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Research articles

Nematicity and structure in $LaFe_{1-x}Co_xAsO$

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ABSTRACT

We present thermal expansion studies on single-crystalline $LaFe_{1-x}Co_xAsO$ with doping levels of $x = 0$, 0.026, and 0.034, and on $BaFe_2As_2$. Anomalies in the thermal expansion coefficient α enable reading off the structural and magnetic transition temperatures T_S and T_N. The evolution of the orthorhombic distortion at T_S is illustrated by length changes of the *a*- and *b*-axes and the experimentally determined orthorhombicity δ. The results are compared to $BaFe_2As_2$. Finally, the electronic phase diagram is discussed.

1. Introduction

Fe-based unconventional superconductors have raised enormous attention since their discovery in $LaFeAsO_{1-x}F_x$ in 2008 [1]. In addition to this '1111' structure type, there is a variety of related structures such as '122', '111', or '11', each of which featuring layers of tetrahedrally coordinated Fe [2]. Many materials of this class, among them $LaFe_{1-x}Co_xAsO$ studied here, show antiferromagnetic (AFM) order in close proximity to the emerging superconducting phase. In addition, there is a symmetry reduction from the C_4 (tetragonal) rotational symmetry at high temperatures to C_2 (orthorhombic) at low temperatures, which is referred to as an Ising-nematic phase. The structural transition may be accompanied by the magnetic one (e.g., in $BaFe_2As_2$), or separated from it (e.g., in $LaFeAsO_{1-x}F_x$). Both in 1111 and 122 systems, it was shown that electronic doping by substitution of Fe for Co may lead to a suppression of magnetism and the appearance of superconductivity [3–6]. In $BaFe_2As_2$, the concomitant magneto-structural phase transition of the parent compound splits into two well-defined transitions upon Co substitution. The presence of an orthorhombic but paramagnetic phase in which electronic nematicity is observed [7] has put the simple picture of the orthorhombic distortion being a mere consequence of AFM into question and has sparked great interest in the nematic phase of the iron-based materials and its microscopic origin [8,9]. In $LaFeAsO_{1-x}F_x$, the tetragonal-to-orthorhombic transition appears well above T_N , giving rise to a nematic phase [10–12]. Specifically, in the parent compound LaFeAsO the structural transition appears at $T_S \approx 155$ K [17,10] and long-range AFM order evolves at $T_N \approx 130$ K [18,10,19,20]. While previous reports however have been based on poly-crystals, single crystals became available only recently [13]. Here, we present the first thermal expansion data on detwinned LaFeAsO single crystals with $x = 0$, 0.026, and 0.034 which enable studying the anomalies of the thermal expansion coefficient at T_N and T_S , the evolution of orthorhombicity, and their doping dependencies. The findings are compared to $BaFe_2As_2$ where the magnetic and structural transitions appear concomitantly in a discontinuous manner.

2. Experiment

$LaFe_{1-x}Co_xAsO$ and $BaFe_2As_2$ single crystals have been grown as described in Refs. [13,14]. For the former, the Co contents have been confirmed by EDX. For the thermal expansion measurements, a three-terminal capacitance dilatometer was utilized [15], which allows an accurate study of crystal length changes. As it was demonstrated previously [16,23,24], the shorter *b*-axis in the low-temperature C_2 phase can be obtained directly by measuring the expansion of the crystal along the $[110]_T$ direction of the original tetragonal cell (see Fig. 3a) because in this configuration the small built-in repulsion force from the dilatometer springs detwins the crystal. The larger *a*-axis, on the other hand, is obtained by combining a 'twinned' measurement (along $[110]_T$) with the 'detwinned' data [16].

3. Results and discussion

Fig. 1 presents the thermal expansion coefficients,

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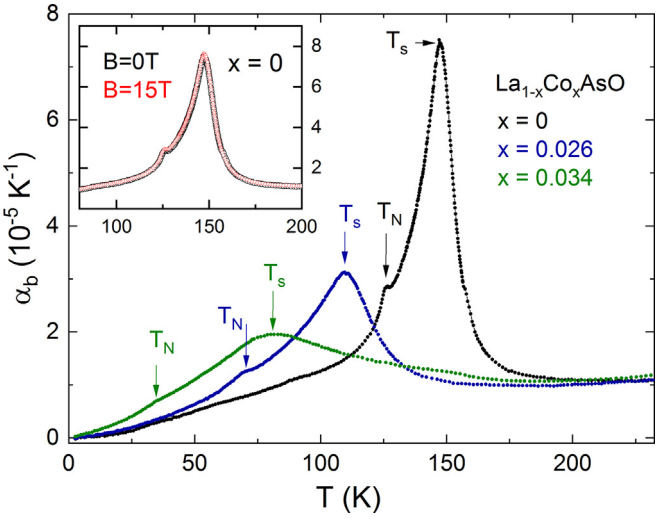

Fig. 1. In-plane thermal expansion coefficients α_b vs. temperature for $x = 0$, 0.026, and 0.034. The insert shows α_b of LaFeAsO at $B = 0$ T and 15 T.

$\alpha_b(T) = 1/L_b \cdot dL_b(T)/dT$ of $LaFe_{1-x}Co_xAsO$. T_S is associated to a pronounced anomaly in α for all doping levels. For $x = 0$, the thermal expansion anomaly maxima imply $T_S = 147$ K and $T_N = 126$ K which is slightly smaller than values from thermal expansion studies on polycrystals where much broader anomalies with maxima at 157 and 137 K have been found [11]. The magnetic transition at T_N is clearly visible at all doping levels under study but appears very weak as compared to the structural one. Applying a magnetic field of 15 T does not yield any significant changes of the anomalies in LaFeAsO as shown in the inset of Fig. 1.

In Fig. 2a, the relative length changes, $\Delta L/L$, along the a- and b-axes of LaFeAsO are presented. In addition, the figure displays respective data on $BaFe_2As_2$ where structural distortion and magnetic order concomitantly appear in a first order transition [16]. The evolution of the orthorhombic distortion, $\delta = (a - b)/(a + b)$, is presented in Fig. 2b. In clear constrast to $BaFe_2As_2$, orthorhombicity in LaFeAsO evolves smoothly and there is a much larger regime of $\gtrsim 50$ K where precursing orthorhombicity is observed. Fig. 2 also displays the T-dependent

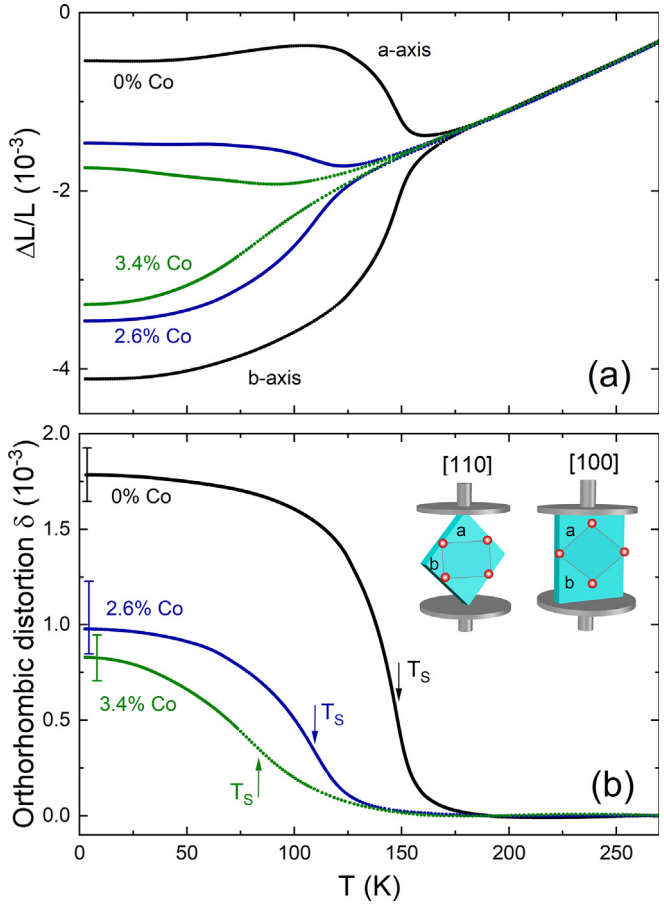

Fig. 3. Relative length changes $\Delta L/L$ vs. temperature of the orthorhombic lattice parameters a and b of $LaFe_{1-x}Co_xAsO$ with $x = 0$, 0.026, and 0.034 as well as (b) of the orthorhombic distortion δ inferred from the data in (a). The sketch illustrates the measurement setup (see the text) [22].

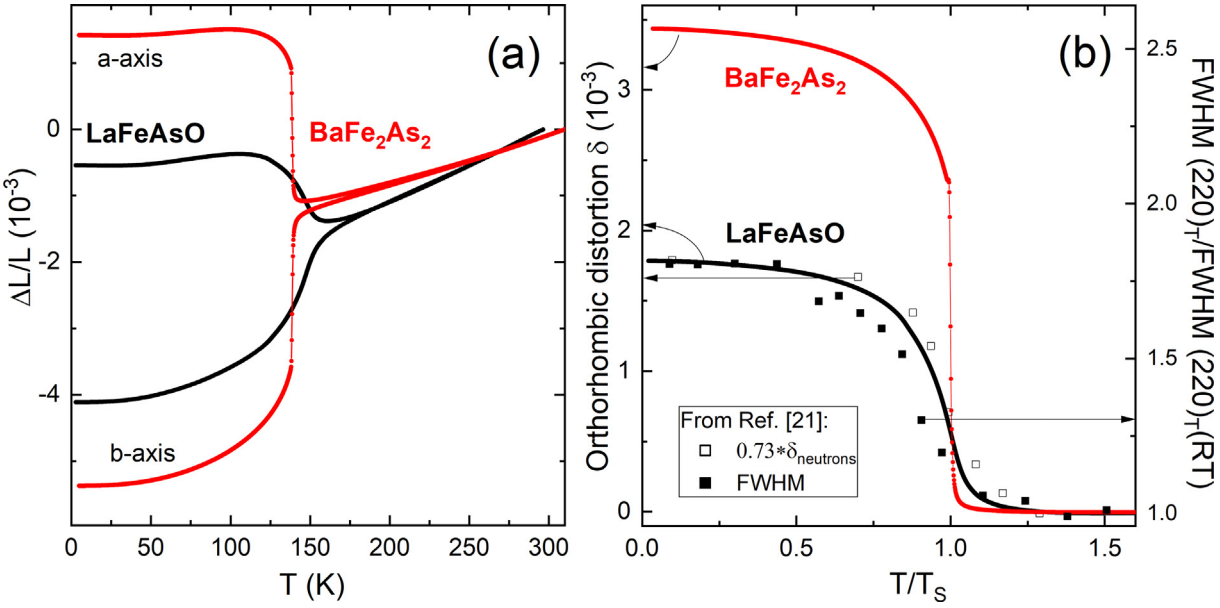

Fig. 2. Relative length changes $\Delta L/L$ vs. temperature of the orthorhombic lattice parameters a and b of LaFeAsO and $BaFe_2As_2$, and (b) orthorhombic distortion $\delta = (a - b)/(a + b)$ inferred from the data in (a). Filled and open squares show the normalised orthorhombicity from neutron diffraction and the FWHM of the splitting of the $(220)_T$-reflection obtained on a polycrstalline sample with $T_S = 152$ K [21].

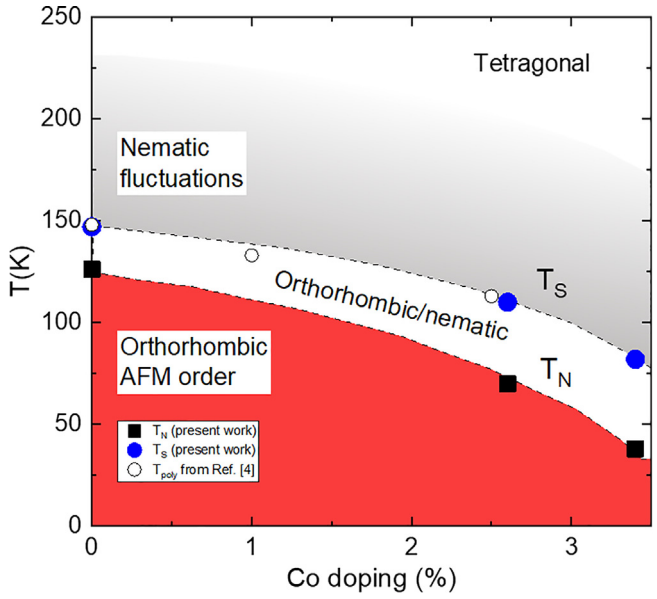

Fig. 4. Electronic phase diagram of $LaFe_{1-x}Co_xAsO$ obtained from the thermal expansion anomalies from Fig. 1 and Fig. 3. Open squares show anomaly temperatures T_{poly} obtained on poly-crystals from Ref. [4].

orthorhombicity (multiplied by the arbitrary factor 0.73) from a neutron study on polycrystalline LaFeAsO and the FWHM of the splitting of the$(220)_T$-reflection from XRD [21]. Note, that $T_S = 152$ K of the polycrystal in Ref. [21] slightly differs from T_S of our single crystal. The temperature evolution of δ, scaled to T/T_S, from our thermal expansion study nicely agrees with the splitting of the a and b lattice constants of a polycrystalline sample. However, there is a quantitative discrepancy of $\sim$ 25% between δ from the thermal study at hand and the neutron data.

The evolution of the nematic phase appears in a rather broad transition. The effect of finite uniaxial pressure inevitably applied in the capacitance dilatometer might cause such broadening as discussed for $BaFe_2As_2$ [16,23,24]. However, the observed agreement of temperature dependence of δ obtained by neutron diffration [21] (i.e., at ambient pressure) and by dilatometry (see Fig. 2) might suggest a intrinsic nature of the broadened phase transition as had been discussed in Ref. [21]. Broadening effects have also been found in diffraction studies by McGuire et al. [25] and by Nomura et al. [26] indicating that this feature is not sample specific but an intrinsic property of LaOFeAs. In particular, we conclude that finite pressure applied in the dilatometer is not the driving force for broadening the evolution of nematicity in LaFeAsO.

The length changes of the a- and b-axes and the associated orthorhombicity enable studying the suppression of T_S and of the orthorhombic splitting upon Co-doping. Fig. 3 shows pronounced broadening of the transition for $x > 0$. At $x = 0.034$, a finite δ appears already at $\sim$ 175 K, i.e., at a somehow similar temperature as compared to $x = 0$ and far above T_S ($x = 0.034$) $= 83$ K. At $T = 4$ K, δ decreases by about a factor of 2 for $x = 0.026$ while further doping to $x = 0.034$ yields only small reduction of this value.

The thermal expansion anomalies enable to construct the phase diagram of $LaFe_{1-x}Co_xAsO$ for small doping levels x (see Fig. 4). For comparison, anomaly temperatures derived from resistivity and susceptibility studies on poly-crystalline samples with $x = 0$, 0.01, and 0.025 are shown [4]. The phase diagram also reports the region where nematic fluctuation starts to evolve. While both the structure and magnetic transition temperatures decrease upon Co-doping, the splitting of T_S and T_N and hence the regime of the orthorhombic yet paramagnetic phase slightly increases. A comparison with the phase diagram of $LaFeAsO_{1-x}F_x$ [12] may suggest that upon moderate Co-doping long-range nematic ordering remains present at $T_N \leqslant T \leqslant T_S$. Moreover, above T_S , we conclude the presence of orbital and spin-nematic fluctuations in $LaFe_{1-x}Co_xAsO$ in addition to the observed structural ones.

4. Conclusions

The thermal expansion of single-crystalline $LaFe_{1-x}Co_xAsO$ has been studied for the first time. The data show anomalies associated with the onset of orthorhombic distortion and long-range magnetic order, respectively. The associated transition temperatures are well separated and are not affected by external magnetic fields of $B = 15$ T. In addition, the data enable investigating orthorhombicity and its precursing evolution well above T_S. All features are visible in the doping range $x \leqslant 0.034$ under study. The estimated 'nematic', i.e., orthorhombic yet paramagnetic phase (white region in Fig. 4) is suppressed upon Co-doping. The doping dependence of nematicity as well as of the precursing region of structural fluctuations need further investigation.

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