High-resolution thermal expansion of isovalently substituted BaFe$_2$(As$_{1-x}$P$_x$)$_2$

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Abstract. We have investigated the isovalently substituted system BaFe$_2$(As$_{1-x}$P$_x$)$_2$ by high-resolution thermal expansion using a home-built capacitive dilatometer. Accurate measurements succeeded despite the very small size of the available single crystals ($\sim 500 \times 500 \times 100 \mu m^3$). Information on the uniaxial pressure derivatives of the transition temperatures is obtained using thermodynamic relations. In-plane and out-of-plane pressure derivatives have opposite sign, which demonstrates the sensitivity of the compound to uniaxial pressure. The structural and the superconducting transition always respond oppositely to uniaxial pressure, which signals their coupling and competition.

1. Introduction
In the intensively studied 122 family of iron-based superconductors, superconductivity can be induced by various substitutions, both charged [1, 2] and isovalent [3, 4] ("chemical pressure"), and by hydrostatic pressure [5]. Uniaxial pressure effects are expected to play a key role in these compounds due to their anisotropic crystal structures. Indeed, the uniaxial pressure derivatives of $T_c$ were found to be large and anisotropic in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ by thermal expansion [6, 7]. Uniaxial pressure components are also expected to be important in the parent compound BaFe$_2$As$_2$ [8]. Recently, small high-quality single crystals of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ have been grown [9]. Here, we study uniaxial pressure effects in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ single crystals using high-resolution thermal-expansion measurements. Precise data could be obtained in spite of the extremely small (for dilatometry) crystal sizes ($\sim 500 \times 500 \times 100 \mu m^3$). We discuss the uniaxial pressure derivatives of the transition temperatures as a function of substitution. Their pronounced anisotropy underlines the importance of hydrostaticity when performing high-pressure experiments. Our results are consistent with a coupling and competition between the orthorhombic SDW phase and superconductivity.

2. Experimental
High-quality single crystals of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ were grown from stoichiometric mixtures of the components as described in Ref. [9]. The sample composition was determined by refinement of 4-
Figure 1. (a)-(c) Photographs of the single-crystalline BaFe$_2$(As$_{1-x}$P$_x$)$_2$ samples. (d) The sample of (c) glued to a thin wire. Panels (a)-(d) are on the same scale. (e)-(f) Top view and side view of the sample of (a) fixed into the sample holding spring. (g) Global view of the dilatometer with the capacitor at the bottom and the screw used to press a sample against the capacitor at the top. (h) Magnified view of the central part of (g). Samples are indicated by arrows. Note their tiny size when compared to the setup.

circle x-ray diffraction data. Uniaxial thermal-expansion coefficients were measured with a home-built capacitive dilatometer with a resolution of 0.1 – 0.01 Å. In this setup, the sample is pressed against one plate of a plate-type capacitor with a force of ~ 0.2 N. When samples are inserted such that this pressure is directed along their tetragonal [110] direction, they will be detwinned [10] and thermal expansion along the shorter orthorhombic b-axis will be measured. Comparing this to the twinned case (when the dilatometer pressure is oriented along the tetragonal [100] direction) allows to estimate the response of the orthorhombic a-axis as well.

It is a special challenge to insert the sub-millimeter sized samples into the dilatometer for in-plane measurements and to control accurately the sample orientation, which is important because of the (de)twinning. Fig. 1 illustrates the setup and sample mounting. Images of the measured samples are shown in panels (a)-(c). In order to insert the samples into the dilatometer, they are either fixed to a sample-holding spring (e,f) or to a thin wire (d). Using these aids, the samples can be inserted and their orientation inside the dilatometer can be controlled. Panel (h) shows a sample inserted on a wire as an example. Note the small dimension of the sample compared to the dilatometer. Nevertheless, high-resolution thermal-expansion data could still be obtained.

3. Results and Discussion

Fig. 2 presents the measured uniaxial thermal-expansion coefficients $\alpha_i = (1/L_i) \frac{dL_i}{dT}$ ($L$ is the sample length and “i” stands for the direction) of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ ($x = 0, 0.12, 0.25, 0.33$). Thanks to the careful sample mounting, thermal expansion could be measured along all three independent crystallographic directions: along the c-axis, along the tetragonal [100] direction (twinned, a- and b-axis) and along [110] (detwinned, orthorhombic b-axis). The strong enhancement of the anomalies in $\alpha$ along the [110] direction demonstrates successful detwinning. The magneto-structural transition of the undoped compound [11] manifests as a sharp peak of the $\alpha_i$’s at $T_{sm} = 135$ K. $T_{sm}$ decreases upon P substitution and it is found to split into two transitions for $x = 0.25$. By comparing measurements in the twinned and the detwinned configuration, we estimate that the structural transition is at $T_s = 56$ K and the SDW transition at $T_{SDW} = 49$ K.
Figure 2. Measured uniaxial thermal-expansion coefficients of BaFe$_2$(As$_{1-x}$P$_x$)$_2$. (a) Twinned in-plane measurements that give an average of the $a$- and the $b$-axis. The inset shows an expanded view of the low-temperature region. (b) Measurements in the detwinned configuration for the (underdoped) samples that undergo the tetragonal-to-orthorhombic phase transition. The inset shows how the magneto-structural transition of the $x = 0.25$ sample can be decomposed into two transitions. (c) Measured thermal-expansion coefficients along the $c$-axis.

The samples with $x = 0.25$ and $x = 0.33$ show superconductivity below $T_c = 20\,\text{K}$ and $T_c = 30\,\text{K}$, respectively. The $x = 0.25$ sample is underdoped and undergoes the superconducting transition below the structural and the magnetic transitions. For this underdoped sample, $T_c$ manifests itself as a kink in the $\alpha_i$’s, and they reach a minimum (or maximum) somewhat below $T_c$. The superconducting anomaly changes its shape drastically on the overdoped side ($x = 0.33$). It is now step-like and of opposite sign as compared to the underdoped sample.

In general, thermal expansion allows to compute uniaxial pressure derivatives of transition temperatures $dT_i/\partial P$ via the Ehrenfest relation $dT_i/\partial P = V_m \Delta C_p / T_c$. Here, $\Delta C_p$ are the discontinuities in $\alpha$ and the specific heat $C_p$, respectively and $V_m$ is the molar volume. $\Delta C_p$ is always positive. Hence, even in the absence of specific-heat data, the sign and the relative size of the anomalies in the $\alpha_i$ provide interesting information about the anisotropy of the $dT_i/\partial P$.

Due to the very small difference between $T_s$ and $T_{SDW}$ only a combined magneto-structural transition at $T_{sm}$ will be considered. The analysis further assumes that the samples remain single-phase at all temperatures even if we cannot exclude a phase separation below $T_c$. It is readily seen from the in-plane measurements that $T_{sm}$ would increase with an applied in-plane pressure. $T_c$ on the overdoped (underdoped) side of the phase diagram would decrease (increase). The uniaxial pressure derivatives along the $c$-axis have opposite sign in all cases. Further, the effects cancel largely in volume. Interestingly, the anomalies at $T_c$ and $T_{sm}$ always have the same sign in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [6, 7, 12] as in BaFe$_2$(As$_{1-x}$P$_x$)$_2$. Within the tetragonal phase (overdoped side) $T_c$ may be enhanced by increasing the separation between FeAs planes and by reducing their area. Consistently, the ratio $c/a$ increases as one approaches optimal doping from the overdoped side [9].

Comparison of measurements in the twinned (along [100]) and detwinned (along [110]) configurations further reveals a large $ab$-plane anisotropy. All the anomalies are more than twice larger in the detwinned configuration as compared to the twinned one. This means that $a$- and $b$-axis pressure derivatives are of opposite sign. The statement remains true even when...
the detwinning is incomplete or when “twinned” samples are partially detwinned. On the underdoped side, it is seen that $T_c$ and $T_{sm}$, or the mechanisms that are at the origin of the respective phase transitions, are coupled and competing. When the structural distortion is favored, superconductivity is hindered and vice versa. For example, the structural transition is favored (disfavored) by $b$-axis ($a$-axis) pressure which also decreases (enhances) $T_c$. The results may also be summarized by identifying uniaxial pressure with a shift in the phase diagram: stress along the $c$-axis (and $a$-axis on the underdoped side) mimics a higher P-content while stress along the $b$-axis (in-plane axis on the overdoped side) mimics a lower P-content. P substitution and (uniaxial) pressure are thus closely linked, as has been found previously in phosphorus- [13, 14] and also cobalt- [15] doped BaFe$_2$As$_2$.

4. Summary
We succeeded in measuring the uniaxial thermal-expansion coefficients of very small single crystals of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ for a number of substitution levels. Samples with $x = 0$, 0.12 and 0.25 undergo a magneto-structural transition. There is evidence for its splitting at the P content $x = 0.25$. Samples with $x = 0.25$ and $x = 0.33$ are superconducting. A pronounced anisotropy of the uniaxial pressure derivatives of all transition temperatures is found. In-plane and $c$-axis pressure derivatives are of opposite sign, as are $a$- and $b$-axis pressure derivatives. Further, the $dT_c/dP_i$’s change sign close to optimal doping. Derivatives of the magneto-structural and superconducting transition temperatures, $dT_{sm}/dP_i$ and $dT_c/dP_i$, are of opposite sign for all directions $i$ which is consistent with a competition between the orthorhombic SDW phase and superconductivity.

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