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Uniaxial strain as an efficient way to decouple intertwined degrees of freedom in high-temperature cuprate superconductors

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Felix Bloch's description of a particle in a periodic lattice in 1930 defines an important cornerstone of modern condensed matter physics. It showed that quantum mechanics can be used to describe collective phenomena in solid state materials and led to microscopic descriptions of fundamental material properties in metals or insulators. These theories, however, treat the electrons as if they were independent quasiparticles, which is not valid for strongly correlated electron systems. These materials display a multitude of macroscopic quantum phenomena that we can understand only if we resolve the coupling among the electronic spin, charge, orbit and lattice degrees of freedom. One example is copper-oxide materials in which high-temperature superconductivity coexists with charge and spin order. Using uniaxial pressure as an external tuning parameter four recent publications led by Swiss researchers have brought new insight into the coupling among the various degrees of freedom in La-based cuprates, and how their fluctuations enable high-temperature superconductivity.

In correlated electron materials novel coherent phenomena and their inherent functional properties frequently arise from deeply intertwined electronic charge, spin, orbital and lattice degrees of freedom. Prominent examples are unconventional superconductivity, skyrmions, multiferroicity, materials with a giant magnetoresistance, or hidden order states. Although, the importance of coupled degrees of freedoms is widely appreciated, how this coupling is realized microscopically often remains a key issue. A model example is unconventional superconductivity in cuprate materials, where it is thought that the macroscopic zero-resistance state arises from charge (CDW) and spin-density wave (SDW) fluctuations [1–4]. To this date, however, it is unclear how the salient charge and spin degrees of freedom are coupled to enable high-temperature superconductivity.

Almost all high-temperature cuprate superconductors share a generic phase diagram such as shown in Fig. 1a for the case of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO). The inclusion of holes on the Cu-site effectively suppresses the correlated antiferromagnetic Mott state of La_2CuO_4 , and triggers a superconducting d -wave state for $0.05 < p < 0.28$. The superconducting state competes with long-range density-wave order, whose nature varies across the different cuprate families. In $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$, for instance, superconductivity coexists only with CDW order [5, 6]. In LSCO and $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ (LBCO), however, also SDW and signatures for pair-density wave order (a spatially modulated superconducting order parameter) are found [1–3]. It is thought that SDW and CDW order are connected in LSCO and LBCO, but the two orders possess a very different temperature and doping dependencies (see Fig. 1b). This raises the question how the different density-wave states are microscopically coupled in cuprate materials, and how their fluctuations enable the emergence of high-temperature superconductivity. Four peer reviewed

publications led by Swiss scientists have recently brought new insight into these questions [7–10].

Scattering techniques are a very efficient tool to study the electronic and lattice degrees of freedom in solid state materials. In La-based cuprates charge order appears at wavevectors \mathbf{Q}_{CDW} (in reciprocal lattice units (rlu)) which are offset by $\mathbf{q}_{\text{CDW}} = (\delta_{\text{CDW}}, 0, 0)$ and $(0, \delta_{\text{CDW}}, 0)$ with respect to structural Bragg peaks. SDW order is found at wavevectors \mathbf{Q}_{SDW} which are shifted by $\mathbf{q}_{\text{SDW}} = (\delta_{\text{SDW}}, 0, 0)$ and $(0, \delta_{\text{SDW}}, 0)$ away from the antiferromagnetic wavevector $\mathbf{Q}_{\text{AF}} = (1/2, 1/2, 0)$ with $\delta_{\text{SDW}} = 1/2\delta_{\text{CDW}} \approx 0.12$ for $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$. The structure

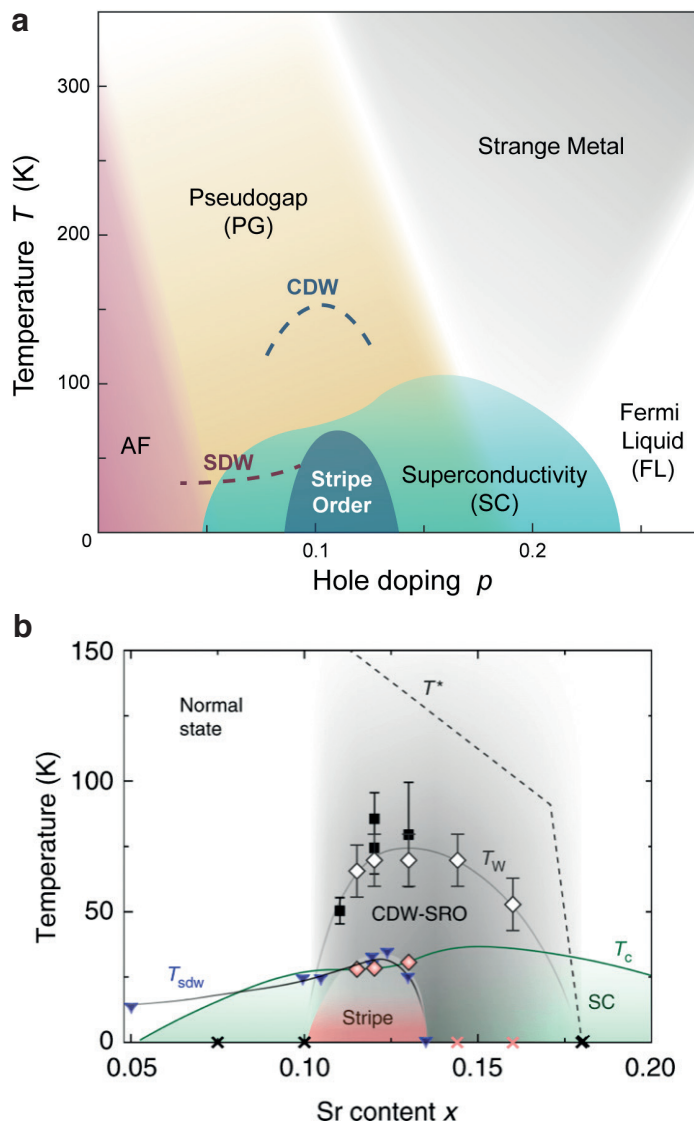


Fig. 1. Representative phase diagram of cuprate high superconductors. **a**: doping and temperature (pT) phase diagram of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, where p denotes the number of holes per Cu site (with courtesy of Jaewon Choi (Diamond Light Source - UK)). **b**: Close up of the phase diagram into the region for $0.05 < p < 0.2$. The holes were included into the system via Sr substitution on the La site (published in [3] under Creative Commons Attribution 4.0 International License).

of the charge-spin density-wave order is widely believed to consist of uniaxial charge-spin stripes [1, 2, 11–14] (see Fig. 2b), although other, checkerboard-type structures were also proposed in the past [15–24] (see Fig. 2a). In fact, regular diffraction experiments do not allow distinguishing the two cases (see Fig. 2a and b for the case of CDW order). This is because a multi-domain state with a single propagation vector in each domain triggers the same diffraction pattern as when two phase-related wavevectors coexist in a single domain. One way to differentiate between the two scenarios is to use an extrinsic tuning parameter that breaks the symmetry on a macroscopic scale [25, 26]. As shown in Fig. 2a and b a repopulation of the CDW/SDW domains is expected in the stripe-order case, whereas the two wavevectors are not expected to be affected differently in checkerboard structures. Three recent experiments have set out to clarify the structures and coupling between the CDW, SDW orders and the lattice in $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ [7–9], using uniaxial pressure cells that were designed at the University of Zurich to optimize the needs for each scattering experiment (see Fig. 2c).

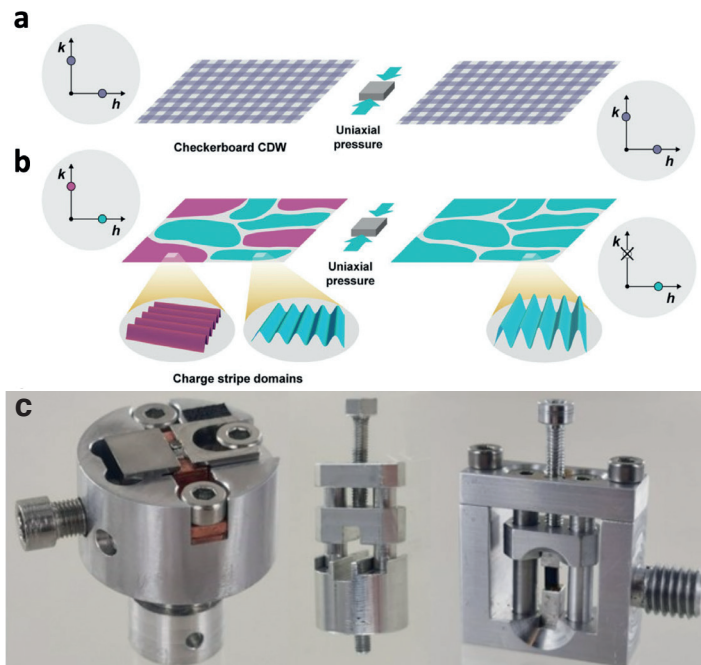


Fig. 2. Revealing the density-wave ground states via uniaxial pressure. **a** and **b**: Schematic representation of the two possible CDW ground states and how they are affected by uniaxial pressure (Reprinted figure with permission from [7]. © (2022) by the American Physical Society.). **c**: Uniaxial pressure devices used in [7–9]

Using elastic x-ray and neutron scattering an international collaboration led by researchers from the University of Zurich and the Paul Scherrer Institut studied the response of CDW and SDW order in $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ upon uniaxial pressure along the Cu-O bond direction [7, 8] (the tetragonal a-axis). Concentrating on two independent Bragg peaks, they observed that the CDW and SDW intensities are redistributed at a compressive strain of $\epsilon_a = \Delta a/a \approx 0.02\%$ while the Bragg peak positions and correlation lengths remained unchanged (see Fig. 3). This provides direct evidence for charge-spin stripe order in La-based cuprates, and effectively rules out other multi-Q single domain structures. The scientists further found that CDW and SDW orders response to uniaxial pressure in a similar manner. In both cases the application of pressure along one of the Cu-O directions favors the domain with wavevector along the perpendicular

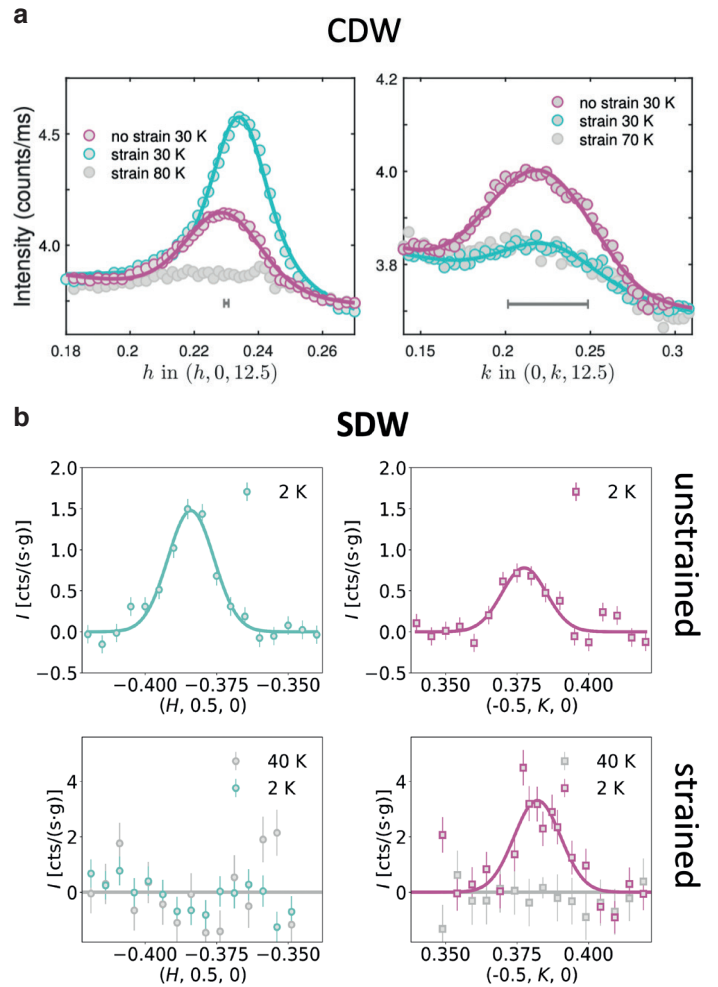


Fig. 3. Uniaxial charge spin-order in La-based cuprates. **a** and **b**: Repopulation of CDW and SDW domains upon uniaxial strain along the Cu-O bond direction, respectively (Figure **a** was reprinted with permission from [7]. © (2022) by the American Physical Society. Figure **b** was taken from [8])

direction. This strongly suggests an intertwined microscopic coupling between CDW and SDW order, and indicates that the putative pair-density wave also consists of uniaxial stripes. Finally, these results attest that uniaxial charge-spin stripe fluctuations are involved in the emergence of high-temperature superconductivity.

Further insight into these fluctuations and their coupling to the lattice degree of freedom was obtained by the same

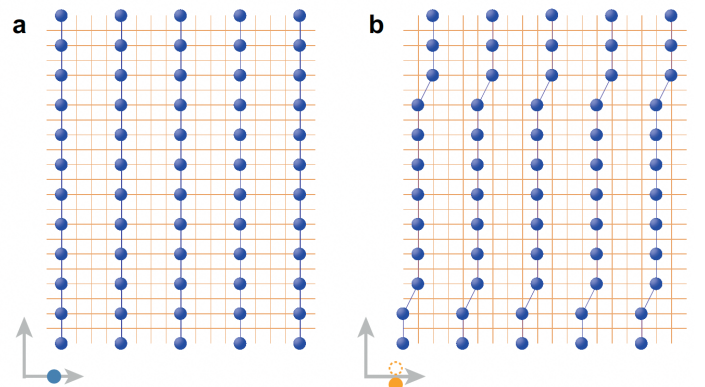


Fig. 4. Modest uniaxial pressure depins transverse stripe kinks. **a** and **b**: Schematic representation of the uniaxial stripes and transverse kinks originating from transverse stripe fluctuations. Modest pressures align the stripes along the Cu-O directions, whereas large strains generate a single stripe domain (published in [9] under Creative Commons Attribution 4.0 International License).

collaboration using resonant x-ray scattering [9]. Transverse stripe fluctuations have been predicted to promote superconductivity by enhancing the Josephson coupling along the c -axis [27]. While such fluctuations are extremely challenging to probe directly, they are also thought to trigger stripe kinks such as shown in Fig. 4a and b. The kinks cause a small incommensuration perpendicular to the CDW wavevector, ie. CDW diffraction peaks are often observed at $\mathbf{q}_{\text{CDW}} = (\delta_{\text{CDW}}, \delta_{\perp}, 0)$ and $(\delta_{\perp}, \delta_{\text{CDW}}, 0)$ with $\delta_{\perp} \neq 0$. Using modest ($\epsilon_a < 0.02\%$) uniaxial strain along the Cu-O bond direction, the scientists studied the transverse pinning properties of the charge stripe order. They found that uniaxial strain first aligns the stripe domains along the Cu-O bond direction, before a single domain state is established at larger pressures. This suggests that low-energetic transverse stripe fluctuations are necessary for high-temperature superconductivity and allows the state to coexist with static stripe order.

The aforementioned publications did not reveal any changes in the competition between superconductivity and stripe order under uniaxial pressure along the Cu-O bond direction of LSCO [7–9]. Interestingly, another international collaboration led by researchers from the Paul Scherrer Institut has observed a different behavior in LBCO when uniaxial pressure is applied along an in-plane axis close to the Cu-Cu bond direction [10]. The pT -phase diagram of LBCO is shown in Fig. 5a, overall following the same behavior as LSCO (see Fig. 1). LBCO, however, features an additional structural transition above $x > 0.09$ enabling the stripe domains to alter along the c -axis (see Fig. 5a). As a result the competition between superconductivity and stripe order is enhanced, leading to an almost complete suppression of superconductivity at $x = 1/8$ where stripe order is strongest. Signatures of the pair-density wave order are often observed as an additional transition in the resistivity and is often referred to two dimensional superconductivity [10]. Combining muon spin rotation and magnetic susceptibility measurements the scientists studied the uniaxial pressure dependence of the SDW order and onset of the two superconducting order parameters in $\text{La}_{1.885}\text{Ba}_{0.115}\text{CuO}_4$. Figure 5b shows their main result, demonstrating a strong competition between stripe order and superconductivity. In fact, a pressure of ~ 0.02 GPa is sufficient to reach an identical transition temperature of all three orders. This demonstrates that the different coherent phenomena are energetically almost degenerate at ambient conditions, and suggests that they arise from stripe fluctuations.

Despite the progress made through these four publications, a plethora of scientific questions remain unresolved and require further investigation. Dedicated studies on LSCO and LBCO under pressure along different crystal directions will gain further insight into the coupling between stripe order and superconductivity. Further scattering experiments will be needed to understand how stripe order is related to the lattice degrees of freedom, and future experiments directly assessing the dynamic properties will be unavoidable to

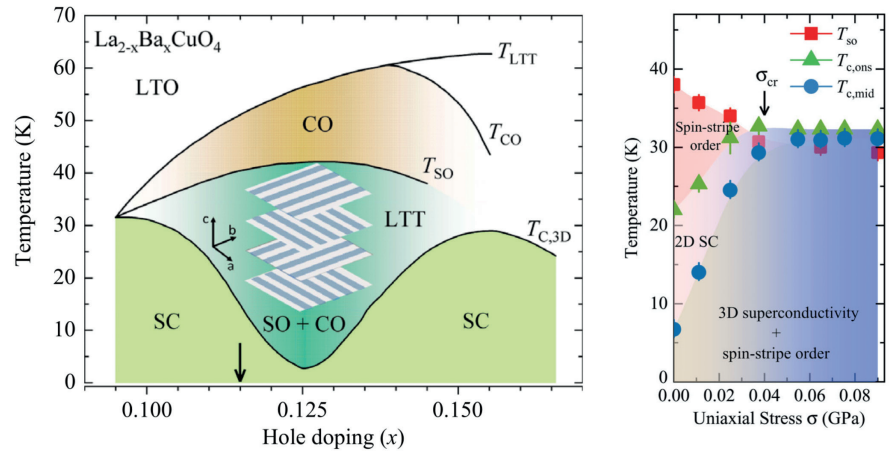


Fig. 5. Uniaxial pressure dependence in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$. **a**: pT -phase diagram, **b**: Evolution of the spin-stripe order and superconductivity under uniaxial strain along an in-plane axis close to the Cu-Cu bond direction (Reprinted figure with permission from [10]. © (2020) by the American Physical Society.)

gain a deeper understanding into the pairing mechanism of high-temperature superconductivity. The aforementioned studies, however demonstrate how uniaxial pressure, a particular clean tuning parameter, can be used to efficiently study the coupling among different degrees of freedom. These developments will be also key in shining new light on other quantum phenomena in correlated electron materials.

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