Absence of nematic instability in LiFeAs

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(Received 7 February 2022; accepted 29 June 2022; published 8 August 2022)

The relationship among unconventional superconductivity, antiferromagnetism, and nematic order in iron-based superconductors (FeSCs) is still highly debated. In many FeSCs superconductivity is in proximity of a nematically and magnetically ordered state. LiFeAs is an exceptional stoichiometric FeSC becoming superconducting below 18 K without undergoing a structural or magnetic transition. However, some recent experimental studies suggested the existence of finite nematic fluctuations and even a nematic superconducting state. In this paper, we employ elastoresistance as a measure of nematic fluctuations in pristine LiFeAs and compare the findings with the elastoresistance of LiFeAs at low Co and V doping levels as well with that of magnetically and nematically ordering NaFeAs. We find LiFeAs and cobalt-doped LiFeAs far away from a nematic instability.

DOI: 10.1103/PhysRevB.106.054508

I. INTRODUCTION

Electronic nematic order is a state that spontaneously breaks the rotational symmetry whereas preserving translational symmetry. It has become a subject of increasing attention in the context of the rich phase interplay in iron-based superconductors (FeSCs) [1–3]. In most FeSCs, nematicity occurs in close vicinity of an antiferromagnetically ordered state, which was repeatedly proposed to be intimately connected to the evolution of the unconventional superconductivity [4–6]. In order to disentangle the relation of superconductivity, magnetism, and nematicity, it is important to investigate different FeSCs, especially those which exhibit just one or two of these phases.

LiFeAs, which crystalizes in the space-group $P4/nmm$ [7], is one of these unusual representatives of the FeSC-family. The role of nematicity in LiFeAs is to date still strongly debated. It is superconducting in the undoped parent compound with a superconducting transition temperature between 16 and 18 K [8–11]. However, several experimental probes including superconducting quantum interference device magnetometry [8,12] and muon-spin rotation [13,14] found no evidence of a magnetic transition or long-range magnetic order. However, it has been reported that LiFeAs could be, in principle, susceptible to nematicity [15,16] or even that it undergoes an unusual symmetry-breaking transition [17,18]. In particular, in Ref. [15], the electric-field gradient in nuclear magnetic resonance (NMR) measurements was used as an indirect probe of local anisotropies in the crystal structure and a finite albeit very small η in LiFeAs has, therefore, been interpreted as the signature of nematic fluctuations. The presence of electronic-nematic fluctuations has also been suggested from an unusual enhancement of the quasiparticle interference (QPI) amplitude in scanning tunnel microscope (STM) measurements [16], and even the stabilization of static nematicity in the superconducting state and of smectic electronic order under uniaxial strain have been reported in angle-resolved photoemission spectroscopy (ARPES) [17] and STM [18] measurements.

The, thus, still unclear role of nematicity in LiFeAs calls for further investigation of nematic fluctuations especially with different probes. One very successful experimental approach to investigate nematic properties in FeSCs is measuring the strain susceptibility of the resistivity $\eta = \frac{d}{d\epsilon}\frac{\Delta\rho}{\rho}$. With this elastoresistance technique, Chu *et al.* [19] were able to establish the temperature-dependent behavior of η as a proof of the electronic origin of nematicity in FeSCs and a measure of the strength of nematic fluctuations, which has since then been confirmed and used by other research groups [20–24].

In this paper, we investigate nematic fluctuations in LiFeAs using elastoresistance measurements on undoped, electron-doped, and hole-doped LiFeAs. In apparent contradiction to the aforementioned reports [15–18], our results clearly indicate that pure LiFeAs is far away from a nematic instability. We compare our results with elastoresistance data on NaFeAs, which is significantly different from LiFeAs.

II. EXPERIMENT

NaFeAs single crystals were prepared according to Ref. [25]. Single crystals of pristine as well as doped LiFeAs were grown by using the self-flux technique as described

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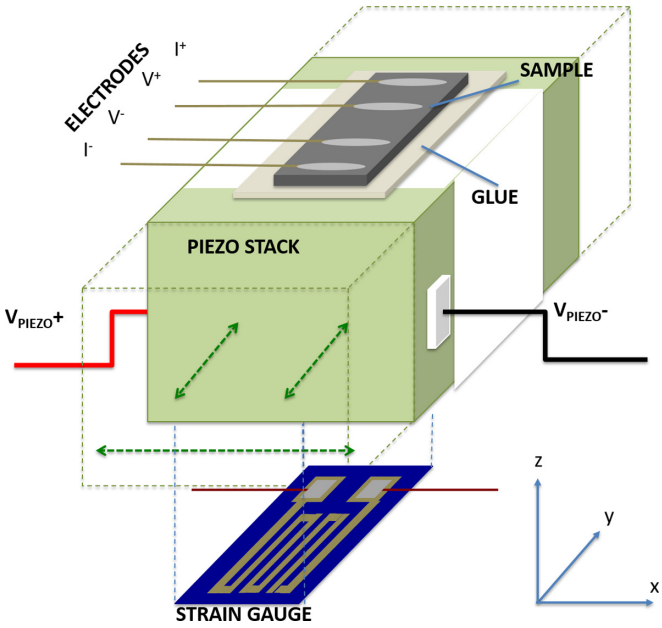

FIG. 1. Experimental setup: Silver wires glued to the sample with the silver paint function as electrodes. Green dashed lines at the piezostack illustrate the expansion under application of a positive voltage V_p. Strain ϵ is measured using a strain gauge on the backside, figure adapted from Ref. [23].

in Refs. [9,26]. Prereacted Li_3As, FeAs, and $FeAs_2$ were used. The mixture was placed in a graphite crucible, which was placed in a Nb container which was later sealed in a quartz ampule to avoid oxidation. This quartz ampule was then placed vertically in a furnace and heated to 1100 °C. After dwelling for 7 h, the furnace was cooled down at a rate of 4.5 K/h to 600 °C and later cooled with 100 °C/h to room temperature. As grown crystals [exemplary image of $Li(Fe_{0.972}V_{0.028})As$ in the inset of Fig. 2(d)] are highly sensitive to air and moisture. Single crystals were characterized by using SEM/x-ray analysis and powder x-ray diffraction, which confirm the stoichiometry of the LiFeAs. All sample preparation steps have been performed in an argon-filled glove box with O_2 and H_2O content less than 0.3 ppm. The glove box is equipped with a second 2-m-long chamber that enables the introduction of a whole measurement probe into the Ar atmosphere. Few selected pieces of as grown single crystals were cut mechanically into rectangular shapes with lengths <4 mm and cleaved to thicknesses of the order of 50 μm, to ensure a homogeneous transmission of strain through the whole thickness (compare Refs. [19,23]). Ag wires were glued on top of a sample in order to perform four-terminal resistance measurements. The samples were glued (using Devcon 14250 5 minute epoxy) along the [110]-crystal axis on top of a piezoelectric actuator that allowed precise strain control depending on an applied voltage V_p. A positive (negative) V_p leads to an expansion (contraction) in y direction (Fig. 1). The strain ϵ was measured using a strain gauge glued onto the backside of the piezo. An exemplary photograph of the setup can be found in Ref. [27]. After making contacts to the sample, it was mounted directly onto the probe, which then was closed inside the Ar box, discharged through the

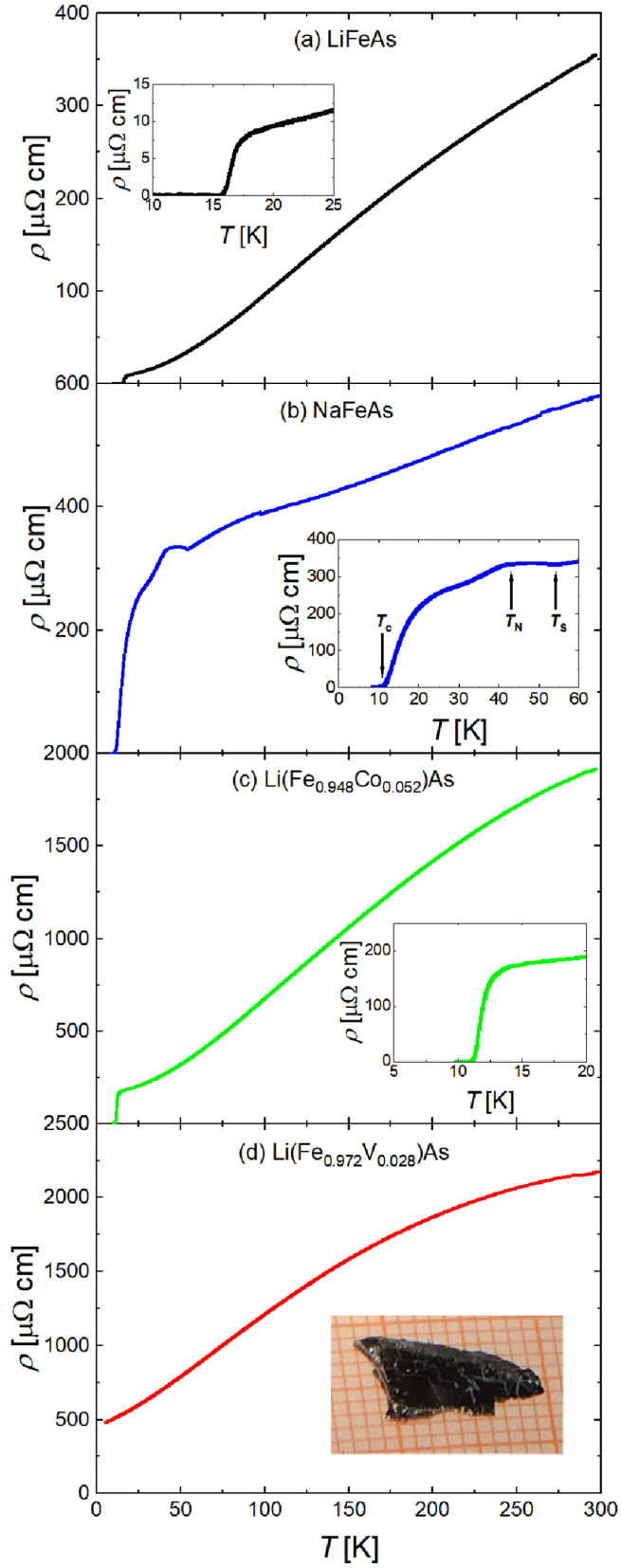

FIG. 2. Temperature dependence of the resistivities $\rho(T)$ of (a) LiFeAs, (b) NaFeAs, (c) $Li(Fe_{0.948}Co_{0.052})As$, (d) $Li(Fe_{0.972}V_{0.028})As$; the insets of (a)–(c): Superconducting transitions, the inset of (d): Photograph of a $Li(Fe_{0.972}V_{0.028})As$ crystal.

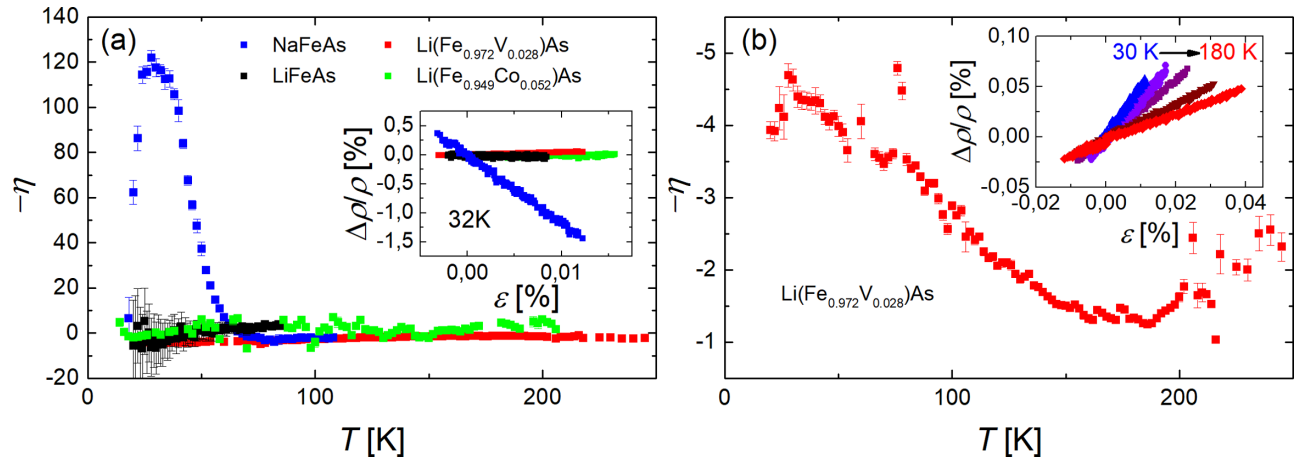

FIG. 3. (a) Elastoresistance η of NaFeAs and Li($Fe_{1-x}TM_x$)As (TM = V, Co). NaFeAs shows a strong divergence, indicating the increasing amount of nematic fluctuations towards the structural transition temperature. The electronic response to strain is significantly smaller, almost nonexistent in Li($Fe_{1-x}TM_x$)As (TM = V, Co). Exemplary $\frac{\Delta\rho}{\rho}$-ϵ relations of the three materials at 32 K in the inset. (b) The amplitude of the elastoresistance of Li($Fe_{0.972}V_{0.028}$)As shows a small increase towards lower temperatures. The error bars of each data point are derived from the statistical error of a linear fit through the $\frac{\Delta\rho}{\rho}$ vs ϵ data. In the tetragonal phase, a negative sign of the nematic susceptibility indicates that the resistance is smaller along the longer crystal axis a than along the shorter b. $\frac{\Delta\rho}{\rho}$-ϵ relations at selected temperatures in the inset.

air lock and immediately connected to a high-vacuum pump, which creates a vacuum of less than 1×10^{-6} mbars to 1×10^{-7} mbars. The evacuated probe was inserted into liquid helium and cooled down. Due to cryopumping, the sample stayed in cryogenic vacuum conditions until it got warmed up again.

III. RESULTS

In Figures 2(a) and 2(b), we present the temperature-dependent resistivities $\rho(T)$ of LiFeAs and NaFeAs, respectively.

The resistivity trend and absolute value of both compounds correspond well to earlier reports [28,29]. The structural and magnetic phase transitions of NaFeAs at $T_S \sim 54$ K and $T_N \sim 43$ K are visible as kinks in $\rho(T)$ [inset in Fig. 2(b)], whereas no kinks or anomalies are observed in the resistivity of LiFeAs as it approaches the sharp superconducting transition at $T_c \sim 16$ K [inset in Fig. 2(a)]. This comparably high transition temperature and the small absolute value of ρ indicate a high quality of the sample.

In LiFeAs, 5.2% Co doping increases the resistivity and lowers the superconducting transition temperature to $T_c \sim 11$ K [Fig. 2(c)]. Apparently, as can be inferred from Fig. 2(d), the impact of V doping on the resistivity is much stronger than that of Co doping. Already at 2.8% V doping the resistivity is higher than that of the Co-doped sample and no sign of a superconducting transition down to 5 K is observed. The ρ-T curves of both doped LiFeAs compounds show no indication of a magnetic or nematic phase transition, consistent with previous reports [30–32].

In Fig. 3(a), the temperature dependence of the elastoresistance η of all four FeSCs investigated in this paper is shown. Remarkably, only the elastoresistance amplitude of NaFeAs shows a diverging behavior towards low temperatures (consistent with Ref. [33]) typical for FeSCs with a nematic phase transition, indicating the increase in nematic fluctuations towards T_S. In contrast, in LiFeAs, at no temperature a clear electronic response $\frac{\Delta\rho}{\rho}$ to strain is observed. This is direct evidence for the absence of nematic fluctuations since local structural distortions connected to local nematic order evidentially change the strain response of the resistivity. The lack of a measurable increase of the elastoresistance amplitude towards low temperatures indicates that the material is far away from a nematic instability.

Having established that pure LiFeAs is far away from a nematic instability opens up the question, whether it is possible to drive the material towards nematicity by tuning external parameters. Here, we investigate the effect of electron and hole doping on the nematic properties of LiFeAs by measuring elastoresistance of Li($Fe_{0.948}Co_{0.052}$)As and Li($Fe_{0.972}V_{0.028}$)As, respectively. In 5.2% Co-doped LiFeAs, similar to the undoped parent compound, no electronic response to strain could be measured which leads to elastoresistance values scattered around zero. The absence of any sign of local structural distortion/nematic fluctuation in LiFeAs does not change upon 5.2% Co doping.

On the other hand, we report a small divergence of the nematic susceptibility towards low temperatures in Li($Fe_{0.972}V_{0.028}$)As [Fig. 3(b)]. Even though the absolute amplitude is low, indicating a small but finite amount of nematic fluctuations, a trend is clearly visible. Note, that the low-temperature absolute value of ρ of Li($Fe_{0.972}V_{0.028}$)As is an order of magnitude higher compared to LiFeAs, thus, enabling a very precise measurement of η—as also noticable in the comparatively small size of the error bars.

IV. DISCUSSION

The absence of nematicity in elastoresistance measurements on LiFeAs might seem at odds with the aforementioned NMR [15], ARPES [17], and STM [16,18] reports. However, a closer inspection reveals a good consistency with these data. More specifically, in Ref. [15], the electric-field gradient of

LiFeAs is reported to be more than one order of magnitude smaller than that of NaFeAs (absolute value smaller than 0.005 even at low temperatures). Such a small-amplitude sign of electronic anisotropy is, in principle, consistent with the small increase in the elastoresistance signal in LiFeAs in Fig. 3. However, also given the significant range of error compared to the small absolute values, the signal cannot unambigously be assigned to nematicity. Furthermore, the nematic order reported from ARPES in Ref. [17] is interpreted as a result of the superconductivity and, therefore, should disappear at the elastoresistance-relevant temperatures above T_c (within the limits of superconducting fluctuations). Finally, the QPI measurements and analysis in Ref. [16] suggest a coupling between electronic and supposedly nematic lattice modes at energies in the range of 10 meV. The energy scale of our elastoresistance measurements is expected to be smaller since we measure in quasistatic conditions in the zero-strain limit. It seems reasonable to expect that under these conditions the nematic modes are energetically too far away to affect the transport data.

Therefore, in consistency with these previous reports on nematic properties in LiFeAs, we conclude that the missing strain response in the elastoresistance measurements presented above strongly suggests that pure, undoped LiFeAs is far away from a nematic instability.

The results on Co-doped LiFeAs seem, in principle, consistent with the observation on canonical FeSCs with nematic ordering parent compounds (i.e., $BaFe_2As_2$ and NaFeAs) that the amplitude of the nematic fluctuations declines upon increasing Co doping to the overdoped side (as seen in elastotransport [21] and NMR [15]). In such a scenario, LiFeAs would be located deep in the electron-doped side of the electronic phase diagram. In this case, hole doping should, in principle, drive the system closer to the nematic phase with a higher amplitude of nematic fluctuations. Indeed, this is what is observed upon V doping, where vanadium could act as an effective hole dopant. In fact, V doping is known to promote the emergence of strong antiferromagnetic spin fluctuations, which seem supportive of this scenario [31,34,35]. However, at the same time the V doping apparently suppresses the superconductivity, which indicates that this doping scheme does not simply drive the system towards lower doping levels in the phase diagram of the canonical electron-doped FeSCs.

V. SUMMARY

To summarize, we employed elastoresistance as direct probe of the nematic properties of undoped, electron-doped, and hole-doped LiFeAs. We showed, that there is no sign of nematic fluctuations detectable in LiFeAs. Furthermore, Co doping does not induce nematic fluctuations, indicating that LiFeAs is on the electron-doped side far away from a nematic instability. Finally, V doping of LiFeAs might induce nematic fluctuations, whereas at the same time suppressing superconductivity.

ACKNOWLEDGMENTS

This project has been supported by the Deutsche Forschungsgemeinschaft through the Research Projects CA 1931/1-1 (F.C.). Furthermore, this project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 Research and Innovation Programme (Grant Agreement No. 647276-MARS-ERC-2014-CoG) (X.H. and C.H.) and Grant No. AS 523/4-1 (S.A.). I.M., A.V., S.A., and B.B. thank DFG and RSF for financial support in the frame of the joint DFG-RSF Project "Weyl and Dirac semimetals and beyond—prediction, synthesis, and characterization of new semimetals."

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