Hints for the nematic pseudogap in the nearly optimally doped La$_{2-x}$Sr$_x$CuO$_4$ superconductor

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The in-plane anisotropy of the Seebeck (S) and Nernst (ν) coefficients of the nearly optimally doped cuprate superconductor La$_{1.6}$Sr$_{0.4}$CuO$_4$ was measured under uniaxial pressure. Regardless of the qualitative differences between the S(T) and ν(T) dependences, both the Seebeck and Nernst anisotropies evolve very similarly with temperature. Namely, they emerge slightly above the structural transition, then change their character, and eventually sign at the temperature close to the temperature of the pseudogap formation. We conclude that the pseudogap possibly breaks rotational symmetry and we indicate an important role played in this phase by small-angle inelastic scattering.

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I. INTRODUCTION

Among the plethora of electronic orders emerging in the phase diagram of copper-based superconductors, the pseudogap is perhaps the most elusive one. Its presence was reported shortly after the discovery of high-Tc superconductors [1,2], and despite the enormous effort that was made to reveal the properties of the pseudogap, a physical origin of this phase is still being debated. The respective scenarios can be basically divided into two categories: these relating the pseudogap to the preformed, but not fully coherent, superconducting Cooper pairs [3–5] and those pointing at other types of electronic order [6–8] that are often claimed to compete with superconductivity [9–11]. Notably, different models for the formation of the pseudogap predict that the electron system in this state should break various symmetries [12–15]. The electronic nematicity, which appears to be common among various families of unconventional superconductors [15–21], breaks the rotational (but preserves translational) symmetry and has been recently suggested to be the true foundation of the pseudogap phase [22,23].

Here, we provide experimental evidence that the pseudogap in a La$_{2-x}$Sr$_x$CuO$_4$ high-Tc superconductor is in fact nematic. A single crystal of nearly optimally doped La$_{2-x}$Sr$_x$CuO$_4$ with $x = 0.141$ was subjected to uniaxial pressure applied within the ab plane which allowed us to determine the in-plane anisotropy of the electrical resistivity as well as the Seebeck and Nernst coefficients. The temperature dependences of the thermoelectrical phenomena indicate that the observed anisotropy is not directly related to the structural transition and a scenario explaining the formation of the pseudogap needs to include the presence of twofold in-plane anisotropy of the small-angle inelastic scattering.

II. METHODS

Single crystals of La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) were grown by the traveling-solvent floating-zone method under flowing O$_2$ gas of 4 bars [24]. The as-grown single-crystalline rod was annealed in flowing O$_2$ gas of 1 bar at 900°C for 50 h, cooled down to 500°C at a rate of 8°C/h, kept at 500°C for 50 h, and then cooled down to room temperature at a rate of 8°C/h. The Sr content of the sample was analyzed by inductively coupled-plasma optical-emission spectrometry (ICP-OES). The quality of the samples was checked by x-ray back-Laue photography to be good. The samples were also checked by powder x-ray diffraction, in which no Bragg peaks due to impurities were observed. The full width at half maximum of the (006) rocking curve was 0.1°–0.2°, which is comparable to that reported in the previous literature [24,25]. The distribution of the Sr content in a sample was also checked using an electron probe microanalyzer (EPMA) to be homogeneous within experimental accuracy.

For the transport measurements a square-like plate with dimensions $2 \times 2 \times 0.1$ mm$^3$ was cut from the rod. The crystallographic c axis was oriented along the shortest side of the sample, whereas the tetragonal a axis was at 45° with the sides of the face square. Such a sample was mounted between two clamps made of phosphor bronze and subjected to uniaxial pressure applied along its sides by a beryllium copper spring controlled with a stepper motor. For the resistivity measurements, the electrical contacts were placed at the corners of a sample and the orientations of the voltage and current leads were switched repetitively during the experiment. This
allowed the determination of the resistivity anisotropy using the Montgomery method [26].

The Seebeck and Nernst coefficients were measured along and across the strain direction in two separate runs where the magnetic field (parallel to the c axis) was varied from −12.5 to +12.5 T. The temperature difference along a sample was determined using two Cernox thermometers as well as a calibrated in magnetic field constantan-chromel thermocouple attached to the sample through a few-millimeters-long and calibrated phosphor bronze wires. The thermoelectrical resistivity anisotropy measured in two experimental configurations differed within several percent in the high-temperature limit (due to slightly different geometrical factors), and were matched by applying a multiplicative correction factor close to 1. More details about the experimental setup are given in the Supplemental Material [27].

III. RESULTS

In Fig. 1 the normalized in-plane resistivity anisotropy ($\Delta\rho_{\text{norm}} = \Delta\rho_{a} - \Delta\rho_{b}$) for La$_{1.36}$Sr$_{0.14}$CuO$_4$ under uniaxial pressures of approximately 2.5, 7.5, 10, 13, and 15 MPa. The inset presents the temperature derivative of the resistivity anisotropy using the analogous temperature dependences of the Nernst coefficient.

FIG. 2. Temperature dependences of the thermoelectric power for La$_{1.36}$Sr$_{0.14}$CuO$_4$ measured along a (red points) and b (blue points) crystallographic axes under uniaxial pressure of approximately 15 MPa. Green points that lie between $S_a$ and $S_b$ depict the Seebeck coefficient measured at ambient pressure. The inset presents the analogous temperature dependences of the Nernst coefficient.
sign change at \( T \approx 80 \text{ K} \), the relative Seebeck anisotropy \( (S_a - S_b)/(S_x + S_y) \) at its maximum at \( T \approx 50 \text{ K} \) reaches 6\%, which is about two orders of magnitude higher than the relative crystallographic anisotropy. Therefore, the orthorhombicity cannot be the direct cause of the thermoelectric anisotropy but it rather serves as a weak external field that makes it possible to align the nematic domains. This in turn allows one to observe the macroscopic consequences of a tendency of the electronic system to self-organize in a way that breaks rotational symmetry.

In order to further investigate this phenomenon, the Seebeck and Nernst anisotropies (\( \Delta S = S_a - S_b \) and \( \Delta \nu = \nu_a - \nu_b \), respectively, divided by \( T \) to account for temperature changes of entropy) are plotted along with the second temperature derivative of the electrical resistivity \( \partial^2 \rho/\partial T^2 \) to highlight a correspondence between these three dependences. First, one can identify a maximum in \( \partial^2 \rho/\partial T^2 \) at \( T \approx 210 \text{ K} \) which occurs at the tetragonal/orthorhombic transition [36] and slightly above the onset of this transition both Seebeck and Nernst coefficients deviate from zero. Subsequently, \( \partial^2 \rho/\partial T^2 \) changes sign at \( T^* \approx 170 \text{ K} \), which marks the inflection point of \( \rho(T) \). According to Ando et al. [36], this is related to the onset of the pseudogap and we use it to define \( T^* \) which agrees well with the expected value for a given composition. Again, slightly above this temperature both \( \Delta S(T)/T \) and \( \Delta \nu(T)/T \) change their character, go through extrema (reaching \( \Delta S_{\text{max}} \) and \( \Delta \nu_{\text{min}} \), respectively), and consequently change sign at \( T \approx 150 \text{ K} \). This observation means that the thermoelectrical anisotropies caused by the structural transition and the emergence of the pseudogap have opposite signs. Interestingly, \( \Delta \rho_{\text{norm}} \) does not exhibit any anomaly at \( T^* \), whereas the evolution of \( \Delta S(T)/T \) and \( \Delta \nu(T)/T \) is very similar despite \( S(T) \) and \( \nu(T) \) being substantially different.

This raises a question about a possible connection between the Nernst and Seebeck coefficients. Actually, these two are similar despite \( \Delta S_a \approx \Delta \nu_x \approx \Delta S_{B_{\text{str}}} \approx \Delta S_{\text{max}} \approx \Delta S_{\text{min}} \), while the evolution of \( \Delta S_x/T \) and \( \Delta \nu/T \) change their character, go through extrema (\( \Delta S_{\text{max}} \) and \( \Delta \nu_{\text{min}} \), respectively), and consequently change sign at \( T \approx 150 \text{ K} \). This observation means that the thermoelectrical anisotropies caused by the structural transition and the emergence of the pseudogap have opposite signs. Interestingly, \( \Delta \rho_{\text{norm}} \) does not exhibit any anomaly at \( T^* \), whereas the evolution of \( \Delta S(T)/T \) and \( \Delta \nu(T)/T \) is very similar despite \( S(T) \) and \( \nu(T) \) being substantially different.

IV. DISCUSSION

There are still controversies over the origin of the pseudogap, but also over basic properties of this enigmatic state. For instance, there is an ongoing debate over whether the pseudogap emerges in the phase transition [39,40], and if it is a “crossover” phenomenon [41] or rather a kind of fluctuating order [42]. It is also not clear which symmetries are broken below \( T^* \), although some reports indicate that in this region the electron system breaks in-plane fourfold lattice symmetry [15,23]. On the other hand, studies of the transport properties in \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) point to a contribution from Cu-O chains as a possible source of the anisotropy [38]. \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) differs from \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) in this respect, since its crystallographic structure lacks Cu-O chains. The lattice obviously breaks fourfold symmetry in the low-temperature orthorhombic phase, but the crystallographic anisotropy is much smaller than the one observed in transport. Moreover, the electrical resistivity in thin films of \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \), which were forced to retain tetragonal symmetry, still exhibited twofold symmetry.
 FIG. 4. Temperature dependences of the Seebeck (blue diamonds, left axis) and Nernst (red points, right axis) anisotropies shown in a semilogarithmic scale for La$_{1.85}$Sr$_{0.15}$CuO$_4$. The plots include offsets ($\Delta S_{\text{max}}$ and $\Delta \nu_{\text{max}}$, respectively) described in the text. Solid lines are fits to the Haller equation. The temperature dependence of the CDW peak height for La$_{1.85}$Sr$_{0.15}$CuO$_4$ taken from Ref. [56] (normalized to the value of $-\Delta S - \Delta S_{\text{max}}$ at $T = 50$ K) are superimposed on the plot as yellow stars. The inset presents exemplary temperature dependences of $\Delta \nu/T$ for YBa$_2$Cu$_3$O$_7$ with a hole concentration $p = 0.12$ (YBCO) [38], Ba(Fe$_{0.95}$Co$_{0.05}$)$_2$As$_2$ (Ba-122) [37], and FeTe [21], along with the respective Haller equation fits. 

with the director loosely related to the orientation of the lattice [43].

Here, we show that while the onset of transport anisotropy correlates with the structural transition, $\Delta S(T)/T$ and $\Delta \nu(T)/T$ at lower temperatures change their character (and eventually sign), likely in response to the emergence of nematic order. Since the Seebeck and Nernst coefficients can be generally understood as a measure of the entropy flux [44], this indicates that the in-plane entropy transport becomes anisotropic (or, more specifically, suppressed along the uniaxial strain according to the signs of $\Delta S$ and $\Delta \nu$) below $T^*$. We would like to mention that we do not expect domain walls to play an important role in determining the in-plane anisotropy, since our sample at a uniaxial pressure of $P \approx 15$ MPa should be mostly detwinned. Early results of x-ray diffraction measurements on stressed La$_{2-x}$Sr$_x$CuO$_4$ single crystals showed that for $P = 1.5$ and 1.8 MPa the detwinning ratio was about 90% [45] and 76% [46], respectively.

There are several possible scenarios explaining anisotropic entropy flow in high-$T_c$ superconductors. First, a contribution from vortexlike excitations [34] can be likely excluded, because this should mostly affect the transverse thermoelectric response. The superconducting fluctuations are also expected to influence the diagonal and off-diagonal elements of the Peltier tensor ($\alpha_{xx}$ and $\alpha_{xy}$, respectively) differently [47]. We propose that the in-plane anisotropy can originate from an anisotropic scattering and since the resistivity and thermoelectrical anisotropies behave differently, we point at the small-angle inelastic scattering as a possible underlying mechanism. Such processes disturb mostly the entropy current, leaving the electrical transport almost unaffected [48,49]. Their abundance is characteristic for strongly correlated electron systems described, for example, as a hydrodynamic electron fluid [50,51], where the energy dissipation is dominated by small-angle electron-phonon scattering [52]. Alternatively, the electron-electron relaxation can be another source of momentum conserving collisions in the high-$T_c$ superconductors [53], whereas in FeSe it can result from the interaction of charge carriers with anisotropic spin fluctuations [54]. Another possibility is the scattering of quasiparticles by charge density wave (CDW) fluctuations that get ordered in the weak strain field, and which were shown to be a source of small-angle inelastic scattering [55].

Noteworthy, CDW short-range order was recently detected in an extended range of temperatures for La$_{2-x}$Sr$_x$CuO$_4$ [56] and electron nematicity may arise via a thermal or quantum phase transition from the proliferation of topological defects of the stripe (i.e., smectic) state [57]. A similar behavior was also observed in NbSe$_2$, where short-range CDW survived beyond the phase coherence transition [58]. Using data from Ref. [56], we have superimposed the height of the CDW peak for similarly doped La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.144$) on the temperature dependences of the thermoelectrical anisotropies in Fig. 4. Evidently, all three quantities exhibit the same type of temperature dependence, and a similar, but not the same, slope.

The last subject we want to address is the temperature dependence of the Nernst coefficient that appears to be common for different unconventional superconductors, both in iron-based superconductors [20,21,37] and cuprates [38] (see the inset in Fig. 4). In Fig. 4 we fit the $\Delta S(T)/T$ and $\Delta \nu(T)/T$ data with the equation proposed to describe the nematic order parameter in liquid crystals [59], $\Delta \nu/T = \alpha (1 - T/T_{\text{th}})^\beta$, where $\alpha$ is the proportional constant, $\beta$ is the pseudocritical exponent, and $T_{\text{th}}$ is a characteristic temperature somewhat lower than the actual temperature of the discontinuous nematic-isotropic transition. Interestingly, this fit seems to be universally applicable to the Nernst data from many different compounds, although the obtained values of $\beta$ are much different (e.g., $\beta \approx 2.6$ for La$_{1.85}$Sr$_{0.15}$CuO$_4$) than values expected for liquid crystals, i.e., $\beta \approx 0.25$ [60].

V. CONCLUSION

The pseudogap is considered to be a key to understanding the mechanism leading to the formation of high-$T_c$ superconductivity [61]. This belief has driven extensive studies aimed at the discovery of the true nature of the pseudogap, which led to the development of new experimental techniques, including measurements under uniaxial pressure. While there are still no definitive conclusions, there seems to be growing evidence that the pseudogap breaks rotational symmetry. One example is a very recent work by Ishida et al. [62] that reports nematic quantum criticality in lead-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$+x. The presence of such a quantum critical point could be linked to superconductivity and so-called strange metallic behavior.

In general, the spontaneous tendency of an electron system to break rotational symmetry appears to be common in materials with strong electron correlations. In compounds with an orthorhombic crystalline structure, the observation of the macroscopic consequences of nematicity might be easier,
because even a slight difference between the $a$ and $b$ lattice parameters provides a symmetry breaking field, which allows for the alignment of nematic domains [57]. However, this also can cause doubts about whether the detected anisotropy is not related to the orthorhombic itself.

Our data show that the thermoelectric anisotropy in La$_{1.06}$Sr$_{0.14}$CuO$_4$ indeed appears at the structural transition, but then it changes sign at $T^\ast$, which implies that the low-temperature anisotropy is related to the pseudogap rather than a difference in the lattice parameters. Correspondingly, a recent study of La$_{2-x}$Sr$_x$CuO$_4$ thin films under uniaxial strain shows that the nematicity is of an electronic origin, with very weak coupling to the crystal lattice [63]. Remarkably, while all the Seebeck, Nernst, and resistivity anisotropies emerge around the tetragonal/orthorhombic structural transition, only the thermoelectric ones seem to be sensitive to the onset of a pseudogap at $T^\ast$.

We conclude that the mechanism responsible for such a behavior is anisotropic small-angle scattering and we point at the short-range unidirectional charge density order as a possible root cause.

All of the relevant data that support the findings of this study are available from the corresponding author upon reasonable request.

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