

Nematicity in $\text{LaFeAsO}_{1-x}\text{F}_x$

C. Hess^{*,1,2}, H. Grafe¹, A. Kondrat¹, G. Lang^{1,3}, F. Hammerath^{1,4}, L. Wang^{†,1}, R. Klingeler⁵,
G. Behr^{‡,1}, and B. Büchner^{1,2,4}

¹ Leibniz-Institute for Solid State and Materials Research, IFW-Dresden, 01069 Dresden, Germany

² Center for Transport and Devices of Emergent Materials, TU Dresden, 01069 Dresden, Germany

³ ESPCI, PSL Research University; CNRS; Sorbonne University, UPMC; LPEM, 10 rue Vauquelin, 75005 Paris, France

⁴ Institute for Solid State Physics, TU Dresden, 01069 Dresden, Germany

⁵ Kirchhoff Institute of Physics, Heidelberg University, 69120 Heidelberg, Germany

Received 13 April 2016, revised 9 June 2016, accepted 20 June 2016

Published online 7 July 2016

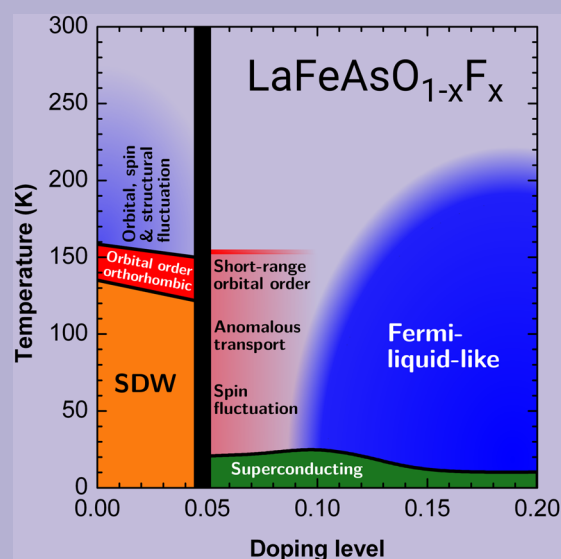
Keywords iron pnictide superconductors, magnetic resonance, nematic order, transport

* Corresponding author: e-mail c.hess@ifw-dresden.de, Phone: +49-351-4659533, Fax: +49-351-4659313

† Present address: Institute for Solid-State Physics, Karlsruhe Institute of Technology, 76021 Karlsruhe, Germany.

‡ Deceased.

Orbital ordering has recently emerged as another important state in iron-based superconductors, and its role for superconductivity as well as its connection to magnetic order and orthorhombic lattice distortion are heavily debated. In order to search for signatures of this so-called nematic phase in oxypnictides, we revisit the normal state properties of the pnictide superconductor $\text{LaFeAsO}_{1-x}\text{F}_x$ with a focus on resistivity, Nernst effect, thermal expansion, and ^{75}As nuclear magnetic resonance (NMR) data. The transport properties at the underdoped level $x = 0.05$ exhibit pronounced anomalies at about the same temperature where undoped LaFeAsO develops long-range nematic ordering, i.e., at about 160 K. Furthermore, the ^{75}As -NMR spin-lattice relaxation rate $(T_1 T)^{-1}$ reveals a progressive slowing down of spin fluctuations. Yet, long-range magnetic order and also a detectable orthorhombic lattice distortion are absent. Thus, we conclude from the data that short-range orbital-nematic ordering or a slowly fluctuating form of it sets in near 160 K. Remarkably, all anomalies in the transport and also the indications of slow spin fluctuations disappear close to optimal doping $x = 0.1$ which suggests that in $\text{LaFeAsO}_{1-x}\text{F}_x$ the nematic phase actually competes with superconductivity.



Schematic electronic phase diagram of $\text{LaFeAsO}_{1-x}\text{F}_x$.

© 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction The discovery of superconductivity in $\text{LaFeAsO}_{1-x}\text{F}_x$ [1] initiated a tremendous research effort which yielded soon after a large variety of new superconducting iron pnictide compounds with T_c up to 55 K [2]. All compounds feature Fe_2As_2 -layers as the common structural unit, with typical examples being RFeAsO ($\text{R} = \text{La}$ or Rare Earth), AFe_2As_2 ($\text{A} = \text{alkaline earth or Eu}$), LiFeAs , and FeSe , which commonly are referred to as “1111”, “122”, “111”, and “11” compounds, respectively (note that Se replaces As in the last example). While

the latter two compounds exhibit superconductivity already in their stoichiometric form, the parent materials RFeAsO and AFe_2As_2 are poor metals which exhibit an antiferromagnetic spin density wave (SDW) ground state. The temperature-dependent transition to this state is always accompanied by a tetragonal-to-orthorhombic transition either at the same or at somewhat higher temperature. Chemical doping destabilizes this state in favor of superconductivity. The obvious proximity of superconductivity and antiferromagnetism very early has lead to the conjecture that spin

fluctuations are the driving mechanism of superconductivity [3]. It has been noticed [4–6] that the structural transition in the parent compounds is unlikely due to steric effects, i.e., phonon-driven, but rather electronically driven. In fact, evidence for additional orbital ordering occurring concomitantly with the structural distortion has recently inferred, e.g., from resistivity anisotropy [7, 8] and angular resolved photoemission spectroscopy (ARPES) [9]. Thus, there are three types of long-range order which occur upon entering the low-temperature orthorhombic SDW state, *viz.*, orbital ordering, the orthorhombic distortion, and the antiferromagnetic SDW order. All these types of ordering are coupled with each other and break C_4 -symmetry and it is an ongoing question which of the three order parameters is driving the transition to the low-temperature phase that has been dubbed “nematic” phase, and to which extent it supports or competes with superconductivity [6].

The most comprehensive investigations of nematic order and fluctuations of it concern 122 and 11 phases [7–12] whereas relatively little is known for the 1111 phases. Viable routes to reach superconductivity in RFeAsO through chemical doping are the substitution of interlayer O by F [1] and the substitution of Co for Fe within the Fe_2As_2 layers in both 1111 and 122 compounds [13]. A generic observation upon doping is that the SDW phase is destabilized, i.e., the T_s and T_N gradually decrease and at some finite doping level superconductivity emerges. The actual nature of the transition from SDW to superconductivity is much under debate. There is evidence that in $\text{LaFeAsO}_{1-x}\text{F}_x$, on which we focus in this paper, the transition is abrupt and first order-like toward a homogeneous superconducting phase [14, 15] while in all other systems experiments suggest a finite doping interval where superconductivity and static magnetism coexist [16].

In the parent compound LaFeAsO the structural tetragonal-to-orthorhombic transition takes place at $T_s \approx 160$ K [14, 17] and the long-range antiferromagnetic SDW order sets in at $T_N \approx 137$ K [14, 18–20]. These successive phase transitions can be well detected due to pronounced signatures in transport, thermodynamic, magnetic, and structural quantities. In this paper, we revisit previous experimental results for $\text{LaFeAsO}_{1-x}\text{F}_x$ in the light of the more recently established scenario for an electronic-nematic ordering that accompanies the structural transition in the parent compound investigate the implications for finite doping levels. We compare transport data of $\text{LaFeAsO}_{1-x}\text{F}_x$ from resistivity ρ [15] and the Nernst coefficient ν [21] with thermal expansion [22] and nuclear magnetic resonance (NMR) [23] data obtained on the same samples at selected doping levels $x = 0, 0.05$, and 0.1 . This allows to place and rationalize the previous results and conclusions in the larger picture of nematicity in $\text{LaFeAsO}_{1-x}\text{F}_x$. In particular, we observe that the distinct anomalies, present in both resistivity and the Nernst coefficient at the underdoped level $x = 0.05$, occur at roughly the same temperature (~ 160 K) as the nematic ordering temperature in the parent compound. The ^{75}As spin-lattice relaxation rate $(T_1 T)^{-1}$ shows a strong increase upon cooling, signaling the slowing-down of spin fluctuations [23], but long-range

magnetic order as well as a global orthorhombic distortion is never established [14, 17, 20, 24].

These qualitative features disappear close to optimal doping $x = 0.1$. We conclude from these findings that short-range or fluctuating orbital ordering is still present in underdoped superconducting $\text{LaFeAsO}_{1-x}\text{F}_x$ but disappears when optimal superconductivity is reached. The results suggest further that in $\text{LaFeAsO}_{1-x}\text{F}_x$ the nematic phase competes with superconductivity.

2 Experimental Sample preparation and all experimental work have been reported separately in earlier works [15, 17, 22, 23], and experimental details can be inferred from the respective publications.

3 Results

3.1 The parent compound LaOFeAs The electrical resistivity, $\rho(T)$, of LaFeAsO (see Fig. 1a) develops a deviation from a standard metallic linear T -dependence near 300 K upon cooling which leads to a maximum at T_s and a subsequent sharp drop with an inflection point at T_N [1, 15, 19, 25]. A further decrease of temperature T leads to a minimum of $\rho(T)$ at ~ 90 K followed by a strong low- T upturn. The observed anomalies in the resistivity upon passing through T_s and T_N coarsely represent a canonical behavior of the resistivity upon entering the SDW ground state not only of many other iron pnictides such as BaFe_2As_2 but also other very different SDW compounds like as Mn_3Si [26] or $\text{Yb}_2\text{Pt}_2\text{Pb}$ [27]. In $\text{LaFeAsO}_{1-x}\text{F}_x$, due to the complex interplay of orbital, magnetic, and structural degrees of freedom, the situation is, however, a bit more involved as compared to the transition to a presumably pure SDW state in the latter two compounds. The detailed origin of such anomalous transport properties is currently not clear. Qualitatively, it seems straightforward, however, to rationalize the observed anomalies in terms of enhanced scattering at $T > T_s$, presumably arising from some form of nematic fluctuations, and a reduced carrier density together with a dramatically enhanced transport relaxation time in the long-range orbital ordered orthorhombic state, signaled by the dramatic drop of $\rho(T)$ below T_s . The inflection point upon cooling through T_N signals that the decrease of the resistivity becomes weaker in the magnetically ordered state, presumably due to the additional opening of an SDW gap which further reduces the carrier density.

The actual nature of the nematic fluctuations which give rise to the enhanced $\rho(T)$ at $T > T_s$ is uncertain. However, there is strong evidence that fluctuations of orbital, lattice, and spin degrees of freedom are ubiquitous which all potentially couple to the resistivity: on the one hand, the Nernst coefficient $\nu(T)$ [21] as shown in Fig. 1b not only exhibits a dramatic enhancement upon cooling through T_s , there is furthermore a clear fluctuation regime of an already enhanced $\nu(T)$ that extends almost up to room temperature, see inset of Fig. 1b. This quantity is known as a sensitive probe for reconstructions of the Fermi surface due to electronic-nematic or SDW order [26, 28–33]. Pure lattice effects typically are of secondary importance. Previously, the “giant” enhancement

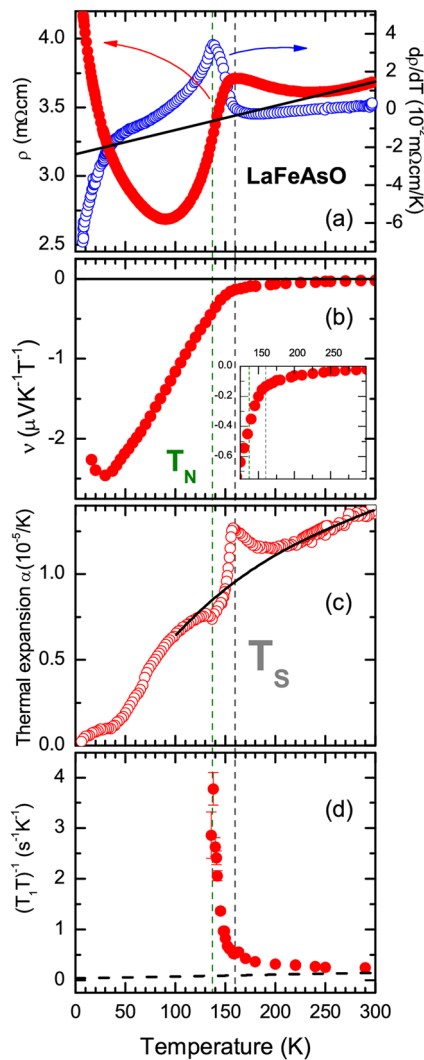


Figure 1 Normalized resistivity $\rho(T)$ and the derivative $d\rho/dT$ [15] (a), the Nernst coefficient $\nu(T)$ [21] (b), thermal expansion [22] (c), and $(T_1 T)^{-1}$ [23] (d) of LaFeAsO. Solid and dashed lines are guides to the eye.

of the Nernst coefficient in LaFeAsO below T_s has quite generally been attributed to the Fermi surface reconstructions due to the SDW state [21]. However, the fact that the enhancement clearly sets in close to T_s , i.e., above the magnetic ordering temperature T_s , with practically no specific anomaly at T_N , implies the Nernst effect to respond primarily to orbital ordering. This suggests the fluctuation regime of $\nu(T)$ at $T > T_s$, to be rather specifically connected to orbital fluctuations.

On the other hand, an extended *lattice* fluctuation regime is observed in thermal expansion data at $T > T_s$ [22], see Fig. 1c. These anomalous contributions to $\alpha(T)$ can be interpreted as a signature of elastic lattice softening. In the setup applied here where the samples experience a moderate compression along the direction of the detected length changes [34], elastic softening is associated with shrinking of the sam-

ple showing up as a positive anomaly in $\alpha(T)$. The observed extended regime of anomalous length changes well above T_N hence probes the elastic soft mode which in case of an electronic origin of the structural transition can be associated with the nematic susceptibility [8, 10]. Measurements of the elastic shear modulus performed in a capacitance dilatometer have indeed been used to obtain the nematic susceptibility of electron- and hole-doped BaFe₂As₂ and FeSe [11, 35].

The presence of lattice fluctuations well above T_N is corroborated by the predominantly phononic heat conductivity κ of LaFeAsO which exhibits a strong dip-like anomaly at $T \gtrsim T_s$ [17, 25] which also signals that structural fluctuations are relevant. Finally, *magnetic* fluctuations at least up to about 200 K can be clearly inferred from the ⁷⁵As-NMR spin lattice relaxation rate, $(T_1 T)^{-1}$, which diverges at T_N [36] (see Fig. 1d). Since the NMR relaxation rate is proportional [37] to the imaginary part of the dynamic spin susceptibility at the Larmor frequency, $(T_1 T)^{-1} \sim \chi''(\mathbf{q}, \omega_0)$, (with the Larmor frequency ω_0 and the wave vector \mathbf{q}), this increase shows the slowing down of spin fluctuations prior to the SDW ordering.

3.2 Underdoped LaO_{1-x}F_xFeAs It is enlightening to directly compare the above observations for the parent compound LaFeAsO with data at finite doping levels. Figure 2 presents our data for $\rho(T)$, $\nu(T)$, and $(T_1 T)^{-1}$ of LaFeAsO_{1-x}F_x at the underdoped level $x = 0.05$, close to the “border” to the long-range ordered nematic phase. As can be inferred very clearly from the data, the two transport quantities exhibit very clear anomalies at practically the same temperature ($T \sim 160$ K) as in the parent compound: the dramatic drop of $\rho(T)$ at about 160 K seen in the parent compound has been replaced by a much weaker but distinct anomaly (Fig. 2a): $\rho(T)$ drops below the low- T extrapolation of its linear high-temperature behavior at about 160 K, which suggests, as in the parent compound, an enhancement of the transport relaxation time [15]. Note, that prior to the onset of T_c an upturn is present which indicates the localization of charge carriers.

The Nernst coefficient shown in Fig. 2b exhibits a very clear change of slope at $T \sim 160$ K and a dramatic drop including a sign change upon cooling. In view of the above interpretation of the Nernst coefficient of the parent compound, this strong anomaly suggests the onset of orbital or SDW ordering at $T \sim 160$ K. Note, however, that the magnitude of the negative contribution that appears at this temperature is about two orders of magnitude smaller than that in the parent compound, i.e., the overall effect is much more subtle. Thus, both resistivity and Nernst effect reveal that a remnant feature of the nematic order is still governing the physics of this compound. For completeness, we mention the further upturn of $\nu(T)$ at lower temperature due to the onset of vortex motion in the superconducting state [21]. It will not be discussed further in this paper as it solely focuses on the normal state properties.

In order to elucidate to which degree magnetism is affected, we plot $(T_1 T)^{-1}$ in Fig. 2c [23]. $(T_1 T)^{-1}$ exhibits a

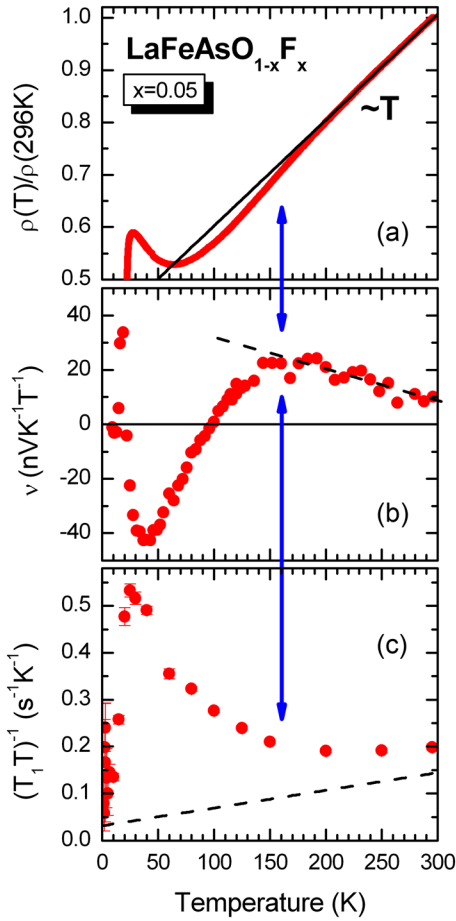


Figure 2 Normalized resistivity $\rho(T)$ [15] (a), Nernst coefficient $\nu(T)$ [21] (b), and ^{75}As -NMR spin-lattice relaxation rate $(T_1 T)^{-1}$ [23] (c) of $\text{LaFeAsO}_{1-x}\text{F}_x$ at $x = 0.05$ with $T_c = 20.6$ K [17]. Solid and dashed lines in (a) and (b) are guides to the eye. Dashed line in (c): see text.

weak positive slope at high temperature $T \gtrsim 200$ K and approaches a linear increase (dashed line in Figs. 1c, 2c, and 3c) [23]. This behavior resembles the generic linear of the static susceptibility [20] that has been heavily debated in the past [20, 38–46], but is not at focus here. However, at lower temperature $(T_1 T)^{-1}$ strongly deviates from this behavior and develops a strong increase with decreasing T until it starts to decrease again after reaching a peak at about 28 K. Note that the magnitude of the increase is about one order of magnitude smaller than that related to the SDW ordering in the parent compound. The observed low-temperature peak in $(T_1 T)^{-1}$ can be well rationalized within the Bloembergen–Purcell–Pound (BPP) model and signals the progressive slowing-down of spin fluctuations of the conduction electrons [23]. These data show that the spin dynamics at this doping level, in consistency with Mößbauer and muon spin rotation (μSR) results [14], are close to but never reach long-range magnetic order.

The thermal expansion at this doping level (not shown, see Fig. 2 of Ref. [22]) as well as the orthorhombic split-

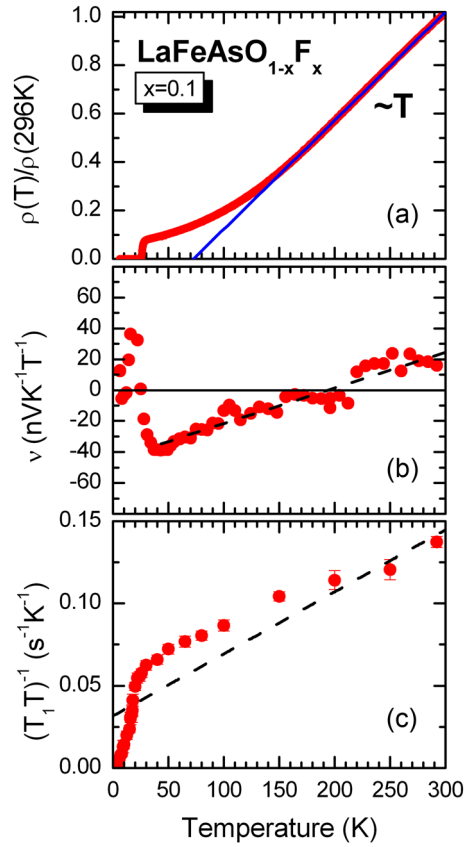


Figure 3 Normalized resistivity $\rho(T)$ [15] (a), Nernst coefficient $\nu(T)$ [21] (b), and ^{75}As -NMR spin-lattice relaxation rate $(T_1 T)^{-1}$ [23, 39] (c) of $\text{LaFeAsO}_{1-x}\text{F}_x$ at $x = 0.1$ with $T_c = 26.8$ K [17]. Solid and dashed lines in (a) and (b) are guides to the eye. Dashed line in (c): see text.

ting in diffraction experiments [14, 17, 24] do not show any anomaly around 160 K. The global lattice degrees of freedom are therefore apparently unaffected by the remnant nematic ordering, i.e., a long-range structural influence is absent. Thus, the well-defined anomalies in the resistivity and the Nernst coefficient signal some form of orbital ordering, which must be of short-range nature because otherwise a global response in the lattice degrees of freedom would be inevitable.

3.3 Optimally doped $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$ We point out that the salient features of the remnant orbital ordering in the transport properties as well as the signature of progressive slowing-down of spin fluctuations as evidenced by the nuclear spin relaxation persist also up to higher doping levels ($x = 0.075$) [15, 23] and vanish only close to optimal doping $x \approx 0.1$. Figure 3 shows our results for the resistivity $\rho(T)$ [15], the Nernst effect $\nu(T)$ [21], and the spin-lattice relaxation rate $(T_1 T)^{-1}$ [23] of $\text{LaFeAsO}_{1-x}\text{F}_x$ at $x = 0.1$, which is close to optimal doping. In the resistivity, see Fig. 3a, the temperature dependence is completely different as compared to the underdoped levels. $\rho(T) \sim T^2$ is found for $T \lesssim 150$ K which smoothly develops into a linear tem-

perature dependence at higher temperature. Remarkably, the drop-like anomaly around 160 K and the low-temperature upturn have completely disappeared [15].

Despite a similar magnitude as for $x = 0.05$, the normal state Nernst coefficient as shown in Fig. 3b [21] displays a completely different normal-state behavior as compared to the underdoped compound. At $T \gtrsim 40$ K, $\nu(T)$ is just roughly linear with a weak positive slope. In particular, no anomaly at about 160 K is present.

Figure 3c shows $(T_1 T)^{-1}$ at $x = 0.1$. The low-temperature increase of $(T_1 T)^{-1}$ has vanished completely at this doping level and one observes a monotonic increase which approaches the same linear increase as in Fig. 2 [23, 39]. However, the deviation from such a linear increase, if any, is very small. This implies the essential absence of slow spin fluctuations at this doping level. Similarly to the underdoped case at $x = 0.05$, no anomaly is present in the thermal expansion [22] and x-ray diffraction [17].

4 Discussion Our main observation derives from the data in Fig. 2, *viz.*, the apparent occurrence of some form of orbital order in underdoped $\text{LaFeAsO}_{1-x}\text{F}_x$ at approximately the same temperature as in the parent compound while magnetic ordering is clearly absent. We recall the rationale which leads to this conclusion: the clear anomalies in the resistivity and the Nernst coefficient in Fig. 2 at about 160 K signal a decrease of the transport relaxation rate (and/or the effective electron mass) and, more generally, a static Fermi surface reconstruction. *A priori*, these can be connected with static orbital ordering, spin ordering, or a phonon-driven orthorhombic distortion. However, the latter two are not good candidates for the primary origin of the transport anomalies: firstly, all magnetic probes, *i.e.*, μSR , and Mößbauer spectroscopy prove the absence of static magnetic order at about 160 K [14, 20], whereas the here presented NMR data of the ^{75}As $(T_1 T)^{-1}$ reveal only progressively slowing down spin fluctuations with a relatively small magnitude. Secondly, a long-range orthorhombic distortion is absent as revealed by diffraction experiments and thermal expansion data [17, 22, 24, 47]. Note, that both magnetic and structural probes do not show any anomaly around 160 K. Thus, it is orbital-driven nematicity which remains as the most plausible possibility for the origin of the transport anomalies.

This inferred orbital nematicity evidently is very subtle, because the transport anomalies are much less pronounced as compared to the parent compound LaFeAsO where all degrees of freedom, *viz.*, orbitals, spins, and the lattice orthorhombic distortion participate in the ordering. This subtlety implies short-range orbital ordering or a slowly fluctuating variant, with length and time scales that are comparable to the electronic mean free path, respectively, the transport relaxation time.

If one considers the doping evolution of orbital ordering with the data at hand, the following scenario is apparent: in the parent compound, *i.e.*, in $\text{LaFeAsO}_{1-x}\text{F}_x$ at $x = 0$, long-range orbital order develops at $T_s = 160$ K, with an extended regime of fast fluctuations at higher temperatures.

As this orbital ordering is long range, the lattice can follow the orbital structure and the long-range orthorhombic distortion concomitantly develops. Long-range antiferromagnetic order then establishes at somewhat lower temperature at T_N , permitted by the broken C_4 symmetry. At underdoping ($x = 0.05$), short-range orbital order sets in at about 160 K, *i.e.*, at about the same temperature where long-range orbital order develops in the undoped case. It is thinkable, that the orbital ordering is only quasi-static, *i.e.*, the short-range orbital ordering is slowly fluctuating with a time scale that is much slower than the fast fluctuations in the parent compound at $T > 160$ K. In any case, the C_4 symmetry is only broken on a local scale. In this situation, the lattice can at most follow on the local scale, *i.e.*, a long-range orthorhombic distortion is impossible. However, the emergence of orthorhombically distorted nanoscale domains which are induced by the orbital structure but are invisible to the global lattice probes cannot be excluded. In an analogous way, the long-range magnetic order is prevented. Nevertheless, the short-range orbital order fosters the slowing-down of spin fluctuations. Note, that a fluctuation regime at $T > 160$ K of the short-range orbital ordering is not recognizable from the transport data, which implies that the overall amplitude of the ordering and of its fluctuations are relatively small. Finally, orbital ordering, even of short-range nature, is apparently completely absent at the near optimal doping level $x = 0.1$ as no anomalies occur at about 160 K.

The actual origin for the sketched doping evolution of nematicity in $\text{LaFeAsO}_{1-x}\text{F}_x$ remains unclear. One might speculate that the orbital ordering becomes increasingly unfavorable upon electronic band filling with doping. This scenario seems, however, rather unlikely because on a local scale the energetics of the orbital ordering obviously stay the same upon doping. This derives from the fact that the actual temperature where the ordering sets in is the same for $x = 0$ and $x = 0.05$. Alternatively, one might conclude from the evolution from long- to short-range ordering a spatially inhomogeneous electronic structure due to the doped charges. In such a scenario, orbital ordering could occur locally in sample areas which electronically resemble much the parent compound and would be quenched in the direct vicinity of a doped charge. The transition from long-range ordering to short-range ordering as a function of doping can then be understood in terms of a percolation threshold which separates the electronic phase diagram into a low-doping regime with long-range nematic order without superconductivity and an underdoped regime with quenched, *i.e.*, short-range (or fluctuating) nematic order which allows superconductivity. Remarkably, optimal superconductivity is reached when no signature of the short-range nematic order is present. If one stays in the sketched scenario, it is plausible that a total quenching of nematicity is reached at about 10% doped charges where distance between doped charges is already very small. We point out that this picture of the evolution of nematicity in $\text{LaFeAsO}_{1-x}\text{F}_x$ is supported by clear-cut experimental evidence from nuclear quadrupole resonance (NQR) measurements [48, 49] for two microscopically distinct charge

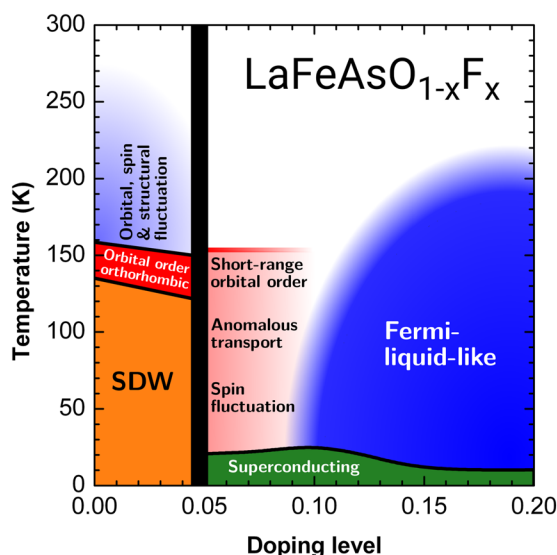


Figure 4 Schematic electronic phase diagram of $\text{LaFeAsO}_{1-x}\text{F}_x$ with three distinct doping regimes: (I) Small F-concentrations $x < 0.05$ including the parent compound, with long-range nematic order at $T \lesssim 160$ K. (II) the underdoped superconducting regime $0.05 \gtrsim x < 0.1$ with short-range orbital ordering (or a slowly fluctuating variant) at $T \lesssim 160$ K and progressively slowing down spin fluctuations. (III) Optimal and higher doping levels $x \gtrsim 0.1$ without nematicity and concomitantly highest T_c , as well as Fermi-liquid-like resistivity behavior. The shaded bar at $x \approx 0.05$ indicates the crossover region between regimes I and II.

environments at the As nucleus for $x < 0.1$, with one corresponding to that of the parent compound and the other to that of higher-doped compounds. It has also been suggested based on NQR data that percolation would indeed control the occurrence of orthorhombicity and static magnetism [49].

We summarize our qualitative findings in a schematic phase diagram of $\text{LaFeAsO}_{1-x}\text{F}_x$ which is shown in Fig. 4 which expands our earlier results [15]. The parent compound and non-superconducting compounds at low doping levels exhibit a long-range ordered orbital-driven nematic state at low temperature. At higher temperature, i.e., prior to the transition to the nematic state, orbital, structural, and magnetic fluctuations govern the physics of the material. This situation persists at small doping levels where the nematic order is somewhat destabilized due to the doped charges but remains long range. Above a critical doping value $x < 0.05$, the long-range nematic order state is completely suppressed, presumably due to an exceeded percolation limit which quenches the long-range order, and superconductivity occurs. The clear anomalies in the resistivity and the Nernst coefficient at about 160 K and the absence of a structural distortion and of magnetic order provide the evidence for short-range orbital order (which might also be slowly fluctuating) at the same temperature as the long-range order in the parent compound. Naturally, due to the inevitable coupling of orbital and lattice degrees of freedom, one can expect nanoscale orthorhombically distorted lattice domains, which are induced by the orbital ordering. At the same time, this phase possesses signatures

of incipient magnetic order, evidenced by slow spin fluctuations. This short-range or fluctuating nematic state fades out at about optimal doping where the resistivity approaches a less unconventional normal state with a $\sim T^2$ dependence at low temperature, signaling Fermi-liquid-like behavior, and a linear temperature dependence at higher temperatures.

It is interesting to note that the orbital driven nematic order in $\text{LaFeAsO}_{1-x}\text{F}_x$ bears some resemblance with the recently reported nematicity in FeSe [12, 35], in the sense that the long-range orbital ordering, accompanied by an orthorhombic distortion, occurs without magnetic order, with the difference that in LaFeAsO long-range magnetic order sets in just a few Kelvin below whereas it remains absent in FeSe. It is also worth to mention that the doping evolution of nematic fluctuations in $\text{LaFeAsO}_{1-x}\text{F}_x$ is quite different from that in Co- and K-doped BaFe_2As_2 . In $\text{LaFeAsO}_{1-x}\text{F}_x$ short-range orbital order (or fluctuations of it) seem to disappear toward optimal doping whereas in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ and $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ nematic fluctuations have been reported to be present over the entire superconducting dome [11] with a tight connection between magnetic and lattice fluctuations [10]. From these findings for the BaFe_2As_2 -based compounds, it has been conjectured the nematic fluctuations could be involved in the superconducting pairing. Our finding of optimal superconductivity with concomitantly disappearing nematic fluctuations renders this possibility less likely for $\text{LaFeAsO}_{1-x}\text{F}_x$, and suggest that the nematic phase actually competes with superconductivity.

5 Summary We have investigated the iron-oxypnictide superconductor $\text{LaFeAsO}_{1-x}\text{F}_x$ in the normal state with a focus on signatures of nematic phases in resistivity, Nernst effect, and ^{75}As NMR data. For undoped LaFeAsO , very pronounced anomalies in all these quantities signal the onset of long-range nematic order. At the underdoped level $x = 0.05$ much weaker but still distinct anomalies at about the same temperature where undoped LaFeAsO develops long-range nematic ordering remain present in the transport coefficients. Despite these signatures of a nematic state, a detectable orthorhombic lattice distortion and magnetic order are absent, and the ^{75}As -NMR spin-lattice relaxation rate $(T_1 T)^{-1}$ reveals a progressive slowing down of spin fluctuations. We conclude from these observations that short-range or slowly fluctuating orbital ordering sets in near 160 K, which is the onset temperature of long-range nematic order in the parent compound LaFeAsO . All anomalies in the transport coefficients and also the indications of slow spin fluctuations are absent at $x = 0.1$, i.e., near optimal doping. The results therefore suggest that the nematic phase competes with superconductivity as it is gradually destroyed by doping and vanishes upon reaching a maximum critical temperature at optimal doping.

Acknowledgements This work has been supported by the Deutsche Forschungsgemeinschaft (DFG) through the Priority Programme SPP1458 (Grants BE1749/12 and GR3330/2).

References

- [1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *J. Am. Chem. Soc.* **130**, 3296–3297 (2008).
- [2] D. C. Johnston, *Adv. Phys.* **59**, 803–1061 (2010).
- [3] I. I. Mazin, D. J. Singh, M. D. Johannes, and M. H. Du, *Phys. Rev. Lett.* **101**, 057003 (2008).
- [4] C. Xu, M. Müller, and S. Sachdev, *Phys. Rev. B* **78**, 020501 (2008).
- [5] C. Fang, H. Yao, W. F. Tsai, J. Hu, and S. A. Kivelson, *Phys. Rev. B* **77**, 224509 (2008).
- [6] R. M. Fernandes, A. V. Chubukov, and J. Schmalian, *Nature Phys.* **10**, 97–104 (2014).
- [7] J. H. Chu, J. G. Analytis, K. De Greve, P. L. McMahon, Z. Islam, Y. Yamamoto, and I. R. Fisher, *Science* **329**, 824–826 (2010).
- [8] J. H. Chu, H. H. Kuo, J. G. Analytis, and I. R. Fisher, *Science* **337**, 710–712 (2012).
- [9] M. Yi, D. Lu, J. H. Chu, J. G. Analytis, A. P. Sorini, A. F. Kemper, B. Moritz, S. K. Mo, R. G. Moore, M. Hashimoto, W. S. Lee, Z. Hussain, T. P. Devereaux, I. R. Fisher, and Z. X. Shen, *Proc. Natl. Acad. Sci. USA* **108**, 6878–6883 (2011).
- [10] R. M. Fernandes, A. E. Böhmer, C. Meingast, and J. Schmalian, *Phys. Rev. Lett.* **111**, 137001 (2013).
- [11] A. E. Böhmer, P. Burger, F. Hardy, T. Wolf, P. Schweiss, R. Fromknecht, M. Reinecker, W. Schranz, and C. Meingast, *Phys. Rev. Lett.* **112**, 047001 (2014).
- [12] S. H. Baek, D. V. Efremov, J. M. Ok, J. S. Kim, J. van den Brink, and B. Büchner, *Nature Mater.* **14**, 210–214 (2015).
- [13] A. S. Sefat, A. Huq, M. A. McGuire, R. Jin, B. C. Sales, D. Mandrus, L. M. D. Cranswick, P. W. Stephens, and K. H. Stone, *Phys. Rev. B* **78**, 104505 (2008).
- [14] H. Luetkens, H. H. Klauss, M. Kraken, F. J. Litterst, T. Dellmann, R. Klingeler, C. Hess, R. Khasanov, A. Amato, C. Baines, M. Kosmala, O. J. Schumann, M. Braden, J. Hamann-Borrero, N. Leps, A. Kondrat, G. Behr, J. Werner, and B. Büchner, *Nature Mater.* **8**, 305–309 (2009).
- [15] C. Hess, A. Kondrat, A. Narduzzo, J. E. Hamann-Borrero, R. Klingeler, J. Werner, G. Behr, and B. Büchner, *EPL (Europhys. Lett.)* **87**, 17005 (2009).
- [16] A. J. Drew, C. Niedermayer, P. J. Baker, F. L. Pratt, S. J. Blundell, T. Lancaster, R. H. Liu, G. Wu, X. H. Chen, I. Watanabe, V. K. Malik, A. Dubroka, M. Rossle, K. W. Kim, C. Baines, and C. Bernhard, *Nature Mater.* **8**, 310–314 (2009).
- [17] A. Kondrat, J. E. Hamann-Borrero, N. Leps, M. Kosmala, O. Schumann, A. Köhler, J. Werner, G. Behr, M. Braden, R. Klingeler, B. Büchner, and C. Hess, *Eur. Phys. J. B* **70**, 461–468 (2009).
- [18] C. de la Cruz, Q. Huang, J. W. Lynn, J. Li, W. R. II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang, and P. Dai, *Nature* **453**, 899–902 (2008).
- [19] H. H. Klauss, H. Luetkens, R. Klingeler, C. Hess, F. J. Litterst, M. Kraken, M. M. Korshunov, I. Eremin, S. L. Drechsler, R. Khasanov, A. Amato, J. Hamann-Borrero, N. Leps, A. Kondrat, G. Behr, J. Werner, and B. Büchner, *Phys. Rev. Lett.* **101**, 077005 (2008).
- [20] R. Klingeler, N. Leps, I. Hellmann, A. Popa, U. Stockert, C. Hess, V. Kataev, H. J. Grafe, F. Hammerath, G. Lang, S. Wurmehl, G. Behr, L. Harnagea, S. Singh, and B. Büchner, *Phys. Rev. B* **81**, 024506 (2010).
- [21] A. Kondrat, G. Behr, B. Büchner, and C. Hess, *Phys. Rev. B* **83**, 092507 (2011).
- [22] L. Wang, U. Köhler, N. Leps, A. Kondrat, M. Nale, A. Gasparini, A. de Visser, G. Behr, C. Hess, R. Klingeler, and B. Büchner, *Phys. Rev. B* **80**, 094512 (2009).
- [23] F. Hammerath, U. Gräfe, T. Kühne, H. Kühne, P. L. Kuhns, A. P. Reyes, G. Lang, S. Wurmehl, B. Büchner, P. Carretta, and H. J. Grafe, *Phys. Rev. B* **88**, 104503 (2013).
- [24] N. Qureshi, Y. Drees, J. Werner, S. Wurmehl, C. Hess, R. Klingeler, B. Büchner, M. T. Fernández-Díaz, and M. Braden, *Phys. Rev. B* **82**, 184521 (2010).
- [25] M. A. McGuire, A. D. Christianson, A. S. Sefat, B. C. Sales, M. D. Lumsden, R. Jin, E. A. Payzant, D. Mandrus, Y. Luan, V. Keppens, V. Varadarajan, J. W. Brill, R. P. Hermann, M. T. Sougrati, F. Grandjean, and G. J. Long, *Phys. Rev. B* **78**, 094517 (2008).
- [26] F. Steckel, S. Rodan, R. Hermann, C. G. F. Blum, S. Wurmehl, B. Büchner, C. Hess, *Phys. Rev. B* **90**, 134411 (2014).
- [27] M. S. Kim, M. C. Bennett, and M. C. Aronson, *Phys. Rev. B* **77**, 144425 (2008).
- [28] C. Hess, E. Ahmed, U. Ammerahl, A. Revcolevschi, and B. Büchner, *Eur. Phys. J. Spec. Top.* **188**, 103–112 (2010).
- [29] R. Bel, K. Behnia, and H. Berger, *Phys. Rev. Lett.* **91**, 066602 (2003).
- [30] O. Cyr-Choinière, R. Daou, F. Laliberté, D. LeBoeuf, N. Doiron-Leyraud, J. Chang, J. Q. Yan, J. G. Cheng, J. S. Zhou, J. B. Goodenough, S. Pyon, T. Takayama, H. Takagi, Y. Tanaka, and L. Taillefer, *Nature* **458**, 743–745 (2009).
- [31] R. Daou, J. Chang, D. LeBoeuf, O. Cyr-Choinière, F. Laliberté, N. Doiron-Leyraud, B. J. Ramshaw, R. Liang, D. A. Bonn, W. N. Hardy, and L. Taillefer, *Nature* **463**, 519–522 (2010).
- [32] A. Hackl and M. Vojta, *Phys. Rev. B* **80**, 220514 (2009).
- [33] A. Hackl, M. Vojta, and S. Sachdev, *Phys. Rev. B* **81**, 045102 (2010).
- [34] The samples are fixed by a small force in the capacitance dilatometer resulting in a pressure of the order of ≈ 100 kPa applied along the direction of the measured length changes.
- [35] A. E. Böhmer, T. Arai, F. Hardy, T. Hattori, T. Iye, T. Wolf, H. v. Löhneysen, K. Ishida, and C. Meingast, *Phys. Rev. Lett.* **114**, 027001 (2015).
- [36] Y. Nakai, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, *J. Phys. Soc. Jpn.* **77**, 073701 (2008).
- [37] T. Moriya, *J. Phys. Soc. Jpn.* **18**, 516–520 (1963).
- [38] H. J. Grafe, G. Lang, F. Hammerath, D. Paar, K. Manthey, K. Koch, H. Rosner, N. J. Curro, G. Behr, J. Werner, N. Leps, R. Klingeler, and B. Büchner, *New J. Phys.* **11**, 035002 (2008).
- [39] H. J. Grafe, D. Paar, G. Lang, N. J. Curro, G. Behr, J. Werner, J. Hamann-Borrero, C. Hess, N. Leps, R. Klingeler, and B. Büchner, *Phys. Rev. Lett.* **101**, 047003 (2008).
- [40] M. M. Korshunov and I. Eremin, *EPL (Europhys. Lett.)* **83**, 67003 (2008).
- [41] M. M. Korshunov, I. Eremin, D. V. Efremov, D. L. Maslov, and A. V. Chubukov, *Phys. Rev. Lett.* **102**, 236403 (2009).
- [42] G. M. Zhang, Y. H. Su, Z. Y. Lu, Z. Y. Weng, D. H. Lee, and T. Xiang, *EPL (Europhys. Lett.)* **86**, 37006 (2009).
- [43] M. Berciu, I. Elfimov, and G. A. Sawatzky, *Phys. Rev. B* **79**, 214507 (2009).
- [44] G. A. Sawatzky, I. S. Elfimov, J. van den Brink, and J. Zaanen, *EPL (Europhys. Lett.)* **86**, 17006 (2009).
- [45] S. L. Skornyakov, A. A. Katanin, and V. I. Anisimov, *Phys. Rev. Lett.* **106**, 047007 (2011).

- [46] J. Chaloupka and G. Khaliullin, Phys. Rev. Lett. **110**, 207205 (2013).
- [47] H. Luetkens, H. H. Klauss, R. Khasanov, A. Amato, R. Klingeler, I. Hellmann, N. Leps, A. Kondrat, C. Hess, A. Köhler, G. Behr, J. Werner, and B. Büchner, Phys. Rev. Lett. **101**, 097009 (2008).
- [48] G. Lang, H. J. Grafe, D. Paar, F. Hammerath, K. Manthey, G. Behr, J. Werner, and B. Büchner, Phys. Rev. Lett. **104**, 097001 (2010).
- [49] G. Lang, L. Veyrat, U. Graefe, F. Hammerath, D. Paar, G. Behr, S. Wurmehl, and H. J. Grafe, arXiv:1508.06532 (2015).