Thermoelectric signature of the nematic phase in hole-doped iron-based superconductor

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Studies of the copper-based superconductors demonstrate how their phase diagram becomes more complex as experimental probes improve, able to distinguish among subtly different electronic phases. One of those phases, nematicity, has become the matter of great interest also in the iron-based superconductors, where it is detected deep in the tetragonal state. Here we present the evolution of the in-plane Nernst effect anisotropy in the strain detwinned Ca(Fe1-xCo_x)2As2 single crystals, whose behaviour can be explained within the approach developed to describe the nematic order parameter in liquid crystals [1,2]. Furthermore, the employed method turns out to be universally applicable to data from other superconductors: Ba(Fe1-xCo_x)2As2 [3] as well as YBa2Cu3O7 [4]. We conclude the observed broken rotational symmetry of the electronic system is a consequence of the emerging thermodynamic electronic nematic order at a temperature much higher than onset of the magnetic and structural transitions.
Introduction

The phase diagram of the copper-based superconductors, which initially consisted of the superconducting and antiferromagnetic phases [5] was subsequently supplemented with regions of occurrence of other electronic orders. These include the pseudo-gap [6], spin density wave (SDW) [7] as well as three-dimensional (3D) and two-dimensional (2D) charge density wave (CDW) [8,9]. It appears the presence of some kind of electronic nematic order is a pervasive characteristic of unconventional superconductors [10,11], which applies also to iron-based superconductors, where the electronic nematic phase is a distinguished feature of their phase diagram [12]. While there is no agreement yet about a mechanism that leads to breaking of rotational symmetry in these electron systems, the phenomenon itself has gathered wide experimental support. It was extensively investigated by spectroscopic [13,14] and macroscopic [15,16] probes including studies of the thermoelectrical response anisotropy [3,17]. The latter has turned out to be an invaluable source of information about the nematic phase in cuprates [4,18]. Furthermore, it was argued that this method could detect a type of nematicity that was unreachable by electrical resistivity studies [4].

Here we report measurements of the Nernst effect anisotropy, which indicate emergence of the nematic phase in the electron doped 122 iron-based superconductors. Notably, the data suggest that at high temperatures we do not observe fluctuations of the low-temperature nematic order, but rather a development of a fully set nematicity. A clue that this might be the case comes from the recent studies of the in-plane thermoelectric anisotropy in the strain-detwinned Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ iron-based superconductor [3]. Those results include data on the anisotropy of the Nernst effect ($\Delta\nu$) defined as a difference between the Nernst coefficient determined in the magnetic field parallel to the crystallographic $c$ axis with the thermal gradient imposed either along the $b$ axis ($\nu_b$) or the $a$ axis ($\nu_a$). The striking feature is that $\Delta\nu$ in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is not affected by the magnetic/structural transition at $T_{tr}$ and $\Delta\nu(T)$ develops smoothly down to
the lowest temperatures changing almost linearly in the semi-logarithmic scale. The onset of
the Nernst anisotropy occurs at a temperature significantly higher than \( T_{tr} \), although the fact that
the magnetic/structural transition in \( \text{Ba}(\text{Fe}_{1-x}\text{Co}_x)\text{As}_2 \) is identified as being second order [19]
does not allow one to exclude that non-zero \( \Delta \nu \) comes as a result of the critical nematic or
magnetic fluctuations. The case of \( \text{Ca}(\text{Fe}_{1-x}\text{Co}_x)\text{As}_2 \) discussed here is different because the
transition in this compound has been classified as being first-order (discontinuous) [20,21], thus
no substantial contribution from fluctuations is expected.

**Methods**

Single crystals of \( \text{Ca}(\text{Fe}_{1-x}\text{Co}_x)\text{As}_2 \) were grown using the Sn-flux method. The Ca, Fe,
As, Co, and Sn elements in molar ratios of \( 1 : (2-x) : 2 : x : 30 \) (\( x = 0, 0.06 \) and 0.2 respectively
for \( \text{CaCo0} \; \text{CaCo3} \) and \( \text{CaCo7} \)) were loaded into alumina crucibles and sealed in quartz ampules
under vacuum. The ampules were heated slowly to 1050 C, kept at this temperature for several
hours, and then cooled down slowly to 650 C at a rate of 2 C/h. Next, the liquid tin was decanted
from the crucibles. The Sn residues on the crystals were removed via etching in diluted
hydrochloric acid. The chemical composition of a Co-doped single crystal was determined by
the energy dispersive x-ray analysis.

The Hall coefficient was measured in unstrained crystals in a magnetic field of
\( B = 12.5 \text{ T} \). Then a sample was mounted between two clamps made of phosphor bronze and
subjected to a 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 electrical resistivities \( \rho_a \) and \( \rho_b \) to be
determined using the Montgomery method [22]. The uniaxial pressure was increased step-by-
step and measurements of the resistivity were repeated until a saturation of the anisotropy,
indicating maximal detwinning, was achieved. The maximal pressure determined in this way was used during subsequent thermoelectric experiments.

The Nernst coefficient was measured along and across the strain direction in two separate runs with the magnetic field (parallel to $c$-axis) varied from -12.5 T 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 millimetres long and 100 µm thick silver wires. Signal leads were made up of long pairs of 25 µm phosphor bronze wires. $\nu_a$ and $\nu_b$, which differ within several percent due to slightly different geometrical factors in two experimental configurations are matched by applying a multiplicative correction factor close to 1. More details about the experimental setup are given in Ref. 3.

Results

Figure 1a presents electrical resistivity measurements performed along the long ($a$) and short ($b$) crystallographic axes in a series of the strain-detwinned $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)\text{As}_2$ single crystals with $x = 0$ (CaCo0), 0.03 (CaCo3), and 0.07 (CaCo7). In all samples the transition to the orthorhombic SDW state affect mostly the electrical transport along the $b$ axis. The anisotropy of the resistivity disappears rather quickly in the tetragonal state as shown in Fig. 1b, where the normalized temperature dependences of the resistive anisotropy ($\Delta\rho = (\rho_b - \rho_a)/(\rho_b + \rho_a)$) form a sharp step in CaCo0 at 167 K, a two steps transition in CaCo3 at 148 K (where the structural and magnetic transitions are separated by about 10 K) and a rather smooth development of $\Delta\rho$ below 100 K in CaCo7. The Hall coefficient, which temperature dependences in the vicinity of the transitions are presented in Fig. 1c, exhibits an anomaly at the respective temperatures in all three samples, which is not exactly the case for the Nernst coefficient presented in Fig. 2a. While $\nu(T)$ in CaCo0 and CaCo3 exhibit a clear
step-like change at the structural/magnetic transition (which might be related to the presence of the Dirac fermions in the SDW phase of the 122 iron based superconductors [23,24]) such an anomaly is absent in CaCo7. Figure 2b, presenting the temperature and doping evolution of the Nernst anisotropy calculated as \( \Delta \nu = \nu_a - \nu_b \), indicates that \( \Delta \nu \) in CaCo0 and CaCo3 changes sign below \( T_{tr} \), whereas for all three samples \( \Delta \nu \) is positive in the tetragonal state. This is different from Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\), where \((\nu_a - \nu_b)\) is negative for all samples being studied except of the most doped Ba(Fe\(_{0.94}\)Co\(_{0.06}\))\(_2\)As\(_2\), where \( \Delta \nu \) is small but positive. Interestingly the onset of \( \Delta \nu \) shown in Fig. 2c occurs at the temperatures significantly higher than each respective \( T_{tr} \). Namely, they are: \( T^* \approx 275, 280 \) and 250 K for CaCo0, CaCo3, and CaCo7, respectively. Remarkably, these values placed on the \( x-T \) phase diagram shown in Fig. 3 closely correspond to the analogous temperatures determined for the Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) series [3]. The phase diagrams for both compounds are normalised to match the range of existence of the SDW phase, ie. \( T \) values are divided by the \( T_{SDW} \) for respective parent compounds (BaFe\(_2\)As\(_2\) or CaFe\(_2\)As\(_2\)), whereas \( x \) values are divided by the respective \( x_{SDW} \) that is the cobalt content at which SDW disappears in \( T = 0 \) K limit.

Perhaps the most important outcome of this work is the set of temperature dependencies of the absolute value of \( \Delta \nu \) shown in Fig. 4a. Plotted is the Nernst anisotropy divided by \( T \) to account for the fact that the Nernst coefficient, being a measure of transverse entropy flow [25], inevitably decreases with decreasing temperature. At the structural/magnetic transition \( |\Delta \nu|/T \) changes abruptly in CaCo0 and CaCo3, perhaps due to a potent contribution to the Nernst coefficient from the afore mentioned Dirac fermions or as a signature of the strong first order character of the transition [12]. In contrast, \( |\Delta \nu|/T \) in CaCo7 seems to be unaffected by the formation of the orthorhombic/SDW state. Notably, the character of the \( |\Delta \nu|/T \) temperature dependence is shared among all the samples studied, and behaviour of \( |\Delta \nu|/T(T) \) is far from the \( \frac{c}{T-T_c} \) form expected for the Curie-Weiss behaviour. The latter was suggested to be caused by
(with some deviations) critical fluctuations of the nematic state in iron-based superconductors studied by means of elastoresistance measurements [16]. As a matter of fact, our data are much better fitted by the equation proposed by I. Haller to describe behaviour of the nematic order parameter \( Q \) in liquid crystals:\[ Q = (1 - \frac{T}{T^*})^\beta, \] where \( T^* \) is a characteristic temperature somewhat lower than the actual temperature of the discontinuous nematic-isotropic transition.

The actual fits: \( \frac{|\Delta\nu|}{T} = \alpha(1 - \frac{T}{T^*})^\beta \) have only two free parameters (\( \alpha \) and \( \beta \)) since \( T^* \) values are taken as the temperatures of the Nernst anisotropy onset (Fig. 2c). Here \( \alpha \) is the proportional constant and \( \beta \) is the pseudo-critical exponent, which according to A. Ranjkesh et al. is supposed to rise with concentrations of impurities [2]. Indeed, as it is shown in Fig. 4b, \( \beta \) in the cobalt doped 122 iron-based superconductors satisfies this rule increasing with \( x \) (since growing substitution of iron with cobalt leads inevitably to an increased disorder). On the other hand, the value of \( \beta \) is about one order of magnitude higher in \((Ca,Ba)(Fe_{1-x}Co_x)_{2}As_2\) than observed in liquid crystals [1,2]. Also significant is that the data from an analogous experiment performed on \( YBa_2Cu_3O_y \) [4], presented in Fig 4c, can also be very well fitted with the Haller equation (this time \( T^* \) is taken as a free parameter).

Such a universal behaviour of the \( |\Delta\nu|/T \) temperature dependence along with the fact that \( |\Delta\nu|/T \) is unaffected by the structural/magnetic transition in \( CaCo7 \) as well as in \( Ba(Fe_{1-x}Co_x)_{2}As_2 \) with \( x = 0.02 \) and 0.04 [3], suggests the anisotropy of the Nernst coefficient might be a proxy of the electronic-nematic order parameter. This idea was already considered by R. Daou et al. [18], which were inspired by the analogous proposition by R.A. Borzi et al. [26] to define the phenomenological nematic order parameter in \( Sr_3Ru_2O_7 \) as the in-plane anisotropy of the electrical resistivity. A remaining question is why we observe in the thermoelectrical studies the broken rotational symmetry, which is inaccessible to electrical measurements and vice versa – that is, why the anomaly at the transition clearly visible in \( \Delta\rho(T) \) is absent in \( \Delta\nu(T) \)? While we cannot provide a firm answer, there are possible scenarios that
could explain such an intriguing behaviour. First, there are well known examples of electronic systems with decoupled charge and entropy fluxes. One of them is the superconducting state where Cooper pairs conduct charge without dissipation but do not contribute in entropy transport. Another are small angle electron-phonon scattering processes, which effectively disturb entropy transport with only little effect on the charge transfer. Remarkably, the latter play an important role in strongly correlated electron system, where charge transport could be described as the flow of a hydrodynamic electron fluid [27]. In such a case the energy dissipation is dominated by momentum conserving small angle electron-phonon scattering [28]. Therefore, if charge carriers in the electronic-nematic state suffer from the anisotropic, but small-angle scattering, it will be seen in the anisotropy of the Nernst effect, but be insignificant for the electrical resistivity measurements. A similar phenomenon was recently observed in YBa$_2$Cu$_3$O$_y$ and attributed to the emergence of the short-range charge density wave modulations [4]. On the other hand, a possible reason why the anomaly at the magnetic/structural transition is absent in the $|\Delta \nu|/T$ temperature dependence was proposed in Ref. 3. Namely, the large angle elastic scattering might cancel out for the Nernst anisotropy which consists of contributions from both $a$- and $b$-direction transport. In fact, $\Delta \nu$ can be expressed as a subtraction of a two Sondheimer cancellations [29], one for $a$-direction, another one for $b$-direction [3], and in metals the resulting difference might be insignificant.

**Conclusion**

In summary, we show that in the phase diagram of iron-based superconductors, in a way similar to their copper-based counterparts, there is room for another electronically driven ordered state. The electronic-nematic phase emerges at high temperature and can develop smoothly despite the occurrence of the magnetic/structural transition. The present form of the phase diagram does not exclude that the electronic-nematic phase ceases when the
superconducting critical temperature reaches its maximum, which would once again be a common behaviour for iron-based and copper-based superconductors [30]. Perhaps it might even suggest an intimate relation between the electronic nematicity and superconductivity [31].
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Figures

Figure 1.

(Color online) **In-plane electrical transport in the Ca(Fe$_{1-x}$Co$_x$)$_2$As$_2$ series.** a, Temperature dependences of the in-plane electrical resistivity (normalised to the high temperature, $T = 290$ K, value) for the detwinned single crystals. There is a clear difference between $\rho$ measured along the long ($a$) and short ($b$) crystallographic axis in the orthorhombic/SDW state. b, Temperature dependences of the normalised resistivity anisotropy $(\rho_b - \rho_a)/(\rho_a + \rho_b)$ showing no sign of a significant difference between $\rho_a$ and $\rho_b$ high above the structural transition. c, Anomalies in the temperature dependences of the Hall coefficient occurring at the structural/magnetic transition.
Figure 2.
(Color online) **In-plane magneto-thermoelectric response in the Ca(Fe$_{1-x}$Co$_x$)$_2$As$_2$ series.**  
**a,** Temperature dependences of the Nernst coefficient for the heat current being imposed along either $a$ or $b$ orthorhombic axis. CaCo3 and CaCo0 plots are shifted vertically by $1 \times 10^{-10}$ and $2 \times 10^{-10}$ V K$^{-1}$ T$^{-1}$, respectively, for the sake of clarity with dashed lines denoting the corresponding $\nu = 0$ V K$^{-1}$ T$^{-1}$ levels. 
**b,** Temperature dependence of the Nernst coefficient anisotropy defined as $\Delta \nu = \nu_a - \nu_b$.  
**c,** High temperature onset of the Nernst anisotropy.
Figure 3.

(Color online) Schematic $x$-$T$ phase diagram of Ca(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [32] and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [33] with marked positions of the onset of the Nernst anisotropy. Results for Ca(Fe$_{1-x}$Co$_x$)$_2$As$_2$ are marked with full red points and for Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ with hollow red points. Yellow and dark yellow regions in the phase diagram denote spin density wave phase; pink and dark pink -- orthorhombic, nonmagnetic phase; blue and dark blue -- superconducting phase.
Anisotropy of the Nernst coefficient as a measure of the electronic nematic order parameter in iron- and copper-based superconductors. a, Temperature dependences of the absolute value of $\Delta\nu/T$ for the Ca(Fe$_{1-x}$Co$_x$)$_2$As$_2$ (solid symbols) and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ (hollow symbols) series, where “(+)” and “(-)” marks denote the positive and negative sign of $\Delta\nu/T$, respectively. Solid lines are fits to the temperature dependence of the nematic order parameters considered in liquid crystals [1], whereas dashed line is an exemplary Curie – Weiss type dependence $\Delta\nu/T = 5 \times 10^{-10} \frac{T}{T-20}$. b, Changes of the $\beta$ parameter with doping that are supposedly reflecting the increasing level of disorder. c, Nernst anisotropy in YBa$_2$Cu$_3$O$_y$, where $\Delta\nu_{200} = \Delta\nu(T) - \Delta\nu(200\,\text{K})$ and the temperature axis is also normalized $^4$. Solid lines give fits for the nematic order parameter as mentioned in the text.
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