Anisotropy of the Seebeck Coefficient in the Cuprate Superconductor YBa$_2$Cu$_3$O$_y$: Fermi-Surface Reconstruction by Bidirectional Charge Order

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The Seebeck coefficient $S$ of the cuprate YBa$_2$Cu$_3$O$_y$ was measured in magnetic fields large enough to suppress superconductivity, at hole dopings $p = 0.11$ and $p = 0.12$, for heat currents along the $a$ and $b$ directions of the orthorhombic crystal structure. For both directions, $S/T$ decreases and becomes negative at low temperature, a signature that the Fermi surface undergoes a reconstruction due to broken translational symmetry. Above a clear threshold field, a strong new feature appears in $S_b$, for conduction along the $b$ axis only. We attribute this feature to the onset of 3D-coherent unidirectional charge-density-wave modulations seen by x-ray diffraction, also along the $b$ axis only. Because these modulations have a sharp onset temperature well below the temperature where $S/T$ starts to drop towards negative values, we infer that they are not the cause of Fermi-surface reconstruction. Instead, the reconstruction must be caused by the quasi-2D bidirectional modulations that develop at significantly higher temperature.

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I. INTRODUCTION

In the last decade, various transport measurements in high magnetic fields have revealed that the Fermi surface of hole-doped cuprate superconductors undergoes a reconstruction at low temperature in a doping interval centered at $p \approx 0.12$ [1]. The key feature of this Fermi-surface reconstruction (FSR) is the presence of a small electron-like pocket, detected by quantum oscillations [2–5], combined with sign changes in the temperature dependence of the Hall ($R_H$) and Seebeck ($S$) coefficients, from positive at high temperature to negative at low temperature. A negative $R_H$ or $S$ has now been observed in seven hole-doped cuprates: YBa$_2$Cu$_3$O$_y$ (YBCO) [6–9], YBa$_2$Cu$_4$O$_8$ (Y124) [6], HgBa$_2$CuO$_{4+x}$ (Hg1201) [10], La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [11, 12], La$_{1.8-x}$Eu$_{0.2}$Sr$_2$CuO$_4$ (Eu-LSCO) [9], La$_{1.6-x}$Nd$_{0.4}$Sr$_2$CuO$_4$ (Nd-LSCO) [13, 14], and La$_{2-x}$Ba$_x$CuO$_4$ (LBCO) [15].

There is compelling evidence that this FSR is caused by charge-density-wave (CDW) order. Indeed, in all materials and at every doping where FSR has been detected, CDW modulations have also been observed by x-ray diffraction (XRD) [16–21] (except in Y124, where no XRD search has been reported). Having said this, the mechanism by which CDW order produces a small electron pocket in the Fermi surface of hole-doped cuprates remains a puzzle. This is because CDW order is thought to be unidirectional (or “stripe-like”) in at least some cuprates and a unidirectional CDW modulation does not in general produce a closed electron pocket [22], at least not at “nodal” locations in the Brillouin zone, away from the anti-nodal pseudogap [23]. By contrast, bidirectional CDW order (with in-plane modulations along both high-symmetry directions of the tetragonal or orthorhombic lattice) readily produces a closed electron pocket at “nodal” locations [24, 25].

This paradox has recently become vivid in the orthorhombic cuprate YBCO at $p = 0.12$, where XRD studies in high magnetic fields detect long-range three-dimensional (3D) CDW order [26], with modulations that run only along the $b$ axis [27, 28], above a sharply defined threshold field that coincides with an anomaly in the sound velocity considered to be the thermodynamic signature of CDW order in YBCO [29]. Is this field-induced unidirectional CDW order causing the FSR in YBCO?

Here, we report measurements of the Seebeck coefficient $S$ of YBCO along the $a$ and $b$ axes at $p = 0.11$ and $p = 0.12$, in magnetic fields high enough to reach the normal state. For both directions, we observe a negative $S$ at low temperature, signature of the FSR giving an electron pocket. In addition, we detect a pronounced minimum in $S_a(H)$, not present in $S_b(H)$, whose onset field and temperature match the onset of the 3D unidirectional CDW order seen by XRD. However, since this onset temperature is well below the temperature where $S/T$ starts to drop towards negative values, we infer that the primary cause of the FSR are the 2D bidirectional CDW modulations that develop in tandem with the gradual drop in $S/T$. 

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FIG. 1. Seebeck coefficient of YBCO at $p = 0.11$, for a heat current along the a axis (a) and b axis (b) of the orthorhombic crystal structure, plotted as $S / T$ vs magnetic field $H$, at various temperatures, as indicated. The negative value of $S / T$ at low temperature and high field is the signature of Fermi-surface reconstruction. In the isotherms of $S_a / T$ vs $H$ (b), a clear dip develops below $T \approx 40$ K, at $H \approx 18$ T.

II. METHODS

Single crystals of YBa$_2$Cu$_3$O$_y$ (YBCO) were prepared by flux growth [30]. Their hole concentration (doping) $p$ is determined from the superconducting transition temperature $T_c$ [31], defined as the temperature below which the zero-field resistance is zero. A high degree of oxygen order was achieved for samples with $p = 0.11$ ($y = 6.54$, ortho-II order, $T_c = 61.5$ K) and $p = 0.12$ ($y = 6.67$, ortho-VIII order, $T_c = 65.4$ K). The Seebeck coefficient $S$ – the longitudinal voltage generated by a longitudinal thermal gradient – was measured, as described elsewhere [9], on two pairs of a-axis and b-axis YBCO samples, with dopings $p = 0.11$ and $p = 0.12$. $S(H)$ was measured as a function of magnetic field up to $H = 34$ T, applied along the c axis, in YBCO samples with $p = 0.11$ at the Laboratoire National des Champs Magnétiques Intenses, (LNCMI) in Grenoble and in samples with $p = 0.12$ at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee. Our b-axis sample with $p = 0.11$ was also measured up to $H = 45$ T, at the NHMFL. At $p = 0.11$ and 0.12, the critical field for suppressing superconductivity in YBCO is $H_{c2} = 25$ T [32].

III. RESULTS

In Fig. 1, the Seebeck coefficient of YBCO at $p = 0.11$ is plotted as $S / T$ vs $H$, for several temperatures. Our data on $S_a$ agree well with previous measurements of the Seebeck coefficient in YBCO [8, 9]. To our knowledge, there are no prior high-field measurements of $S_b$,
in YBCO. We see that for both directions, $S / T$ at high field goes from positive at high temperature to negative at low temperature, the signature that FSR is occurring upon cooling, resulting in a Fermi surface at low temperature that contains a small electron pocket \cite{1}. Note that the magnitude of $S / T$ at $T \to 0 \ (\simeq -0.8 \mu V / K^2)$ is consistent with theoretical expectation \cite{33} in the sense that $S / T = -(\pi^2/2) (k_B/e) (1/T_F) = -1.0 \mu V / K^2$ \cite{9}, if we use the Fermi temperature $T_F = 410 \ K$ measured by quantum oscillations in YBCO at $p = 0.11 \ [2, 34]$.

The isotherms of $S_a$ in Fig. 1(b) reveal a new and pronounced feature, essentially absent in $S_b$. Indeed, on top of the overall field and temperature dependence as observed in $S_a / T$, $S_b / T$ exhibits an upturn at high field, producing a dip at $H \simeq 18 \ T$ that deepens as temperature is reduced. In Fig. 2(a), we focus on this feature by comparing $S_a / T$ (blue) and $S_b / T$ (red) vs $H$ at $T = 20 \ K$. At low field (up to about $16 \ T$), both curves are identical: zero in the vortex-solid state, then slightly positive, followed by a dramatic drop to large negative values. At fields above $16 \ T$, a striking anisotropy between the two directions appears, as a pronounced upturn develops in $S_b$, but not in $S_a$. If we identify the field at which $S_b$ reaches a minimum as $H_{\text{Seebeck}}$, equal to $19 \pm 1 \ T$ at $T = 20 \ K$. Fig. 2(b) presents the same comparison at $p = 0.12$, in crystals with a different oxygen order (ortho-VIII instead of ortho-II). We observe a very similar Seebeck anisotropy, again characterized by an upturn in $S_b$, appearing above $H_{\text{Seebeck}} = 16 \pm 1 \ T$.

To study the temperature dependence of $H_{\text{Seebeck}}$ in detail, we measured closely-spaced isotherms of $S_b$ up to $45 \ T$, plotted in Fig. 3. We see that the minimum in $S_b / T$ vs $H$ is present at temperatures up to at least $30 \ K$, remaining in roughly the same position. In Fig. 4, we plot $H_{\text{Seebeck}}$ on the $H - T$ phase diagram of YBCO at $p = 0.11$ (yellow squares). It is essentially constant in temperature up to $30 \ K$.

In Fig. 5(a), $S_a(T)$ and $S_b(T)$ measured at $H = 34 \ T$ are compared directly, plotted as $S / T$ vs $T$. We see that down to $45 \ K$, the two curves are approximately parallel, with a roughly constant difference between them. Indeed, a smooth fit through the $a$-axis data (blue line) makes a good fit through the $b$-axis data if the line is simply shifted down rigidly (red line). Below $45 \ K$, $S_a / T$ continues its monotonic decrease, but the anomalous feature in $S_b$ produces a striking departure of $S_b / T$ from its fit line (red), initially as a plateau which persists down to $\simeq 30 \ K$. To capture this extra anisotropy, we plot the difference between $b$-axis data and red fit line in the inset of Fig. 5(a). We see that it appears below $T_{\text{Seebeck}} = 47 \pm 5 \ K$. $T_{\text{Seebeck}}$ is plotted on the $H - T$ phase diagram of Fig. 4, for three different fields.
at $p=0.11$ and 0.12, respectively (Fig. 2). This value of $H_{\text{Seebeck}}$ at $p=0.11$ is in agreement with the anomaly in the sound velocity [29] that marks the phase transition to CDW order (Fig. 4). Those field values are also in agreement with the threshold field detected in the thermal Hall conductivity $\kappa_{xy}$ [35], for both $p=0.11$ (Fig. 4) and $p=0.12$. Therefore, it is clear that $H_{\text{Seebeck}}$ coincides with the onset of 3D unidirectional CDW order.

The onset temperature for that order ($T \simeq 47$ K) [27, 28] is not far below the onset of the NMR splitting associated with CDW order [37]. There is little doubt that $T \simeq 47 \pm 5$ K coincides with the onset of 3D unidirectional CDW order.

The fact that we can clearly detect the onset of 3D unidirectional CDW order in the Seebeck coefficient allows us to examine whether it causes the FSR in YBCO. In Fig. 5(b), we plot $S_a / T$ vs $T$ at $p=0.12$ for $H = 10$, 16, and 34 T. We see that $S_a / T$ starts to deviate downward from its high-temperature behaviour below $T \simeq 130$ K, it peaks at 105 K and then it drops to become negative below $\sim 60$ K. This is a gradual process, which starts in parallel with the gradual growth of short-range 2D CDW modulations seen in XRD below $\simeq 140$ K [19]. (Down to $T_c$, both the Seebeck and the XRD intensity are independent of magnetic field.)

The decrease in $S / T$ upon cooling is the signature of the FSR that leads to the formation of a small electron pocket in the Fermi surface at low temperature, detected via quantum oscillations, whose Fermi energy is consistent with the value of $S / T$ at $T \to 0$ (Fig. 5(b)) [9]. In other words, the entire evolution of $S_a / T$ vs $T$ is quantitatively consistent (in temperature and in amplitude) with a scenario whereby FSR is caused by the 2D CDW modulations (CDW-1). The fact that this evolution is completely unaffected by the sharp onset of the 3D unidirectional CDW order at 47 K, measured in YBCO at the same doping ($p=0.12$) and the same field ($H=16$ T) [27], indicates that it doesn’t play a fundamental role in causing the FSR. It appears to only confer an extra anisotropy.

The fact that the 2D CDW modulations are bi-directional, i.e. that they run along both the $a$ and $b$ directions in the CuO$_2$ planes of YBCO, provides a natural mechanism for the formation of a small electron pocket in the reconstructed Fermi surface [24, 25], located in nodal positions where the states are believed to be in under-doped cuprates with an anti-nodal pseudogap. An analysis of the anomalies in the sound velocities concluded that the order responsible for the observed transition must be bi-directional [29].

Note that the CDW modulations observed in Hg1201 [20] are very similar to the 2D CDW modulations in YBCO, and they cause a very similar FSR [38], with negative Hall and Seebeck coefficients at low temperature [10]. Therefore, attributing the cause of the FSR to these 2D CDW modulations is consistent with the fact that so far no field-induced 3D CDW order has been observed in Hg1201.

FIG. 5. a) Seebeck coefficient of YBCO at $p=0.11$ along the $a$ axis (blue) and $b$ axis (red), at $H=34$ T, plotted as $S / T$ vs $T$. The blue line is a smooth fit to the $S_a$ data; the red line is the same line shifted down by 0.1 $\mu$V / K$^2$. The anomalous feature seen in the field dependence of $S_a$ (Fig. 2) shows up in the temperature dependence of $S_a / T$ initially as a plateau. Inset: Difference between the $b$-axis data points and the red line (fit) in the top panel (red dots). The green dots are obtained using the $S_b$ data of Fig. 1(b). The onset temperature for the extra anisotropy is $T_{\text{FSR}} = 47 \pm 5$ K (arrow). b) $S_a / T$ vs $T$, in YBCO at $p=0.12$, for three field values as indicated. At all fields, 2D bi-directional CDW modulations (CDW-1) are observed in the blue and red regions [36], while 3D uni-directional CDW order (CDW-2) is observed only in the red region and only when $H > 15$ T [27, 28]. The dashed line is a smooth extension of the high-$T$ data below its inflexion point at $T=130$ K. The dotted line is a linear extension of the data below $T=40$ K.

IV. DISCUSSION

The anomaly in $S_b$ we observe in YBCO at $p=0.11$ is confined to a region of the $H - T$ diagram (Fig. 4) that is essentially the same region where 3D unidirectional CDW order has been observed by XRD [26-28]. This order was detected in YBCO above an onset field $H=18$ T at $p=0.11$ [28] and above $H=15$ T at $p=0.12$ [26, 27], in good agreement with $H_{\text{Seebeck}} = 19 \pm 1$ T and 16 $\pm 1$ T
Given that 2D CDW modulations exist in the superconducting state at $H = 0$ [18, 19], one might ask: are there signatures of the FSR inside the superconducting phase, i.e. inside the green region of the $H - T$ phase diagram (Fig. 4)? The answer is yes: in YBCO at $p = 0.11$, $R_H$ at $T = 15$ K is negative for all fields down to $H = H_{\text{c}1} \simeq 10$ T, the field below which the vortex solid forms and $R_H = 0$ [35]. So a negative $R_H$ is observed even when $H < H_{\text{Seebeck}}$. In the vortex-liquid state between $H_{\text{c}1}$ and $H_{\text{c}2}$, the negative $R_H$ could come from states inside the vortex core.

On the other hand, the thermal Hall conductivity $\kappa_{xy}$ is dominated by $d$-wave quasiparticles outside the vortex cores. In YBCO at $p = 0.12$, $\kappa_{xy}$ is negative in the normal state just above $T_c$ [35], even in the limit $H = 0$, as is the electrical Hall conductivity [6]. Immediately below $T_c$, $\kappa_{xy}$ becomes positive [35]. This sudden change of sign could be due to a sudden increase in the quasiparticle mean free path as the inelastic scattering is gapped out, as found in YBCO immediately below $T_c$ [39]. Because the correlation length of the 2D CDW modulations is rather short in YBCO (and even shorter in Hg1201), the longer electronic mean free path in the superconducting state may well average over the short-range CDW and wipe out the FSR. Increasing the field to suppress superconductivity makes $\kappa_{xy}$ negative again [35]. The threshold field at which this change of sign happens coincides with $H_{\text{Seebeck}}$ (Fig. 4), i.e. with the onset field for 3D CDW order. This can be understood as follows: 3D CDW order competes with superconductivity, its onset precipitates the demise of superconductivity, which causes a reduction in the mean free path, making the FSR by short-range 2D CDW modulations possible again. In other words, 3D CDW order triggers the transition out of the superconducting phase and this is where $\kappa_{xy}$ starts its transition from zero to its normal-state (negative) value [35].

V. SUMMARY

In summary, the Seebeck coefficient $S$ of YBCO at $p = 0.11$ and 0.12 responds to two aspects of the complex CDW ordering in this material. First, as temperature is decreased from room temperature, $S_a / T$ deviates gradually downward from its dependence at high temperature in parallel with the gradual growth in the 2D bi-directional CDW modulations detected by XRD well above $T_c$. $S_a / T$ decreases below $T \simeq 100$ K to eventually become negative, extrapolating to a large negative value at $T \to 0$ that is quantitatively consistent with the small electron pocket in the normal-state Fermi surface detected by quantum oscillations at low temperature. We infer that the 2D bi-directional CDW modulations reconstruct the Fermi surface of YBCO, and produce the electron pocket. The same is true for Hg1201.

Secondly, a pronounced anomaly appears in $S_b$ below a temperature and above a field that are both consistent with the onset temperature and field of the 3D unidirectional CDW order detected in YBCO by high-field XRD at $p = 0.11$ and 0.12. We conclude that the extra anisotropy is due to that low-temperature order, which is not, however, the primary cause of the FSR. Nevertheless, given that the two types of CDW modulations (CDW-1 and CDW-2 in Fig. 4) have the same wavelength, they most likely have a common origin. It would be helpful to further elucidate the nature of their interplay.

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