Charge Order and Superconductivity in Underdoped $\mathrm{YBa_2Cu_3O_{7-\delta}}$ under Pressure

Carsten Putzke,[1] Jake Ayres,[1] Jonathan Buhot,[2] Salvatore Licciardello,[2] Nigel E. Hussey,[2]
Sven Friedemann,[1] and Antony Carrington[1]
[1]*H.H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, United Kingdom*
[2]*High Field Magnet Laboratory (HFML-EMFL), Radboud University, Toernooiveld 7, Nijmegen 6525 ED, Netherlands*

(Received 21 July 2017; published 16 March 2018)

In underdoped cuprates, an incommensurate charge density wave (CDW) order is known to coexist with superconductivity. A dip in T_c at the hole doping level where the CDW is strongest ($n_p \simeq 0.12$) suggests that CDW order may suppress superconductivity. We investigate the interplay of charge order with superconductivity in underdoped $\mathrm{YBa_2Cu_3O_{7-\delta}}$ by measuring the temperature dependence of the Hall coefficient $R_H(T)$ at high magnetic field and at high hydrostatic pressure. We find that, although pressure increases T_c by up to 10 K at 2.6 GPa, it has very little effect on $R_H(T)$. This suggests that pressure, at these levels, only weakly affects the CDW and that the increase in T_c with pressure cannot be attributed to a suppression of the CDW. We argue, therefore, that the dip in T_c at $n_p \simeq 0.12$ at ambient pressure is probably not caused by the CDW formation.

DOI: 10.1103/PhysRevLett.120.117002

Cuprate high-T_c superconductors have a rich phase diagram, and the underdoped region is particularly complex, with several distinct phases [1]. Elucidating the microscopic origin of these phases and their relation to the superconductivity remains of great importance. For a hole doping $n_p < 0.18$ and below a characteristic temperature T^*, a phase appears which is characterized by a reduction in the density of states close to the Fermi level [2]. The loss of states which characterizes this pseudogap phase is anisotropic, leading to disconnected arcs of the Fermi surface being observed in angle resolved photoemission spectroscopy [3]. Inside the pseudogap phase, an incommensurate charge density wave (CDW) phase is known to form [4,5]. For the cuprate $\mathrm{YBa_2Cu_3O_{7-\delta}}$ (Y123), the CDW has been detected over the doping range $0.09 \lesssim n_p \lesssim 0.16$ and has the largest x-ray intensity for $n_p \simeq 0.12$ [6]. Close to this point in the phase diagram, other anomalous behavior is observed—most importantly, a relative dip in the superconducting transition temperature T_c [7] producing the well-known 60 K plateau and a marked reduction in the upper critical field H_{c2} [8].

Given the correlation between the strengthening of the CDW at $n_p \simeq 0.12$ and the relative dip in T_c, it is natural to suppose [9] that superconductivity and CDW formation are mutually exclusive competing phases, so the suppression of one leads to the strengthening of the other. The x-ray intensity from the CDW modulation is strongly reduced when the temperature is reduced below T_c in zero magnetic field, but it is increased when superconductivity is suppressed by applying a large magnetic field [10]. This suggests that the CDW is attenuated when the electrons become paired; however, what is less clear is whether the inverse also holds. In other words, does the strengthening of the CDW lead to a suppression of the superconductivity—as is perhaps suggested by the dip in T_c at $n_p \simeq 0.12$—or does this dip have another origin and is the CDW then enhanced simply because superconductivity is suppressed?

To help answer this question, we have performed a series of measurements of the temperature dependence of the Hall coefficient $R_H(T)$ of underdoped Y123 ($n_p \simeq 0.11$) at magnetic fields of up to 36 T as a function of high hydrostatic pressure p. As described below, this allows us to vary T_c and thus to study the correlation between T_c and the CDW without changing the chemical composition.

In optimally doped and underdoped cuprates, the Hall coefficient has a strong and unusual temperature dependence. At high temperatures, $T \gtrsim 100$ K, R_H increases as T decreases, particularly for Y123, $R_H \sim 1/T$ at optimal doping [11,12]. At lower temperature, and for a doping range which approximately coincides with that where the CDW is observed with x rays, $R_H(T)$ displays a maximum and decreases strongly at lower temperature even when the applied magnetic field is sufficiently strong to suppress superconductivity [13]. For $0.08 \lesssim n_p \lesssim 0.15$, R_H changes sign and, at a sufficiently low temperature, becomes temperature independent [14]. In the same range of doping and again at high magnetic field, quantum oscillations (QOs) are observed [15–18]. The low frequency of the QO signal suggests that the Fermi surface undergoes reconstruction, the most likely cause of which is the CDW [19]. Therefore, the temperature dependence of $R_H(T)$ is a sensitive probe of the CDW state and can be used to track its evolution with temperature, field, and pressure.

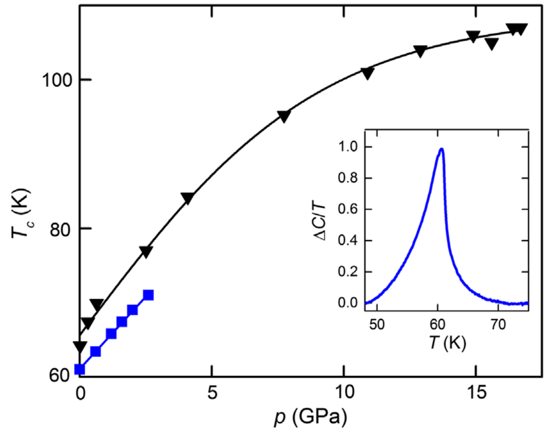

FIG. 1. Superconducting transition temperature vs pressure for the sample used in our work (the squares) compared to the behavior reported by Sadewasser *et al.* [22] (the triangles) for a similar doping. The lines are guides for the eye. (Inset) The measured heat capacity at ambient pressure of our sample close to T_c after the removal of a smooth background and scaled to unity at T_c.

Hydrostatic pressure provides a very useful tuning parameter with which to explore the phase diagram of the cuprates. It can be used to tune T_c in a single sample without introducing the disorder associated with chemical changes in the oxygen content or cation stoichiometry. Hydrostatic pressure increases T_c most rapidly for doping close to $n_p \simeq 0.12$ [9,20]. The increase in T_c is linear at low p and then tends to saturate at higher pressure, with T_c eventually reaching ~107 K at ~17 GPa (Fig. 1). The exact mechanism of this T_c enhancement is not clear. The thermoelectric power (TEP) decreases with increasing pressure [21] suggesting that there is some pressure-induced doping, but this cannot be the sole mechanism because the maximum T_c obtained as a function of pressure is a strong function of n_p [22]. An analysis of the rate of pressure-induced charge transfer suggested by the accompanying changes in the TEP, Knight shift, and quantum oscillation frequency in the sister compound to Y123, $YBa_2Cu_4O_8$ (Y124), suggests that about half of the enhancement of $T_c(p)$ comes from charge transfer [23].

Our measurements of R_H were performed on single crystals of Y123 which were grown by the standard flux technique [24] and annealed at 580 °C in a 2% oxygen in nitrogen mixture for 8 days followed by a rapid quench to room temperature. This resulted in samples with $n_p \simeq 0.11$, as estimated from T_c [7]. The width and homogeneity of the bulk superconducting transition was determined using heat capacity, measured with a membrane calorimeter [25]. A sharp superconducting transition at ambient pressure, $T_c^0 = 60.7 \pm 0.3$ K, was found (see the inset in Fig. 1). Electrical contacts were applied prior to annealing using a combination of sputtered Au and Dupont 4929 silver epoxy. Care was taken to sputter Au along the whole of the c-axis thickness of the sample to ensure homogeneous current flow and accurate values of R_H. The samples were mounted inside a piston cylinder pressure cell made from the high strength, nonmagnetic [26] alloy and cooled by a helium cryostat inside a Bitter magnet at the High Field Magnet Laboratory in Nijmegen, Netherlands. R_H and longitudinal magnetoresistance were measured simultaneously by sweeping the field between the positive and negative extremes at various fixed temperatures. The Hall resistivity was then calculated from the odd part of the magnetoresistance measured between contacts placed on opposite sides of the sample. Once inside the pressure cell, T_c was determined to be the temperature where the resistivity falls below 1% of the normal state value. At ambient pressure, this coincides with the peak in the heat capacity, and the evolution with pressure is in good agreement with a previous report for a similar doping [22] (see Fig. 1). The results obtained here were repeated in a second sample with a slightly higher value of n_p [27].

The field evolution of R_H at fixed temperatures in fields up to 36 T is shown in Fig. 2. At temperatures well above T_c, R_H is essentially independent of field. Below T_c, both the resistivity and the R_H value are zero below the irreversibility field H_{irr}, and R_H acquires a finite negative value and tends towards a constant value at the highest fields. As temperature is decreased, the field induced transition becomes progressively sharper. As pressure is increased, the main change to this behavior is that H_{irr} increases, so the increase of $|R_H|$ from zero below H_{irr} occurs at a higher field. Hence, the lowest temperature where we can approach the normal state value with the available field increased with applied pressure from 4.2 K at ambient pressure to ~25 K at 2.6 GPa.

In Fig. 3 the temperature evolution of this high field R_H value shows a characteristic sign change that is highly suggestive of a reconstruction of the Fermi surface [13]. It evolves from a holelike positive value at high temperature

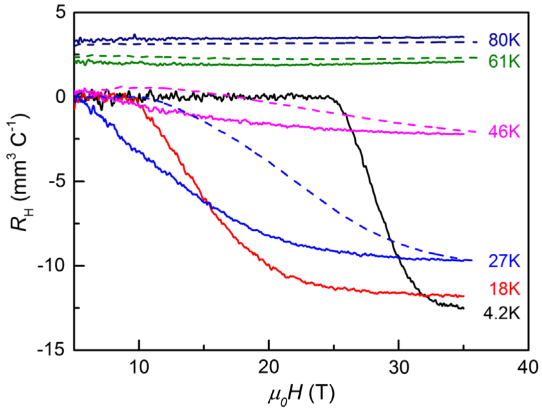

FIG. 2. Hall coefficient vs field for Y123 with $n_p = 0.11$ ($T_c^0 = 60.7$ K) at ambient pressure (the solid lines) and $p = 2.6$ GPa (the dashed lines) ($T_c = 71$ K). The temperature labels refer to the ambient pressure. The temperatures for the $p = 2.6$ GPa data were 80, 61, 43, and 25 K, respectively.

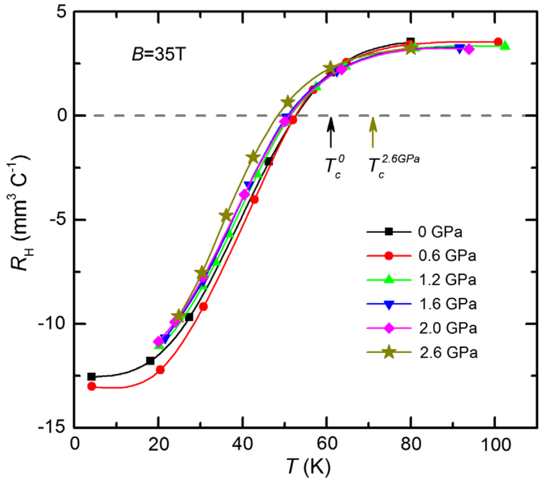

FIG. 3. Hall coefficient at $B = 35$ T vs temperature for Y123 with $n_p = 0.11$ at applied pressures of up to 2.6 GPa. The lines are guides for the eye. The two arrows mark the zero field T_c at ambient pressure and at 2.6 GPa, respectively.

to one which is dominated by an electronlike negative contribution at low temperature. Below ~20 K, R_H tends to a temperature independent value.

Figure 3 also shows our central result, namely, that pressure has only a small effect on the temperature evolution of R_H. There is a weak decrease in the temperature T_0 where R_H changes sign ($dT_0/dp = -1.1 \pm 0.2$ K/GPa [27]), which is small compared to the increase in T_c, which is 10 K (3.8 K/GPa) over this same pressure interval (as marked by the arrows in the figure). This then suggests that the CDW is not strongly affected by pressure up to at least 2.6 GPa, and hence it is unlikely that any pressure-induced weakening of the CDW contributes significantly to the pressure evolution of T_c.

In other CDW systems, it is usually found that pressure does suppress CDW order. This can be understood both from the influence of pressure on the lattice and on the electronic structure: (i) pressure generally leads to an increase of the lattice stiffness and hence raises the phonon frequencies, leading to a decrease in electron-phonon coupling and weakening CDW order; (ii) pressure increases orbital overlap and thus leads to a more isotropic and 3D Fermi surface topology, thus weakening any Fermi surface nesting underlying CDW order. The effect of pressure on the CDW can be highly nonlinear. For example, in 2H-NbSe$_2$ there is only a weak decrease in the CDW transition temperature with increasing pressure until close to the critical pressure [28]. For the Y123 system, this critical pressure appears to be well above our maximum of 2.6 GPa. As $T_c(p)$ evolves monotonically up to the highest pressures and has a maximum slope at ambient pressure [22] (Fig. 1), we conclude that suppression of the CDW with pressure does not contribute significantly to $T_c(p)$. If it did, we would then expect an increase in the slope of $T_c(p)$ where the CDW is suppressed at some higher pressure, which is not observed.

An important consideration in interpreting our data is the nature of the CDW in Y123 at high fields. X-ray studies have shown that, at high temperature and/or low field, the structure is essentially two dimensional with a short c-axis coherence length. Below T_c, this 2D order is suppressed, but it is restored as superconductivity is weakened by a magnetic field [10]. At high fields the b- and c-axis CDW coherence lengths grow in a precursor region, and a different three-dimensional structure is eventually formed *in addition to* the 2D order [29,30]. The 2D order above T_c is independent of the field [10]. The sign change of R_H begins at high temperatures and low fields, with T_0 essentially being field independent [13,14], so it is likely caused by the 2D CDW order rather than the 3D component. This is also consistent with models which give a closed electron pocket for reconstruction based on the 2D, approximately 3×3 biaxial order, but not for the uniaxial 3D order [31–33]. Therefore, although our measurements of R_H are necessarily performed at high field, to suppress the superconductivity, they should be expected to probe the evolution of the same 2D CDW order which has been suggested to cause a dip in T_c at $n_p \simeq 0.12$ at zero field.

In the scenario where the CDW reduces T_c, if the CDW were suppressed at low pressure, we would expect T_c to rise with pressure. Then, if the pressure-suppressed CDW were to reemerge in high field, we would expect a marked decrease in the irreversibility field compared to the behavior at ambient pressure. In Fig. 4 we show that $H_{\mathrm{irr}}(T)$ follows a common behavior for all pressures, when scaled to account for the changes in T_c and $H_{\mathrm{irr}}(T = 4\ \mathrm{K})$ with pressure. Thus, we conclude that either the CDW is not stabilized by a high field or else superconductivity is not affected by the CDW.

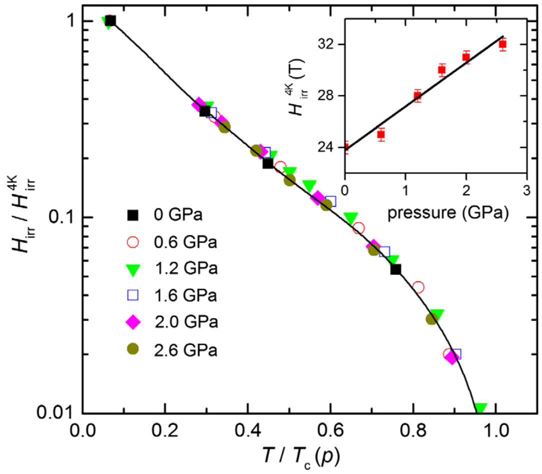

FIG. 4. Evolution of the irreversibility field H_{irr} with pressure and temperature plotted on scaled axes. The scaling field $H_{\mathrm{irr}}^{4\mathrm{K}}$ was determined to best scale the data to the ambient pressure result, and its evolution with pressure is shown in the inset. The line is a guide for the eye.

The evolution of $R_H(T)$ with pressure is distinctly different from that observed as a function of doping. Our value of T_0 for $n_p = 0.11$ is consistent with that reported previously [14]. However, as T_c is increased by oxygen doping, it is found that T_0 increases sharply, by more than 20 K between $n_p = 0.11$ and $n_p = 0.12$ (which corresponds to an increase in T_c of around 6 K). By contrast, here we find only a small *decrease* in T_0 when T_c is increased by 10 K with pressure. The value of R_H at low temperature was also found to be a strong function of n_p [14], whereas here again we see only a very small change as a function of pressure.

Our observation that the CDW is only weakly affected by pressure is consistent with a previous study of Y124 [23]. In Y124, with $n_p \simeq 0.13(1)$ and $T_c^0 = 79$ K, quantum oscillations were observed up to a pressure of 0.84 GPa. The frequency of the oscillations increased slowly and monotonically with an increasing p, suggesting a gradual evolution of the CDW reconstruction potential rather than any profound change in its structure. Furthermore, the effective mass m^* was found to decrease with an increasing p. Within the interpretation that the enhancement of m^* is caused by quantum CDW fluctuations, the decrease in mass would suggest that these fluctuations are reduced by pressure.

A further point to note is that, although the change in sign of R_H approximately coincides with T_c at ambient pressure, the fact that its temperature dependence remains unaltered despite T_c increasing by more than 10 K shows that superconducting fluctuation effects and vortex motion, which have previously been suggested as a cause of the R_H sign change [34], play very little role. Hence, our results provide further proof that the sign change of R_H is a normal state property.

In summary, we have shown that the sign change of the normal state Hall coefficient of $\mathrm{YBa_2Cu_3O_{7-\delta}}$ with a doping of $n_p = 0.11$ ($T_c = 60.7$ K at ambient pressure) is only weakly affected by hydrostatic pressure up to 2.6 GPa, despite T_c being increased by 10 K. The results suggest that the charge ordered phase is not destabilized by pressures in this range and that CDW destruction does not play a significant role in the observed increase in T_c with pressure. It is unlikely that the dip in T_c, characterized by a 60 K plateau, is caused by the CDW formation, and it is rather likely that it has another origin. The strengthening of the CDW at this doping is simply a consequence of the T_c dip.

We thank S. M. Hayden and J. R. Cooper for useful discussions. This work was supported by the Engineering and Physical Sciences Research Council (Grants No. EP/K016709/1, No. EP/L015544/1, No. EP/N026691/1, No. EP/N01085X/1, and No. NS/A000060/1), and by the High Field Magnet Laboratory – Radboud University/Foundation for Fundamental Research on Matter (HMFL-RU/FOM)—a member of the European Magnetic Field Laboratory.

[1] B. Keimer, S. A. Kivelson, M. R. Norman, S. Uchida, and J. Zaanen, Nature (London) **518**, 179 (2015).

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

[3] M. R. Norman, H. Ding, M. Randeria, J. C. Campuzano, T. Yokoya, T. Takeuchi, T. Takahashi, T. Mochiku, K. Kadowaki, P. Guptasarma, and D. G. Hinks, Nature (London) **392**, 157 (1998).

[4] T. Wu, H. Mayaffre, S. Kramer, M. Horvatic, C. Berthier, W. N. Hardy, R. X. Liang, D. A. Bonn, and M. H. Julien, Nature (London) **477**, 191 (2011).

[5] G. Ghiringhelli *et al.*, Science **337**, 821 (2012).

[6] M. Hucker, N. B. Christensen, A. T. Holmes, E. Blackburn, E. M. Forgan, R. X. Liang, D. A. Bonn, W. N. Hardy, O. Gutowski, M. von Zimmermann, S. M. Hayden, and J. Chang, Phys. Rev. B **90**, 054514 (2014).

[7] R. X. Liang, D. A. Bonn, and W. N. Hardy, Phys. Rev. B **73**, 180505 (2006).

[8] G. Grissonnanche *et al.*, Nat. Commun. **5**, 3280 (2014).

[9] O. Cyr-Choinière, D. LeBoeuf, S.Badoux, S. Dufour-Beauséjour, D. A. Bonn, W. N. Hardy, R. Liang, N. Doiron-Leyraud, and L. Taillefer, arXiv:1503.02033.

[10] J. Chang, E. Blackburn, A. T. Holmes, N. B. Christensen, J. Larsen, J. Mesot, R. X. Liang, D. A. Bonn, W. N. Hardy, A. Watenphul, M. von Zimmermann, E. M. Forgan, and S. M. Hayden, Nat. Phys. **8**, 871 (2012).

[11] A. Carrington, A. P. Mackenzie, C. T. Lin, and J. R. Cooper, Phys. Rev. Lett. **69**, 2855 (1992).

[12] A. Carrington, D. J. C. Walker, A. P. Mackenzie, and J. R. Cooper, Phys. Rev. B **48**, 13051 (1993).

[13] 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).

[14] D. LeBoeuf, N. Doiron-Leyraud, B. Vignolle, M. Sutherland, B. J. Ramshaw, J. Levallois, R. Daou, F. Laliberté, O. Cyr-Choinière, J. Chang, Y. J. Jo, L. Balicas, R. Liang, D. A. Bonn, W. N. Hardy, C. Proust, and L. Taillefer, Phys. Rev. B **83**, 054506 (2011).

[15] N. Doiron-Leyraud, C. Proust, D. LeBoeuf, J. Levallois, J. B. Bonnemaison, R. X. Liang, D. A. Bonn, W. N. Hardy, and L. Taillefer, Nature (London) **447**, 565 (2007).

[16] A. F. Bangura, J. D. Fletcher, A. Carrington, J. Levallois, M. Nardone, B. Vignolle, P. J. Heard, N. Doiron-Leyraud, D. LeBoeuf, L. Taillefer, S. Adachi, C. Proust, and N. E. Hussey, Phys. Rev. Lett. **100**, 047004 (2008).

[17] E. A. Yelland, J. Singleton, C. H. Mielke, N. Harrison, F. F. Balakirev, B. Dabrowski, and J. R. Cooper, Phys. Rev. Lett. **100**, 047003 (2008).

[18] B. J. Ramshaw, S. E. Sebastian, R. D. Mcdonald, J. Day, B. S. Tan, Z. Zhu, J. B. Betts, R. X. Liang, D. A. Bonn, W. N. Hardy, and N. Harrison, Science **348**, 317 (2015).

[19] S. E. Sebastian, N. Harrison, and G. G. Lonzarich, Rep. Prog. Phys. **75**, 102501 (2012).

[20] R. Benischke, T. Weber, W. H. Fietz, J. Metzger, K. Grube, T. Wolf, and H. Wuhl, Physica (Amsterdam) **203C**, 293 (1992).

[21] J. S. Zhou and J. B. Goodenough, Phys. Rev. Lett. **77**, 151 (1996).

[22] S. Sadewasser, J. S. Schilling, A. P. Paulikas, and B. W. Veal, Phys. Rev. B **61**, 741 (2000).

[23] C. Putzke, L. Malone, S. Badoux, B. Vignolle, D. Vignolles, W. Tabis, P. Walmsley, M. Bird, N. E. Hussey, C. Proust, and A. Carrington, Sci. Adv. **2**, e1501657 (2016).

[24] E. Stupp and D. M. Ginsberg, in *Physical Properties of High Temperature Superconductors III*, edited by D. M. Ginsberg (World Scientific, Singapore, 1992).

[25] Membrane calorimeter model number XEN-39397, supplied by Xensor Integration (http://www.xensor.nl).

[26] Standard Pressed Steel Technologies MP35N.

[27] See the Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.117002 for the results for the second sample and the detailed pressure dependence of T_0 for sample 1.

[28] C. Berthier, P. Molinie, and D. Jerome, Solid State Commun. **18**, 1393 (1976).

[29] S. Gerber *et al.*, Science **350**, 949 (2015).

[30] J. Chang, E. Blackburn, O. Ivashko, A. T. Holmes, N. B. Christensen, M. Hucker, R. Liang, D. A. Bonn, W. N. Hardy, U. Rutt, M. von Zimmermann, E. M. Forgan, and S. M. Hayden, Nat. Commun. **7**, 11494 (2016).

[31] N. Harrison and S. E. Sebastian, New J. Phys. **14**, 095023 (2012).

[32] A. J. Millis and M. R. Norman, Phys. Rev. B **76**, 220503 (2007).

[33] O. Cyr-Choinière, S. Badoux, G. Grissonnanche, B. Michon, S. A. A. Afshar, S. Fortier, D. LeBoeuf, D. Graf, J. Day, D. A. Bonn, W. N. Hardy, R. Liang, N. Doiron-Leyraud, and L. Taillefer, Phys. Rev. X **7**, 031042 (2017).

[34] J. M. Harris, Y. F. Yan, O. K. C. Tsui, Y. Matsuda, and N. P. Ong, Phys. Rev. Lett. **73**, 1711 (1994).