Exploring competing orders in cuprate superconductors

Clifford W. Hicks#, Andrew P. Mackenzie##

The microscopic structure of the fluctuating states that bring about high-temperature superconductivity remains unknown. Fluctuations of antiferromagnetic order is almost certainly an essential part, but the degree to which charge structure may also be essential to the problem of cuprate superconductivity is not known. Through uniaxial stress, quasi-long-range charge density wave order can be induced in $YBa_2Cu_3O_{6.67}$ under zero applied magnetic field. The way in which this state interacts with the superconductivity provides essential information on the superconducting state itself.

In the doping-temperature phase diagram of hole-doped cuprates there is always an antiferromagnetic phase at low doping and a dome of superconductivity at higher doping. This structure led early on to a hypothesis that antiferromagnetic fluctuations drive the superconductivity [1]. However, doping is only one possible tuning axis. Hole-doped cuprates also show a ubiquitous tendency towards charge order. If the superconductivity is found generally to exist in proximity to both antiferromagnetism and charge order, along distinct tuning axes, it then becomes unclear whether one should look first to fluctuations of the magnetism or the charge order to explain the superconductivity [2].

In unstressed $YBa_2Cu_3O_{6.67}$, below ≈ 150 K there is static charge order that has a very short in-plane correlation length, and is almost completely uncorrelated from layer to layer; this is typically termed the 2D charge order [3]. $YBa_2Cu_3O_{6.67}$ has $p \approx 1/8$ holes per Cu site in the CuO_2 planes, a doping that favors charge order. Under strong magnetic field, which weakens the superconductivity, 3D-correlated charge order appears, with a considerably longer in-plane and much longer inter-plane correlation length [4]. One hypothesis is that this occurs because "patches" of the 2D order lock together when it becomes sufficiently strong, but this interpretation has not been proved.

In Fig. 1, we show X-ray data on $YBa_2Cu_3O_{6.67}$, which reveal that ≈ 1% uniaxial compression along the *a* axis (that is, along the shorter axis, and so increasing the orthorhombicity of the CuO_2 planes) also induces 3D-correlated charge order [5]. What makes this demonstration especially valuable is that stress is homogenous, whereas magnetic field induces a mixed phase. The coexistence of superconductivity and 3D charge order under applied field [6] might, therefore, be a consequence of charge order condensing in vortex cores [7] rather than intrinsic coexistence. Uniaxial stress offers the possibility of understanding the intrinsic behavior. In Fig. 1(b), we show that under 1% compression the 3D peak appears only between ≈40 and ≈70 K. Its disappearance as temperature is reduced indicates that it does not coexist with superconductivity.

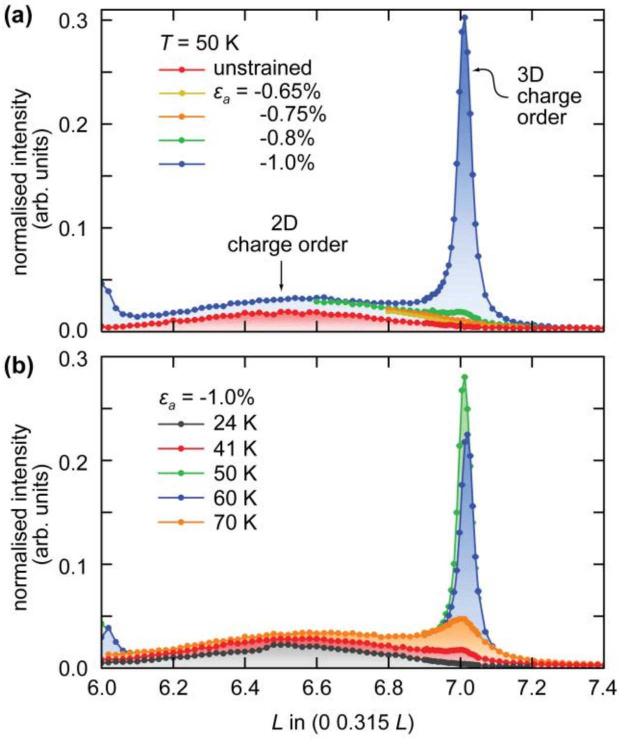

Fig. 1: Elastic X-ray scattering data showing the stress-induced onset of 3D-correlated charge order in $YBa_2Cu_3O_{6.67}$. (a) In both the unstressed sample, and when it is compressed by ≈1% along the a axis, there is a broad peak corresponding to 2D-correlated charge order. Under 1% compression, the 3D peak appears. (b) At $\varepsilon_a \approx -1\%$, the 3D charge order is present only between ≈ 40 and 70 K, indicating that it does not coexist with superconductivity.

The 2D charge order has been observed to weaken when superconductivity onsets, which indicates that it competes with superconductivity [8]. However, the reverse effect, of the charge order on the superconductivity, has not been conclusively demonstrated. It is important to do so to understand whether the charge order could be a form of "alternative" order to the superconductivity, driven by the same interactions. In some La-based compounds there is clear suppression of superconductivity as a consequence of the presence of charge order. Most famously, in $La_{2-x}Ba_xCuO_4$ T_c drops to ≈ 4 K right in the vicinity of x = 0.125, where static charge order appears [9]. But the phenomenology of charge order is different between the La-based compounds and $YBa_2Cu_3O_{6+x}$. In the La-based compounds, it coexists with spin order to form spin-charge stripes, whereas in $YBa_2Cu_3O_{6+x}$ spin and charge order mutually compete [10]. Therefore, the charge order of $YBa_2Cu_3O_{6+x}$ could interact with the superconductivity in a qualitatively different way.

To determine whether onset of 3D charge order in $YBa_2Cu_3O_{6.67}$ affects the superconductivity, the dependence of T_c on uniaxial stress was measured at MPI-CPfS. Results are shown in Fig. 2(a). We note that to achieve precision at high stresses, a number of technical steps were required. One was use of a susceptometer microfabricated from a GaAs/AlGaAs 2DEG, in order to measure T_c over a small length scale, which avoids averaging over long-lengh-scale inhomogeneity. Another was use of a stress cell developed at MPI-CPfS that incorporates both force and displacement sensors, allowing the state of the epoxy that holds the sample to be monitored [11]. In order to place samples that were compressed along the *a* axis on the same plot as one that was compressed along the *b* axis, data are plotted against $\sigma_a - \sigma_b$, where σ_a is stress along the *a* axis and negative values denote compression. At low $|\sigma_a - \sigma_b|$, T_c is seen to have an approximately quadratic dependence on uniaxial stress. For σ_a below about -1 GPa, however, the dependence changes to linear. The red stars in Fig. 2(a) mark the maximum curvature $d^2T_c/d\sigma_a^2$, which we identify as the approximate crossover point from quadratic to linear behavior.

The crossover to linear behavior occurs at a similar stress to the onset of 3D charge order shown in Fig. 1. Furthermore, because the 3D charge order is uniaxial, it is expected to couple linearly to uniaxial stress. Competition between the 3D order and superconductivity would therefore be expected to drive a stress-linear suppression of T_c. The data of Figs. 1(b) and 2(a) therefore show that the 3D order and superconductivity compete: onset of superconductivity suppresses the 3D order, and onset of the 3D order suppresses T_c. The precision of the data so far does not, strictly, rule out a range of coexistence; this will be a

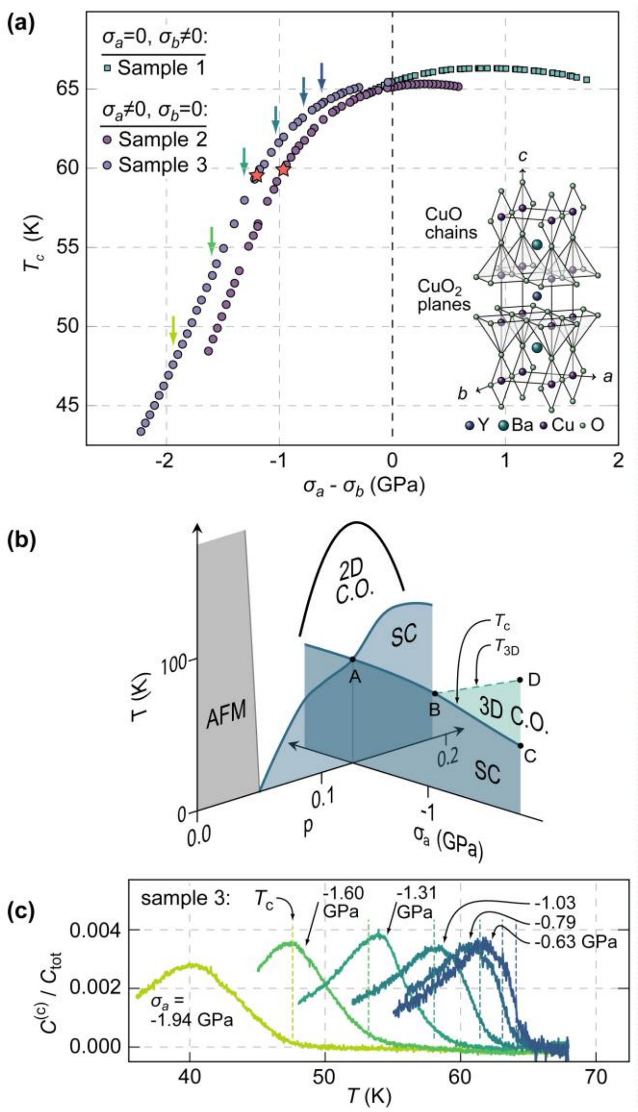

Fig. 2: (a) T_c versus uniaxial stress for three samples of $YBa_2Cu_3O_{6.67}$. T_c was measured through magnetic susceptibility. Uniaxial compression along the a axis drives a steep, stress-linear suppression of T_c, when the stress $|\sigma_a|$ exceeds ~ 1 GPa. The red stars mark maximum curvature $d^2T_c/d\sigma_a^2$, which we take as the crossover points from quadratic to linear stress dependence. (b) Stress-temperature-doping phase diagram of $YBa_2Cu_3O_{6+x}$ as resolved so far. T_{3D} has not yet been resolved in measurements. (c) Heat capacity anomaly at T_c of sample 3 measured through the elastocaloric effect, at various applied stresses. The stresses are indicated by the arrows in panel (a). $C^{(c)}$ is the critical part of the heat capacity, associated with the onset of the superconductivity, while C_{tot} is the total heat capacity.

topic for future inquiry. The stress-temperature-doping phase diagram of $YBa_2Cu_3O_{6+x}$, based on measurements performed so far, is shown in Fig. 2(b).

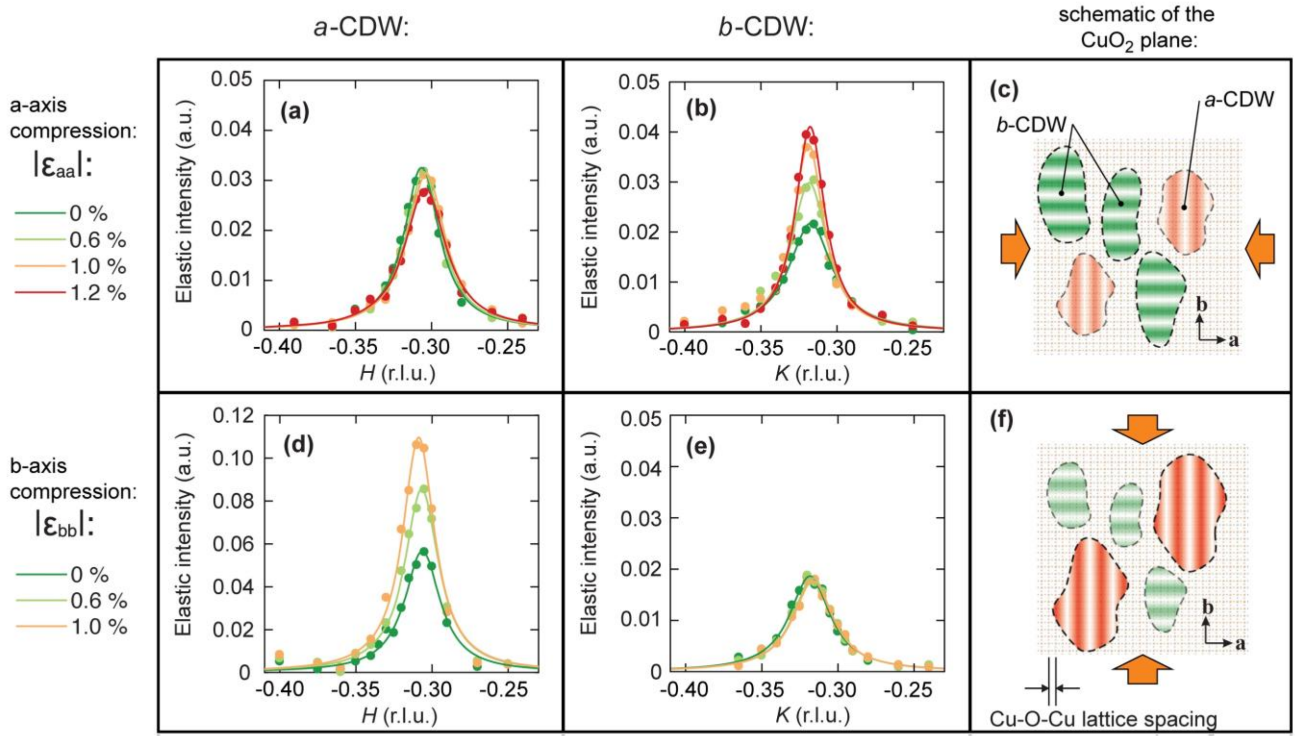

Fig. 3. Resonant x-ray data on the 2D charge order of $YBa_2Cu_3O_{6.67}$ under uniaxial stress. (a) Cuts through the a-axis-oriented component, and (b) through the b-axis-oriented component. (c) Schematic representation of the charge order in real space. Patches of charge order are indicated, and their size corresponds to the observed correlation lengths. (d–f) Equivalent data for b-axis compression.

We investigated this competition further through elastocaloric effect measurements. In $La_{2-x}Ba_xCuO_4$, a very small uniaxial stress can suppress static stripe order and dramatically raise T_c [12], suggesting that the static stripe order is an alternative order to the superconductivity, in the sense of having nearly the same onset temperature and likely a very similar energy. If the 3D charge order of $YBa_2Cu_3O_{6+x}$ has a similarly close relationship to the superconductivity, then the heat capacity anomaly along the transition between 3D charge order and superconductivity might be very small. That is, in the phase diagram of Fig. 2(b), the dominant heat capacity anomaly might follow the line A-B-D, with only a small residual anomaly along the ling B-C. The heat capacity anomaly can be measured through the elastocaloric effect [13], the change in temperature with applied strain. Results are shown in Fig. 2(c): the heat capacity anomaly at T_c does not change substantially as the 3D charge order onsets. In other words, although the 3D charge order interacts with the superconductivity strongly enough to suppress T_c, it probably does not constitute a form of "failed superconductivity" in the manner of stripe order in $La_{2-x}Ba_xCuO_4$.

How does the 2D charge order respond to uniaxial stress? The 2D charge order, unlike the 3D order, is biaxial. In Fig. 3 we show results of a resonant x-ray scattering study in which the effects of *a*-axis and *b*-axis compression are compared [14]. The separate effects of the applied stress on the *a*- and *b*-oriented CDW components are shown, and what the data reveal is a qualitative symmetry: compression along the *a* axis strengthens the *b*-CDW component, while compression along the *b* axis strengthens the *a*-CDW component. It is particularly notable just how sharp the *a*-CDW peak becomes under σ_b = -1 GPa: its correlation length at this stress well exceeds that of the *b*-CDW when, under *a*-axis compression, 3D charge order appears. This observation is not straightforwardly consistent with one major model of the 3D charge order [15], in which it appears when the correlation length of 2D patches exceeds a threshold, allowing them to lock together. Potentially, the chain layers hinder formation of an *a*-axis-oriented 3D CDW.

The Hall effect is commonly interpreted as a good indication of the onset of charge order: In $YBa_2Cu_3O_{6.67}$, the 2D order onsets at $T_{CDW} \approx 150$ K, and at this temperature the Hall coefficient also starts to deviate from its high-temperature trend [16]. The capability to tune the charge order with uniaxial stress allows us to test this hypothesis. We show preliminary data in Fig. 4. In this preliminary study the sample was compressed along the *b* axis, which, at least within the stress range accessed so far, does not induce 3D order. In Fig. 4(a), it is seen that the Hall coefficient barely changes between σ_b = 0 and -1.2 GPa, which could indicate that, despite the rapid grown in correlation of the *a*-CDW shown in Fig. 3(d), the overall strength of the 2D charge order is not altered much by this applied stress, or that the Hall effect is in reality not much affected by the presence charge order.

In Fig. 4(b) we show elastoresistivity data. The elastoresistivity starts to deviate from its high-temperature trend at ≈ 150 K, that is, at the approximate onset temperature of the 2D charge order. At lower temperatures the elastoresistivity becomes quite strong, and positive ($dR/d\sigma > 0$), meaning that compression along the *b* axis causes the *b* axis resistivity to decrease. (Recall that $\sigma < 0$ denotes compression.) The explanation might lie in the behavior shown in Fig. 3. It is seen there that compression along the *b* axis strengthens the *a*-CDW, which would be expected to increase scattering along the *a* axis. The *b*-axis resistivity might fall as a counterpart to this. Potentially the *b*-CDW weakens as the *a*-CDW strengthens, though this was not resolved in the X-ray data.

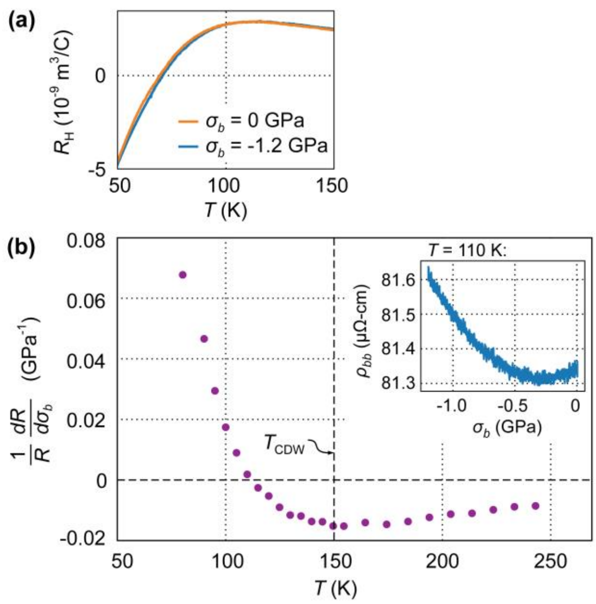

Fig. 4. Preliminary transport data on uniaxially stressed $YBa_2Cu_3O_{6.67}$. Here, pressure is applied along the b axis. (a) The Hall coefficient. Almost no change is observed between σ_b = 0 and -1.2 GPa. (b) The elastoresistivity. Above 110 K, the elastoresistivity has the "wrong" sign: resistivity increases *when the sample is compressed. The elastoresistivity is seen to deviate from the high-temperature trend starting at $T_{CDW} \approx 150$ K.*

There is much more work to do on the cuprate superconductors. One major task is to investigate the extent of the 3D charge order phase at other dopings: is it something unique to $p \approx 0.125$, which would argue against a general connection to superconductivity, or can it be induced at all dopings, with sufficiently strong uniaxial stress? Another task is to investigate the thermodynamic consequences of the 3D order, for example through measurement of the stress-strain relationship, which has been made possible through development of a new stress cell at MPI-CPfS [11], and which is described for Sr_2RuO_4 in https://www1.cpfs.mpg.de:2443/PQM_01. We have demonstrated so far application of uniaxial stresses strong enough to alter the structure of electronic order in a cuprate superconductor, which will allow in time a detailed investigation of the fundamental requirements for high temperature superconductivity.

External Cooperation Partners

Prof. Bernhard Keimer (MPI-FKF, Stuttgart); Prof. Matthieu Le Tacon (Karlsruhe Institute of Technology).

References

[1] *Weak-coupling theory of high-temperature superconductivity in the antiferromagnetically correlated copper oxides, P. Monthoux, A. V. Balatsky, and D. Pines, Phys. Rev. B* ***46*** *(1992) 14803,* *doi.org/10.1103/PhysRevB.46.14803*

[2] *Electronic liquid-crystal phases of a doped Mott insulator, S. A. Kivelson, E. Fradkin, and V. J. Emery, Nature* ***393*** *(1998) 551, doi.org/10.1038/31177*

[3] *Long-range incommensurate charge fluctuations in (Y, Nd)$Ba_2Cu_3O_{6+x}$, G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N. B. Brookes, G. M. De Luca, A. Frano, D. G. Hawthorn, F. He, et al., Science* ***337*** *(2012) 821,* *doi.org/10.1126/science.1223532*

[4] *Three-dimensinoal charge density wave order in $YBa_2Cu_3O_{6.67}$ at high magnetic fields, S. Gerber, H. Jang, H. Nojiri, S. Matsuzawa, H. Yasumura, D. A. Bonn, R. Liang, W. N. Hardy, Z. Islan, A. Mehta, et al., Science* ***350*** *(2015) 949, doi.org/10.1126/science.aac6257*

[5]* *Uniaxial pressure control of competing orderes in a high-temperature superconductor, H.-H. Kim, S. M. Souliou, M. E. Barber, E. Lefrançois, M. Minola, M. Tortora, R. Heid, N. Nandi, R. A. Borzi, G. Garbarino, et al., Science* ***362*** *(2018) 1040, doi.org/10.1126/science.aat4708*

[6] *Thermodynamic phase diagram of static charge order in underdoped $YBa_2Cu_3O_y$, D. LeBoeuf, S. Krämer, W. N. Hardy, R. Liang, D. A. Bonn, and C. Proust, Nat. Phys.* ***9*** *(2013) 79.*

[7] *A four unit cell periodic pattering of quasiparticle states surrounding vortex cores in* $Bi_2Sr_2CaCu_2O_{8+\delta}$, *J. E. Hoffman, E. W. Hudson, K. M. Lang, V. Madhavan, H. Eisaki, S. Uchida, and J. C. Davis, Science* ***295*** *(2002) 466,* doi.org/10.1126/science.1066974
[8] *Direct observation of competition between superconductivity and charge density wave order in* $YBa_2Cu_3O_{6.67}$, *J. Chang, E. Blackburn, A. T. Holmes, N. B. Christensen, J. Larsen, J. Mesot, R. Liang, D. A. Bonn, W. N. Hardy, A. Watenphul, et al., Nat. Phys.* ***8*** *(2012) 871,* doi.org/10.1038/NPHYS2456
[9] *Stripe order in superconducting* $La_{2-x}Ba_xCuO_4$ *(*$0.095 \leq x \leq 0.155$*), M. Hücker, M. v. Zimmermann, G. D. Gu, Z. J. Xu, J. S. Wen, G. Xu, H. J. Kang, A. Zheludev, and J. M. Tranquada, Phys. Rev. B.* ***83*** *(2011) 104506,* doi.org/10.1103/PhysRevB.83.104506
[10] *Momentum-dependent charge correlations in* $YBa_2Cu_3O_{6+\delta}$ *superconductors probed by resonant x-ray scattering: evidence for three competing phases, S. Blanco-Canosa, A. Frano, T. Loew, Y. Lu, J. Porras, G. Ghiringhelli, M. Minola, C. Mazzoli, L. Braicovich, E. Schierle, et al., Phys. Rev. Lett.* ***110*** *(2013) 187001,* doi.org/10.1103/PhysRevLett.110.187001
[11]* *Piezoelectric-based uniaxial pressure cell with integrated force and displacement sensors, M. E. Barber, A. Steppke, A. P. Mackenzie, and C. W. Hicks, Rev. Sci. Inst.* ***90*** *(2019) 023904,* doi.org/10.1063/1.5075485
[12]* *Using uniaxial stress to probe the relationship between competing superconducting states in a cuprate with spin-stripe order, Z. Guguchia, D. Das, C. N. Wang, T. Adachi, N. Kitajima, M. Elender, F. Brückner, S. Ghosh, V. Grinenko, T. Shiroka, et al., Phys. Rev. Lett..* ***125*** *(2020) 097005,* doi.org/10.1103/PhysRevLett.125.097005
[13] *AC elastocaloric effect as a probe for thermodynamic signatures of continuous phase transitions, M. S. Ikeda, J. A. W. Straquadine, A. T. Hristov, T. Worasaran, J. C. Palmstrom, M. Sorensen, P. Walmsley, and I. R. Fisher, Rev. Sci. Inst.* ***90*** *(2019) 083902,* doi.org/10.1063/1.5099924
[14]* *Charge density waves in* $YBa_2Cu_3O_{6.67}$ *probed by resonant x-ray scattering under uniaxial compression, H.-H. Kim, E. Lefrançois, K. Kummer, R. Fumagalli, N. B. Brookes, D. Betto, S. Nakata, M. Tortora, J. Porras, T. Loew, et al., Phys. Rev. Lett.* ***126*** *(2021) 037002,* doi.org/10.1103/PhysRevLett.126.037002
[15] *Dimensional crossover of charge-density wave correlations in the cuprates, Y. Caplan and D. Orgad, Phys. Rev. Lett.* ***119*** *(2017)107002,* doi.org/10.1103/PhysRevLett.119.107002
[16] *Change of carrier density at the pseudogap critical point of a cuprate superconductor, S. Badoux, W. Tabis, F. Laliberté, G. Grissonnanche, B. Voignolle, D. Vignolles, J. Béard, D. A. Bonn, W. N. Hardy, R. Liang, et al., Nature* ***531*** *(2016) 210,* doi.org/10.1038/nature16983

[#] C.Hicks.1@bham.ac.uk; hicks@cpfs.mpg.de
[##] mackenzie@cpfs.mpg.de