

Hall, Seebeck, and Nernst Coefficients of Underdoped $\text{HgBa}_2\text{CuO}_{4+\delta}$: Fermi-Surface Reconstruction in an Archetypal Cuprate Superconductor

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(Received 31 October 2012; published 28 June 2013)

Charge-density-wave order has been observed in cuprate superconductors whose crystal structure breaks the square symmetry of the CuO_2 planes, such as orthorhombic $\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO), but not so far in cuprates that preserve that symmetry, such as tetragonal $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Hg1201). We have measured the Hall (R_H), Seebeck (S), and Nernst (ν) coefficients of underdoped Hg1201 in magnetic fields large enough to suppress superconductivity. The high-field $R_H(T)$ and $S(T)$ are found to drop with decreasing temperature and become negative, as also observed in YBCO at comparable doping. In YBCO, the negative R_H and S are signatures of a small electron pocket caused by Fermi-surface reconstruction, attributed to charge-density-wave modulations observed in the same range of doping and temperature. We deduce that a similar Fermi-surface reconstruction takes place in Hg1201, evidence that density-wave order exists in this material. A striking similarity is also found in the normal-state Nernst coefficient $\nu(T)$, further supporting this interpretation. Given the model nature of Hg1201, Fermi-surface reconstruction appears to be common to all hole-doped cuprates, suggesting that density-wave order is a fundamental property of these materials.

DOI: [10.1103/PhysRevX.3.021019](https://doi.org/10.1103/PhysRevX.3.021019)

Subject Areas: Condensed Matter Physics, Materials Science, Superconductivity

There is a growing body of evidence that competing ordered states shape the phase diagram of the cuprates, and the identification of those states is currently a central challenge of high-temperature superconductivity. In the La_2CuO_4 -based cuprates, whose maximal T_c does not exceed 40 K, the existence of unidirectional density-wave order involving spin and charge modulations, known as stripe order [1,2], is well established, as in $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ [3] and $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ (Eu-LSCO) [4], for instance. This stripe order causes a reconstruction of the Fermi surface [5–8] and may be responsible for the low T_c . The observation of a small electron pocket in the Fermi surface of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO) [9,10], a material with a maximal T_c of 93 K, shows that its Fermi surface also undergoes a reconstruction [11,12]. Comparative measurements of the Seebeck coefficient in YBCO and Eu-LSCO reveal a

detailed similarity [7,8], suggesting that Fermi-surface reconstruction (FSR) in YBCO is caused by some form of stripe order.

Charge-density-wave modulations were recently detected in YBCO, via high-field nuclear magnetic resonance (NMR) [13] and x-ray-scattering [14–18] measurements, in the range of temperature and doping where FSR occurs [8,19]. Although the detailed structure of these modulations remains to be clarified, there is little doubt that they are responsible for the FSR in YBCO.

The fundamental question, then, is whether such charge modulations are a generic property of the cuprates. Because both the low-temperature tetragonal structure of Eu-LSCO and the orthorhombic structure of YBCO distort the square CuO_2 planes and impose a preferred direction, charge modulations are perhaps triggered or stabilized by these particular forms of unidirectional distortion. To answer that question, we need to examine a cuprate material with square CuO_2 planes. For that purpose, the model material is $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Hg1201), a tetragonal cuprate with the highest maximal T_c of all single-layer cuprates (97 K) [20,21], in which no charge or spin modulations have yet been reported. In this article, we present measurements of the Hall, Seebeck, and Nernst coefficients in underdoped Hg1201 that reveal a FSR very similar to that seen in YBCO, consistent with some form of

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density-wave modulation in Hg1201. This observation is evidence that charge-density-wave modulations, as seen in YBCO, are a universal and fundamental property of underdoped cuprates.

Methods.—Two nominally identical high-purity single crystals of underdoped Hg1201 are measured (samples *A* and *B*), with $T_c \simeq 65$ K, prepared as described in Refs. [20,21]. According to the $T_c(p)$ relationship for Hg1201 established in Ref. [22], our samples have a doping $p \simeq 0.075$. We measure the Hall ($R_H \equiv \rho_{xy}/H$), Seebeck ($S \equiv -V_x/\Delta T$), and Nernst [$\nu \equiv N/H \propto (V_y/\Delta T)/H$] coefficients, where ρ_{xy} is the transverse resistivity, and V_x (V_y) is the longitudinal (transverse) voltage in the presence of a longitudinal temperature difference ΔT . For all measurements, the magnetic field H is applied perpendicular to the CuO_2 planes and the current (charge or heat) is within the plane. Hall measurements are performed in pulsed magnetic fields at the LNCMI in Toulouse up to $H = 68$ T, as described in Ref. [10]. The Seebeck coefficient is measured on sample *A*, as described in Ref. [8], up to 28 T at the LNCMI in Grenoble and up to 45 T at the NHMFL in Tallahassee. The Nernst coefficient is measured on sample *A* at Sherbrooke in a field of $H = 10$ T, as described in Ref. [23].

Negative Hall and Seebeck coefficients.—In Fig. 1(a), the Hall coefficient R_H is plotted as a function of magnetic field H up to 68 T, for different temperatures down to $T = 4.2$ K. All isotherms of sample *A* (*B*) saturate at high fields, beyond $H = 53$ T (68 T), except (including) at $T = 4.2$ K. In Fig. 1(b), the high-field value of R_H is plotted versus temperature. In Figs. 2(a) and 2(b), we show the corresponding Seebeck data as a function of magnetic field and temperature, respectively. As expected for a hole-doped material, both R_H and S are positive at high temperature. However, with decreasing temperature, they both start to fall below about $T \simeq 50$ K to eventually become negative below $T_0 = 10 \pm 1$ K. These findings are our central result: The low-temperature high-field normal state of Hg1201 has negative Hall and Seebeck coefficients.

In Figs. 1(b) and 2(b), comparison with corresponding data in underdoped YBCO, with $T_c \simeq 57$ K ($p = 0.1$) [8,10], reveals a striking similarity between the two cuprates. In YBCO, there is compelling evidence that the negative R_H and S at low temperature come from a small electron Fermi surface. This evidence includes quantum oscillations [9,24] with a frequency F and mass m^* , which, at this doping, account precisely for the normal-state Seebeck coefficient S at $T \rightarrow 0$, whereby $S/T \propto m^*/F$, given that S/T is negative [7,8]. By analogy, we deduce that the Fermi surface of underdoped Hg1201 at low temperature also contains an electron pocket. This fact implies that the Fermi surface is reconstructed relative to the topology of cuprates at high doping, namely, a single large holelike cylinder, as observed in the single-layer tetragonal cuprate $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$ at $p \simeq 0.25$ [25–27].

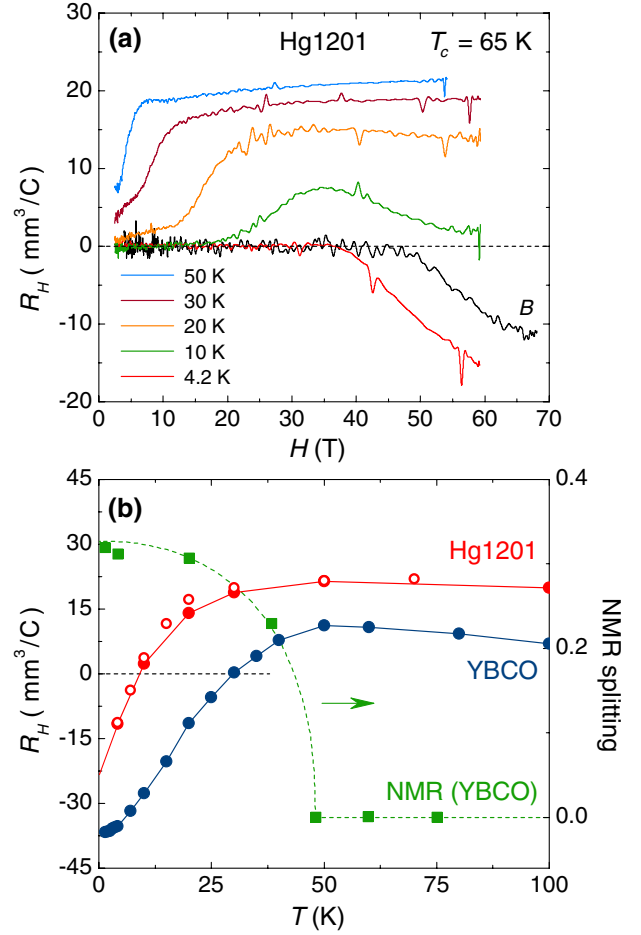


FIG. 1. (a) The Hall coefficient R_H of Hg1201 as a function of magnetic field H , in sample *A* at various temperatures, as indicated. Also shown is an isotherm at $T = 4.2$ K, measured on sample *B* (black curve, labeled *B*). (b) Normal-state Hall coefficient R_H as a function of temperature, at $H = 53$ T (sample *A*; closed red circles, left axis) and $H = 68$ T (sample *B*; open red circles, left axis). Corresponding data are shown for YBCO at $p = 0.10$ and $H = 55$ T (blue circles, left axis; from Ref. [10]). Note how in both materials, $R_H(T)$ drops at low temperature to become negative, a signature of Fermi-surface reconstruction [12]. We reproduce the splitting of NMR lines in YBCO at $p = 0.10$ and $H = 28.5$ T, which reveals the onset of charge order below $T_{\text{CO}} \simeq 50$ K (green squares, right axis; from Ref. [13]).

In other words, the FSR in Hg1201 sets in below a critical doping p^* located somewhere between $p \simeq 0.08$ and $p \simeq 0.25$, as is the case for YBCO [8,19], Eu-LSCO [8], and $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ [5,6,12,28]. This quantum critical point at p^* in the phase diagram of Hg1201 marks the onset of some density-wave order that breaks the translational symmetry of the lattice.

Our data also show that a transformation occurs upon cooling at fixed p , albeit smoothly, with no sign of a sharp transition. The onset of FSR may be associated with the temperature T_{max} at which $R_H(T)$ is maximal, although $R_H(T)$ clearly starts to deviate downward from its

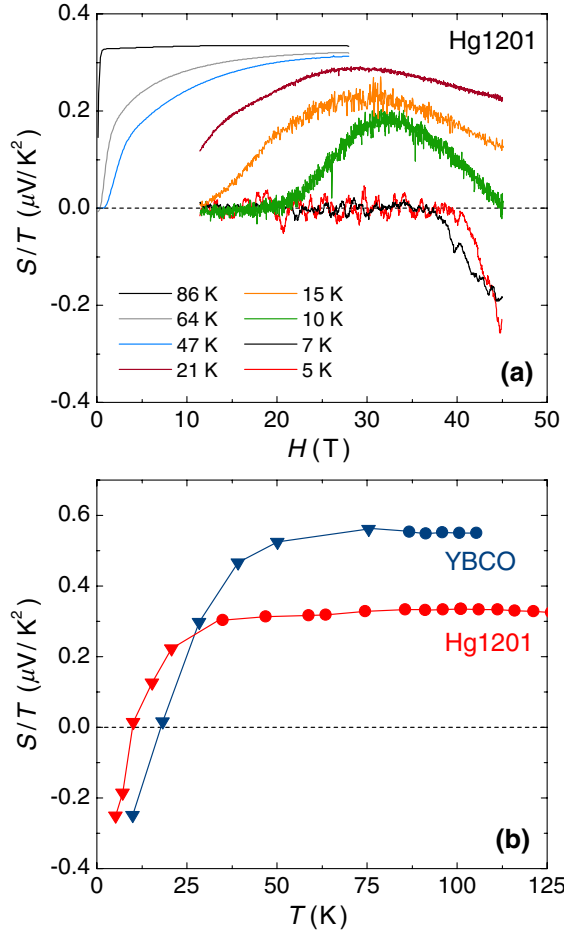


FIG. 2. (a) Seebeck coefficient S of Hg1201 plotted as S/T versus H , in sample A at various temperatures, as indicated. (b) Normal-state Seebeck coefficient S/T of Hg1201 as a function of temperature, at $H = 28$ T (red circles) and $H = 45$ T (red triangles). Corresponding data are shown for YBCO at $p = 0.10$ in $H = 0$ (blue circles) and 28 T (blue triangles), from Ref. [8].

high-temperature behavior well above T_{\max} , at a temperature labeled T_H [19]. Our Hall data on Hg1201 sample B yield approximately $T_{\max} \approx 100$ K and $T_H \approx 240$ K. In YBCO, $T_{\max} \approx 100$ K and $T_H \approx 120$ K at $p = 0.12$ [19]. In Fig. 3, T_{\max} , T_H , and the Hall effect sign-change temperature T_0 in YBCO and Hg1201 are plotted on their respective phase diagrams. This phase diagram is in agreement with the one recently proposed based on the analysis of the dc-resistivity measurements [29]. Notably, the characteristic temperature below which the planar resistivity exhibits Fermi-liquid-like behavior in temperature [29] and frequency [30] approximately matches T_H .

Given that the tetragonal structure of underdoped Hg1201 has no unidirectional character, our findings suggest that density-wave order is a generic tendency of the square CuO_2 plane, and therefore a phenomenon intrinsic to the physics of cuprates. The precise nature of the density-wave order responsible for FSR in Hg1201 remains to be elucidated, e.g., by x-ray scattering.

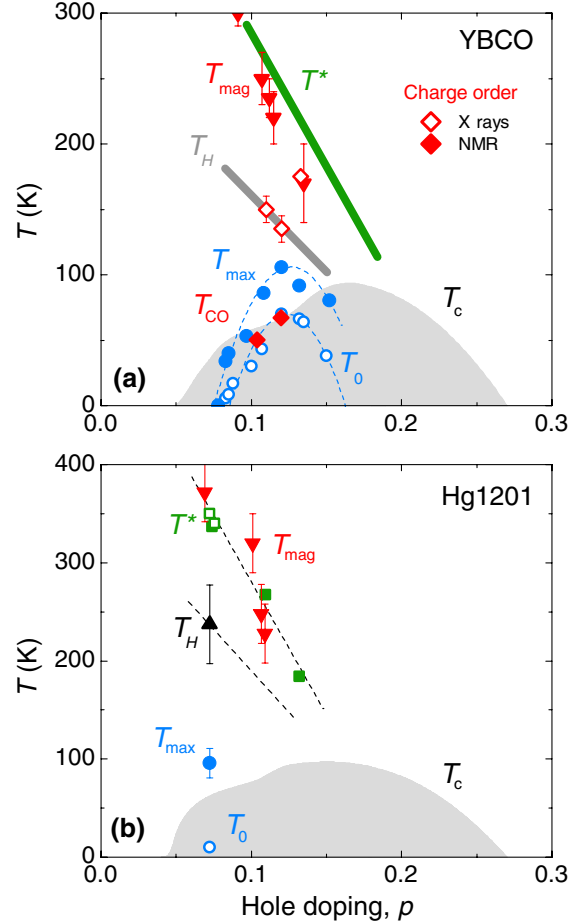


FIG. 3. (a) Temperature-doping phase diagram of YBCO, showing the zero-field superconducting phase below T_c (grey dome, from Ref. [36]) and the onset of $q = 0$ magnetic order below T_{mag} detected by spin-polarized neutron scattering (down triangles, from Ref. [42]). Several characteristic temperatures of the transport properties are displayed: The thick green and grey lines schematically represent the pseudogap temperature T^* , from resistivity and Nernst data [23], and T_H , from $R_H(T)$ [19], respectively. T_{\max} (full circles) and T_0 (empty circles) are determined from $R_H(T)$ [19] (see text). We also show the onset of charge order at T_{CO} via NMR (full diamonds, from Ref. [13]) and the approximate onset of charge modulations via x-ray scattering (open diamonds, from Refs. [14–16]). (b) Corresponding phase diagram for Hg1201, showing T_c (grey dome, from Ref. [22]), T_{mag} (down triangles, from Refs. [39,40]), and T^* from resistivity (full squares, from Refs. [21,39,40]; open squares, from this work). The characteristic temperatures T_{\max} (full circles) and T_0 (empty circles) of $R_H(T)$ are also shown. All dashed lines in both panels are a guide to the eye.

In YBCO at $p = 0.10$ and 0.12 , a modulation of the charge density is detected in the CuO_2 planes by NMR measurements at high fields [13]. It is inferred to be unidirectional, with a period of $4a_0$, where a_0 is the lattice spacing, along the a axis of the orthorhombic lattice for the ortho-II structure at $p = 0.10$. (The pattern could not be determined for the ortho-VIII structure at $p = 0.12$.) Note,

however, that an additional modulation along the b axis cannot be excluded. In Fig. 1(b), we reproduce the NMR data at $p = 0.10$ and see that the onset of charge order, at $T_{\text{CO}} = 50 \pm 10$ K, coincides approximately with the downturn in $R_H(T)$ for a similar doping ($p = 0.10$). The same may be said of $S(T)/T$. Moreover, an increase in doping to $p = 0.12$ causes a parallel increase in both T_{CO} [13] and T_{max} [19] [see Fig. 3(a)], strong evidence that the FSR in YBCO is caused by this charge-density-wave order [31]. In Eu-LSCO, unidirectional stripelike charge order with a period $4a_0$ is detected by x-ray scattering [32], with $T_{\text{CO}} \approx 40$ K at $p = 0.10$ and $T_{\text{CO}} \approx 80$ K at $p = 0.12$ [4], and linked to a drop in $R_H(T)$ [5,12] and in $S(T)/T$ [7,8], again showing that charge order is causing the FSR [31].

Recent x-ray studies of YBCO in zero and low magnetic fields up to 17 T, however, have discovered incommensurate charge modulations (which may not be static) along both the a and b axes, with a period of $3.1a_0$ [14–18]. As seen in Fig. 3, the onset of the x-ray-scattering intensity appears to match T_H , although these crossovers are gradual and there is no sharp anomaly in either the x-ray data or the transport data. Their relation to the charge order seen by NMR at high field below T_{CO} remains to be understood. Recent high-field sound velocity measurements on YBCO at $p = 0.11$ detect the charge order below T_{CO} , and show that it must be a bidirectional charge-density wave (and not domains of two uniaxial density waves) [33]. So, the case of YBCO would appear to differ from the unidirectional charge-stripe scenario observed in the La_2CuO_4 -based materials. But, more work is needed to establish the differences and clarify whether they are fundamental. It has been proposed that a bidirectional charge order is part of the explanation for the reconstructed Fermi surface of underdoped YBCO [34]. Given the striking similarity in the transport properties of YBCO and Hg1201 shown here and in Ref. [29], it is very likely that they host a similar form of charge order.

In YBCO, charge order competes with superconductivity [14–16], probably making T_c fall when FSR sets in [19], below $p \approx 0.16$ (Fig. 3). The competition is manifest in recent measurements of the upper critical field H_{c2} in YBCO [35], which show $H_{c2}(p)$ to be strongly suppressed where charge order exists. Although less pronounced, a similar suppression (with respect to a parabola) with underdoping is observed in the T_c versus p curve, for both YBCO [36] and Hg1201 [22] (see Fig. 3). Certain features of the lattice structure may play a role in stabilizing the charge order, strengthening it more in some materials (e.g., with the low-temperature tetragonal structure). These features would have an impact on the competition between charge order and superconductivity, suppressing T_c more effectively in Eu-LSCO (maximal $T_c \approx 20$ K), where charge order exists at $H = 0$, than in YBCO (where a magnetic field may be needed to fully stabilize charge order [13,33]), for example.

Negative Nernst effect.—In underdoped YBCO, the Nernst coefficient $\nu(T)$ [7,23] also provides hints of FSR. As seen in Fig. 4, the Nernst coefficient ν of YBCO at $p = 0.12$ is small and positive at high temperature, and it drops to large and negative values at low temperatures. (Note that unlike the Hall and Seebeck coefficients, the sign of the Nernst coefficient is not governed directly by the sign of the dominant charge carriers.) This drop was shown to occur at the pseudogap temperature T^* [23], at which the in-plane resistivity $\rho_a(T)$ deviates from its linear temperature dependence at high temperature. Close to T_c , a positive signal due to superconductivity appears [37], but application of a large magnetic field suppresses this signal, revealing that the smooth drop in the normal state ν/T continues monotonically down to $T = 0$ [7,38]. The value of $|\nu/T|$ at $T \rightarrow 0$ is precisely that expected of the electron Fermi surface [8], given its frequency, mass, and mobility, measured via quantum oscillations [9,24]. In other words, the large negative Nernst coefficient in YBCO at low temperature is a consequence of FSR.

As shown in Fig. 4, the Nernst coefficient of Hg1201 is essentially identical to that of YBCO. When plotted versus T/T^* , ν/T has exactly the same temperature dependence in both materials. (T^* in Hg1201 is defined as in YBCO [21,39,40].) We infer that the large negative $\nu(T)$ in Hg1201 is also a manifestation of the FSR. (Note that in YBCO, ν is anisotropic in the ab plane [23,38]. In tetragonal Hg1201, where no such anisotropy is expected, the magnitude of ν lies between the ν_a and ν_b of YBCO.)

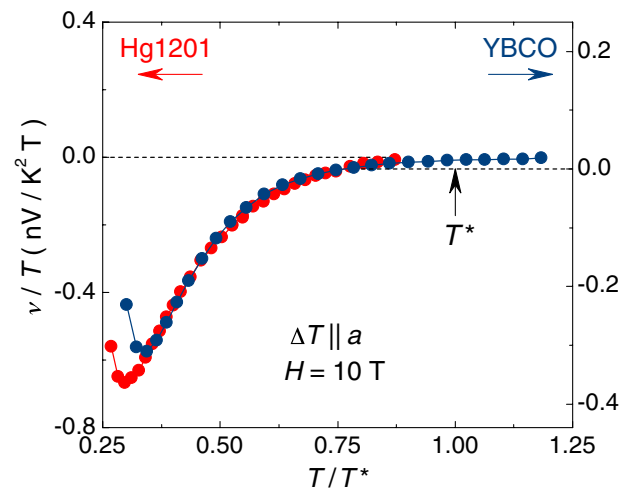


FIG. 4. Nernst coefficient ν of Hg1201 ($T_c = 65$ K; red circles, sample A) and YBCO ($T_c = 66$ K, $p = 0.12$; blue circles, from Ref. [23]), plotted as ν/T versus T/T^* , where T^* is the pseudogap temperature as defined in the main text. Here, $T^* = 340$ K for Hg1201 and $T^* = 250$ K for YBCO. The magnetic field $H = 10$ T is along the c axis, and the heat gradient is along the a axis. Note the different vertical scales for Hg1201 (red arrow, left axis) and YBCO (blue arrow, right axis). Below $T/T^* = 1.0$, the quasiparticle signal falls gradually to reach large negative values, in identical fashion in the two materials.

Summary and outlook.—Our high-field measurements of Hall and Seebeck coefficients in the tetragonal single-layer cuprate Hg1201 reveal that its normal-state Fermi surface undergoes a reconstruction in the underdoped regime at low temperature, which produces an electron pocket. This Fermi-surface reconstruction is compelling evidence for the presence of a density-wave order that breaks translational symmetry. The remarkable similarity of these transport properties with those of the orthorhombic bilayer cuprate YBCO strongly suggests that the charge-density-wave order observed in YBCO is also responsible for the FSR in Hg1201, and is thus a generic property of hole-doped cuprates. The presence of charge-density-wave order in the midst of the phase diagram of cuprate superconductors raises some fundamental questions. Is the enigmatic pseudogap phase a high-temperature precursor of the charge order at low temperature? Is the quantum critical point for the onset of charge order responsible for the anomalous properties of the normal state, such as the linear- T resistivity? Are fluctuations of the charge order involved in pairing? Our findings in Hg1201 broaden the scope for exploring these questions by adding a clean archetypal cuprate to the list of materials that exhibit all the key properties of hole-doped cuprates, including superconductivity with a high T_c , a pseudogap phase with novel magnetism [39–41] (Fig. 3), and Fermi-surface reconstruction from charge-density-wave order.

A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490, the State of Florida, and the U.S. Department of Energy. Part of this work was performed at the Laboratoire National de Champs Magnétiques Intenses, supported by the EU Contract No. 228043 and the ANR Superfield. The work at Sherbrooke was supported by a Canada Research Chair, CIFAR, NSERC, CFI, and FQRNT. The work at the University of Minnesota (crystal growth, annealing, characterization, and contacting of samples) was supported by the U.S. Department of Energy, Office of Basic Energy Sciences. J.C. was supported by the Swiss National Science Foundation. N.B. acknowledges the support of a Marie Curie Fellowship.

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