where it contains small electronlike pockets.[3] A fundamental question is: what causes this change in Fermi surface? Is it the onset of some order? If so, what symmetry is broken?

Recent measurements of the Hall coefficient $R_H(T)$ in the hole-doped cuprate $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ (Nd-LSCO) at $p=0.20$ (Ref. 4) have revealed a pronounced upturn at low temperature (see inset to Fig. 2), which coincides with the onset of stripe order (see Fig. 1). At a slightly higher doping, $p=0.24$, $R_H(T)$ remains flat at low temperature (see inset in top panel of Fig. 2), with the value expected of a large holelike Fermi surface containing $1+p$ holes.[4] This shows that the onset of stripe order at $p^\star \simeq 0.24$ causes a reconstruction of the large Fermi surface.[5] In the large-Fermi-surface state, the normal-state resistivity $\rho(T)$ is linear in temperature down to the lowest temperatures[4] (see inset to Fig. 1) and the Nernst coefficient $\nu(T)$ is small, negative, and featureless.[6] At $p=0.20$, however, both ρ (Ref. 4) and ν/T (Ref. 6) show a pronounced upturn, with respective onset temperatures T_ρ and T_ν that coincide [$T_\rho \simeq T_\nu$ (Ref. 6)] and are a factor of 2 above the onset of stripe order (see Fig. 1). Recent photoemission measurements on Nd-LSCO at $p=0.12$ (Ref. 7) suggest that the pseudogap in Nd-LSCO may have the same features as in other cuprates, with an onset temperature $T^\star$ (Ref. 7) consistent with T_ρ and T_ν. This implies that the pseudogap phase may be a fluctuating precursor of the long-range stripe order that sets in at lower temperature.[5]

In this Rapid Communication we investigate the thermopower $S(T)$ of Nd-LSCO. In general, the thermopower is a complex quantity that involves the energy dependence of the conductivity.[11,12] However, in the limit of dominant impurity scattering, it has been shown theoretically that $S/T \propto (C_e/T)(1/ne)$, where C_e is the electronic specific heat, n is the density of charge carriers, and e is the charge of the electron.[12] Empirically, it has been pointed out that $S/T \approx (C_e/T)(1/ne)$ in the limit of $T \to 0$ for a wide range of strongly correlated electron systems.[11] Therefore, at low temperature the thermopower approximately represents the electronic heat capacity per charge carrier. (Note that it would be difficult to measure $C_e(T)$ accurately in Nd-LSCO given that it is less than 1% of the total specific heat $C(T)$ above 4 K, and the low-temperature behavior is masked by a large Schottky anomaly.[13,14]) We find that the three regimes of behavior seen in the resistivity as upturn for $p<p^\star$, linear for $p=p^\star$, and quadratic for $p>p^\star$, show up in S/T, respectively, as upturn, logarithmic divergence, and nearly flat. This is strongly reminiscent of the electron behavior in metals near a quantum phase transition,[15] suggesting that the pseudogap phase is characterized by some order, which vanishes at a quantum critical point located inside the region of superconductivity in the phase diagram. All evidence points to so-called "stripe" order as the anomalies in transport correlate with the onset of spin/charge modulations.

The two samples of Nd-LSCO used in this study are the same as those used and described in Refs. 4 and 6. They have a doping of $p=0.20$ and $p=0.24$, with respective T_c values of 20 and 17 K. The thermopower was measured using a one-heater, two-thermometer dc technique, with Cernox thermometers. The applied temperature gradient was always less than 7% of the average sample temperature. The thermopower of the resistive leads in the measurement circuit (PtW or phosphor-bronze) was calibrated against optimally doped $YBa_2Cu_3O_y$ (YBCO) ($T_c=93$ K) for $T<90$ K and 6N-pure Pb for $T>90$ K.[16] The $p=0.24$ sample was also measured using a low-frequency two-heater, two-thermometer ac technique,[17] with a sinusoidal excitation of frequency of 5–100 mHz and amplitude of 0.1 K. The signal-to-noise ratio in the ac measurement was ten times better than in the dc case. There was excellent agreement in the data obtained with both techniques.

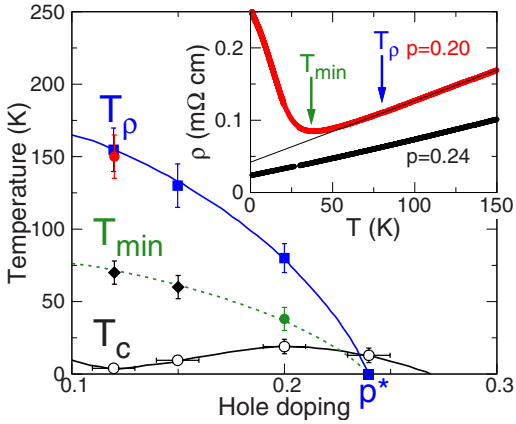

FIG. 1. (Color online) Phase diagram of Nd-LSCO. The temperature T_ρ (blue squares and solid line) is defined as the temperature below which the normal-state resistivity deviates from its linear-T behavior at high temperature (see inset and (Ref. 4). T_ρ coincides with T_ν, the onset of the upturn in the Nernst coefficient (Ref. 6). T_ρ and T_ν correlate with the (extrapolated) opening of the pseudogap seen by photoemission at $p=0.12$ (red circle) (Ref. 7). Since the linear-T dependence extends down to $T\rightarrow 0$ at $p=0.24$ (see inset), $T_\rho=0$ at that doping. Although we define the critical point $p^\star$ where $T_\rho=0$ is at $p=0.24$, it could be slightly lower. The superconducting transition temperature T_c (open black circles) is also plotted, showing that the T_ρ line must end inside the superconducting phase. Data for $p=0.12$ and $p=0.15$ are from Ref. 8; data for $p=0.20$ and $p=0.24$ are from Ref. 4. Also shown is the upturn temperature T_{min} (closed green circle and dashed line) at which the resistivity reaches its minimum value (see inset). The onset of charge order deduced from x-ray diffraction (Refs. 9 and 10) (black diamonds) coincides with T_{min} (see Refs. 4 and 8). Inset: normal-state resistivity of the two Nd-LSCO crystals used in this study, measured in a magnetic field strong enough to entirely suppress superconductivity (from Ref. 4).

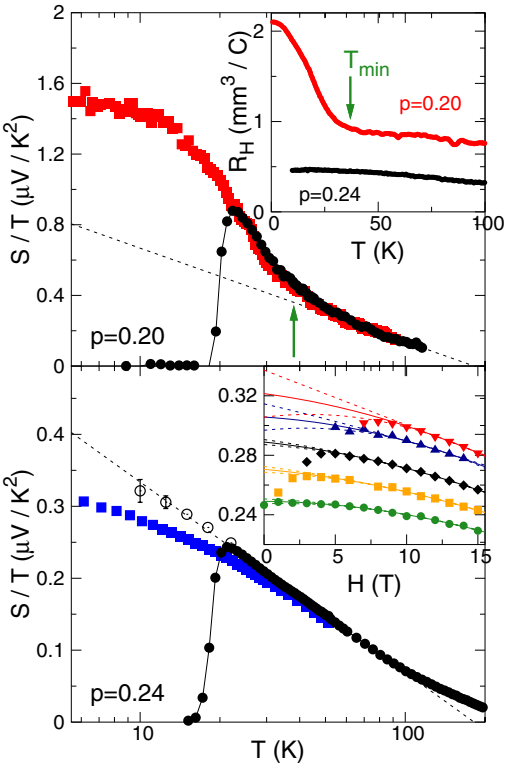

FIG. 2. (Color online) Thermopower $S(T)$ of Nd-LSCO, plotted as S/T vs log T, with (squares) and without (full circles) a magnetic field of 15 T applied along the c axis. Top panel: sample with $p=0.20$, measured with the dc technique (see text). The magnetic field has no discernible effect other than to suppress superconductivity. The dashed line is a linear fit to the data above 50 K. The arrow marks T_{min} (see inset of Fig. 1). Inset: Hall coefficient as a function of temperature, for both Nd-LSCO samples (Ref. 4). At $p=0.20$, the upturn in $R_H(T)$ at low temperature is seen to coincide with T_{min}. Bottom panel: sample with $p=0.24$, measured with the ac technique (see text). The magnetic field is seen to cause a reduction in S for $T<40$ K. To correct for this and to extend the zero-field behavior to $T<T_c$, we extrapolate finite field data to zero field (see inset). The resulting extrapolated values are plotted as open circles. Inset: S/T for Nd-LSCO with $p=0.24$ as a function of magnetic field, at five fixed temperatures: 10, 12.5, 15, 18, and 22 K (top to bottom). Second-order polynomial fits to the field dependence are extrapolated back to zero field. Best (solid lines) and worst (dashed lines) fits are shown, indicative of the uncertainty in the width and position of the superconducting downturn. The corresponding error bars are shown in the main panel (open circles).

In Fig. 2, we plot S/T vs log T for Nd-LSCO at $p=0.20$ and $p=0.24$. The data taken on our $p=0.20$ crystal ($x=0.20$ and $y=0.4$) are in excellent quantitative agreement with previous measurements on $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ at the same values of x and y, over the entire temperature range in zero magnetic field.[18] Our data on Nd-LSCO at $p=0.24$ are in good quantitative agreement with published data on polycrystalline LSCO at $p=0.25$ (only reported in zero field).[19,20]

There is no consensus on the mechanism that governs the thermopower in cuprates. While phonon drag has been invoked to explain the temperature dependence in $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212),[21] it is not satisfactory for the case of Nd-LSCO and YBCO where neither electron-phonon nor mass-enhancement mechanisms are adequate.[22,23] Here we propose an electronic origin for both the temperature and doping dependence of S, at least below 100 K. This is strongly supported by the similarity found in resistivity and Hall effect. At a doping $p=0.24$, close to $p^\star$, S/T in zero magnetic field shows a perfect $\ln(T_0/T)$ dependence from 100 K down to T_c. Application of a magnetic field $H\|c$ =15 T to push T_c down is seen to slightly suppress S/T below this $\ln(T_0/T)$ dependence for $T<40$ K. By extrapolating the field dependence of S/T to $H=0$, as shown in the inset to Fig. 2, we can track the zero-field S/T at temperatures below $T_c(H=0)$. This shows that the $\ln(1/T)$ regime persists at least down to 10 K, within the uncertainty of this extrapolation, i.e., over a full decade of temperature.

This $\ln(T_0/T)$ dependence of S/T is strongly reminiscent of the $\ln(T_0/T)$ dependence observed in C_e/T at the quantum critical point of various heavy-fermion metals.[15] In Fig. 3, we compare S/T in Nd-LSCO with C_e/T in the heavy-fermion metal $CeCu_{6-x}Au_x$,[24] each at three concentrations: below, at, and above their respective critical points, $p^\star$ and x_c. By substituting Au in $CeCu_6$, antiferromagnetic order is

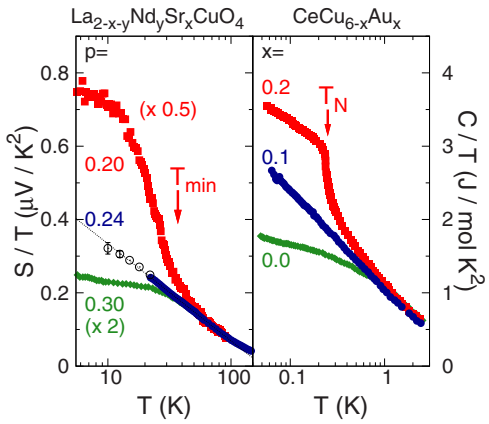

FIG. 3. (Color online) Left panel: thermopower of $La_{2-x-y}Nd_ySr_xCuO_4$ with $p=0.20$ ($\times$ 0.5) and $p=0.24$ ($y=0.4$, this work), compared to that of LSCO with $p=0.30$ [$\times$ 2; $y=0$, from 5–40 K (Ref. 25)], plotted as S/T vs $\log T$. Right panel: specific heat of the heavy-fermion metal $CeCu_{6-x}Au_x$ at $x=0.0$, 0.1, and 0.3, plotted as C_e/T vs $\log T$, showing the evolution across the quantum critical point at $x=x_c=0.1$ where the ordering temperature T_N goes to zero (from Ref. 24).

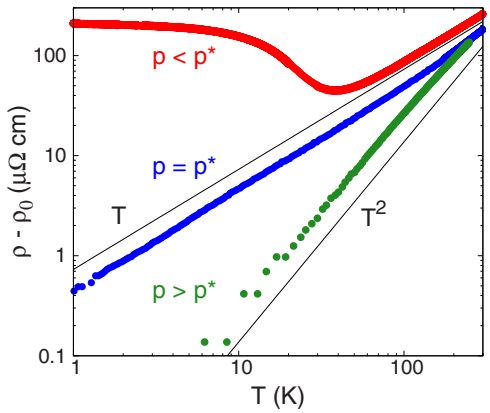

FIG. 4. (Color online) Temperature-dependent part of the resistivity, $\rho(T)-\rho_0$, vs $\log T$ for $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with $p=0.20$ ($p<p^\star$) and $p=0.24$ ($p=p^\star$), from Ref. 4, compared to that of LSCO with $p=0.30$ ($p>p^\star$), from Ref. 26. ρ_0 is the value to which $\rho(T)$ extrapolates at $T=0$; for Nd-LSCO at $p=0.20$, the extrapolation is based only on data above $T^\star=80$ K.

made to appear beyond a critical concentration $x=x_c=0.1$, with an ordering temperature T_N that rises linearly with x.[15,24] In the absence of data on Nd-LSCO at $p>p^\star$, we compare with data on LSCO ($y=0$) at $x=p=0.30$.[25] Given that both materials exhibit virtually identical resistivity and thermopower above $T^\star$,[18] it is reasonable to assume they also do above $p^\star$.

The similarity is remarkable, with both materials displaying the three distinctive regimes of quantum criticality: relatively flat in the Fermi-liquid state, logarithmically divergent at the critical point, and a jump in the ordered state. The characteristic temperature scale T_0 in the $\ln(T_0/T)$ dependence of either S/T or C_e/T is of course vastly different in the two materials, by roughly 2 orders of magnitude, as are the ordering and pseudogap temperatures, T_N and $T^\star$. This qualitative similarity reinforces the case for a quantum phase transition in Nd-LSCO at $p^\star$, previously made on the basis of resistivity,[4] whose three regimes are displayed in Fig. 4: quadratic in the Fermi-liquid state, linear at the critical point, and an upturn below that point.

There is also a strong similarity with the electron-doped cuprate $Pr_{2-x}Ce_xCuO_{4+\delta}$ (PCCO), where the case for a quantum critical point is well established.[27] In the $T\rightarrow 0$ limit, both R_H and S/T in PCCO show an abrupt change as the doping x drops below the critical doping x_c, signaling the change in Fermi surface from a large hole cylinder to a combination of small electron and hole pockets.[28,29] The two coefficients track each other as equivalent measures of the effective carrier density.[28] At $x=x_c$, $\rho(T)$ is again linear in temperature at low T.[30] These typical signatures of a quantum critical point have been attributed to the loss of antiferromagnetic order near x_c (Ref. 31) and the quantum fluctuations thereof.

In a model of charge carriers on a three-dimensional Fermi surface scattered by two-dimensional antiferromagnetic spin fluctuations, transport properties near the magnetic quantum critical point are found to be dominated by "hot spots," points on the Fermi surface connected by the ordering wave vector. In this case, calculations show that $\rho(T)\propto T$, $C_e/T\propto\ln(T_0/T)$, and $S/T\propto\ln(T_0/T)$, where k_BT_0 is an energy scale on the order of the bandwidth.[32] This naturally accounts for the different temperature scales observed in Nd-LSCO and $CeCu_{6-x}Au_x$ where $T_0\simeq 170$ K in the former and 4 K in the latter since the Fermi velocity is about 10^5 m/s in cuprates and 10^3 m/s in heavy-fermion metals.

The strong empirical similarity with both heavy-fermion metals and electron-doped cuprates makes a compelling case for a quantum critical point at $p^\star$ in Nd-LSCO. The nature of the order below $p^\star$ seems to involve both spin and charge degrees of freedom. On the one hand, superlattice Bragg peaks observed in Nd-LSCO by neutron diffraction show that a static (or slow) spin modulation at low temperature persists all the way up to $p\approx p^\star$.[8] On the other hand, the upturn in $\rho(T)$ at $T_{\rm min}$ coincides with the onset of charge order,[4] which occurs at a temperature somewhat above the onset of spin modulation.[8] In other words, the pseudogap phase below $T^\star$ (and $p^\star$) appears to be a phase with "stripe" order, perhaps short-range or fluctuating above $T_{\rm min}$.

The impact of stripe order on the Fermi surface of a hole-doped cuprate has been calculated.[33] The large holelike cylinder is found to reconstruct in a way that depends on the strength of the spin and charge potentials. Calculations of the associated Hall coefficient predict a rise in R_H with the onset of charge order,[34] as observed experimentally in Nd-LSCO when going from $p=0.24$ to $p=0.20$.[4] Spin order can cause a drop in R_H, which can even become negative,[34] as a result of an electron pocket being present in the reconstructed Fermi surface.[33] Such a drop is indeed seen in Nd-LSCO at lower doping, in the vicinity of $p=1/8$, where $R_H(T\rightarrow 0)\approx 0$.[18] The fact that a large drop in $R_H(T)$ also occurs in YBCO near $p=1/8$,[3] starting at a very similar temperature,[5] points to a common underlying cause of Fermi-surface reconstruction.

In conclusion, the combination of resistivity, Hall coefficient, Nernst coefficient, and thermopower in Nd-LSCO makes a compelling case that the pseudogap phase in this

high-T_c superconductor ends at a quantum critical point located inside the superconducting dome at $p \approx 0.24$. All four transport coefficients undergo a simultaneous rise at low temperature which correlates with the onset of charge order seen by other probes. This is compelling evidence that the Fermi surface is reconstructed by "stripe" order. Given that a linear-T resistivity is a universal property of cuprates near optimal doping, it is likely that a common mechanism is at play, associated with such a quantum critical point, in broad analogy with heavy-fermion metals.

We thank K. Behnia and N.E. Hussey for allowing us to show their unpublished thermopower data on LSCO at $x=0.30$.[25] We also thank A. Chubukov, P. Coleman, Y. B. Kim, S. A. Kivelson, G. Kotliar, K. Haule, G. G. Lonzarich, A. J. Millis, M. R. Norman, C. Proust, T. M. Rice, S. Sachdev, T. Senthil, H. Takagi, and A.-M. S. Tremblay for helpful discussions and J. Corbin for his assistance with the experiments. L.T. acknowledges the support of a Canada Research Chair, NSERC, FQRNT and CIfAR. J.S.Z. and J.B.G. were supported by an NSF grant.

*louis.taillefer@physique.usherbrooke.ca

[1] N. Doiron-Leyraud, C. Proust, D. LeBoeuf, J. Levallois, Bonnemaison, R. X. Liang, D. A. Bonn, W. N. Hardy, and L. Taillefer, Nature (London) **447**, 565 (2007).
[2] N. E. Hussey, M. Abdel-Jawad, A. Carrington, A. P. Mackenzie, and L. Balicas, Nature (London) **425**, 814 (2003).
[3] D. LeBoeuf, N. Doiron-Leyraud, J. Levallois, R. Daou, J. B. Bonnemaison, N. E. Hussey, L. Balicas, B. J. Ramshaw, R. X. Liang, D. A. Bonn, W. N. Hardy, S. Adachi, C. Proust, and L. Taillefer, Nature (London) **450**, 533 (2007).
[4] R. Daou, N. Doiron-Leyraud, D. LeBoeuf, S. Y. Li, F. Laliberte, O. Cyr-Choiniere, Y. J. Jo, L. Balicas, J.-Q. Yan, J.-S. Zhou, J. B. Goodenough, and L. Taillefer, Nat. Phys. **5**, 31 (2009).
[5] L. Taillefer, J. Phys.: Condens. Matter **21**, 164212 (2009).
[6] O. Cyr-Choinière, R. Daou, F. Laliberte, D. LeBoeuf, N. Doiron-Leyraud, J. Chang, J.-Q. Yan, J.-G. Cheng, J.-S. Zhou, J. B. Goodenough, S. Pyon, T. Takayama, H. Takagi, Y. Tanaka, and Louis Taillefer, Nature (London) **458**, 743 (2009).
[7] J. Chang, Y. Sassa, S. Guerrero, M. Mansson, M. Shi, S. Pailhes, A. Bendounan, R. Mottl, T. Claesson, O. Tjernberg, L. Patthey, M. Ido, M. Oda, N. Momono, C. Mudry, and J. Mesot, New J. Phys. **10**, 103016 (2008).
[8] N. Ichikawa, S. Uchida, J. M. Tranquada, T. Niemoller, P. M. Gehring, S. H. Lee, and J. R. Schneider, Phys. Rev. Lett. **85**, 1738 (2000).
[9] M. v. Zimmermann, A. Vigliante, T. Niemöller, N. Ichikawa, T. Frello, J. Madsen, P. Wochner, S. Uchida, N. H. Andersen, J. M. Tranquada, D. Gibbs, and J. R. Schneider, Europhys. Lett. **41**, 629 (1998).
[10] T. Niemöller, N. Ichikawa, T. Frello, H. Hunnefeld, N. H. Andersen, S. Uchida, J. R. Schneider, and J. M. Tranquada, Eur. Phys. J. B **12**, 509 (1999).
[11] K. Behnia, D. Jaccard, and J. Flouquet, J. Phys.: Condens. Matter **16**, 5187 (2004).
[12] K. Miyake and H. Kohno, J. Phys. Soc. Jpn. **74**, 254 (2005).
[13] J. Takeda, T. Inukai, and M. Sato, J. Phys. Chem. Solids **62**, 181 (2001).
[14] I. M. Sutjahja, J. Aarts, A. A. Nugroho, M. Diantoro, M. O. Tjia, A. A. Menovsky, and J. J. M. Franse, Physica C **392-396**, 207 (2003).
[15] H. v. Löhneysen, A. Rosch, M. Vojta and P. Wölfle, Rev. Mod. Phys. **79**, 1015 (2007).
[16] R. B. Roberts, Philos. Mag. **36**, 91 (1977).
[17] F. Chen, J. C. Cooley, W. L. Hults, and J. L. Smith, Rev. Sci. Instrum. **72**, 4201 (2001).
[18] Y. Nakamura and S. Uchida, Phys. Rev. B **46**, 5841 (1992).
[19] C. Uher, A. B. Kaiser, E. Gmelin, and L. Walz, Phys. Rev. B **36**, 5676 (1987).
[20] M. V. Elizarova and V. E. Gasumyants, Phys. Rev. B **62**, 5989 (2000).
[21] H. J. Trodahl, Phys. Rev. B **51**, 6175 (1995).
[22] J.-S. Zhou and J. B. Goodenough, Phys. Rev. B **51**, 3104 (1995).
[23] J. L. Tallon, J. R. Cooper, P. S. I. P. N. de Silva, G. V. M. Williams, and J. W. Loram, Phys. Rev. Lett. **75**, 4114 (1995).
[24] H. v. Löhneysen, T. Pietrus, G. Portisch, H. G. Schlager, A. Schröder, M. Sieck, and T. Trappmann, Phys. Rev. Lett. **72**, 3262 (1994).
[25] H. Jin, A. Narduzzo, M. Nohara, H. Takagi, N. E. Hussey, and K. Behnia (unpublished).
[26] S. Nakamae, K. Behnia, N. Mangkorntong, M. Nohara, H. Takagi, S. J. C. Yates, and N. E. Hussey, Phys. Rev. B **68**, 100502(R) (2003).
[27] Y. Dagan, M. M. Qazilbash, C. P. Hill, V. N. Kulkarni, and R. L. Greene, Phys. Rev. Lett. **92**, 167001 (2004).
[28] P. Li, K. Behnia and R. L. Greene, Phys. Rev. B **75**, 020506(R) (2007).
[29] J. Lin and A. J. Millis, Phys. Rev. B **72**, 214506 (2005).
[30] P. Fournier, P. Mohanty, E. Maiser, S. Darzens, T. Venkatesan, C. J. Lobb, G. Czjzek, R. A. Webb, and R. L. Greene, Phys. Rev. Lett. **81**, 4720 (1998).
[31] E. M. Motoyama, G. Yu, I. M. Vishik, O. P. Vajk, P. K. Mang, and M. Greven, Nature (London) **445**, 186 (2007).
[32] I. Paul and G. Kotliar, Phys. Rev. B **64**, 184414 (2001).
[33] A. J. Millis and M. R. Norman, Phys. Rev. B **76**, 220503(R) (2007).
[34] J. Lin and A. J. Millis, Phys. Rev. B **78**, 115108 (2008).