Effect of uniaxial strain on the structural and magnetic phase transitions in BaFe$_2$As$_2$

Chetan Dhillon, Z. Yamani, Wei Tian, J. Zeretsky, A. S. Sefat

1Department of Physics, Boston College, Chestnut Hill, Massachusetts 02167, USA
2Canadian Neutron Beam Centre, National Research Council, Chalk River, Ontario, Canada K0J 1P0
3Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA
4Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA
5Physics Department, University of California, Berkeley, California 94720, USA
6Materials Science Division, Lawrence Berkeley National Lab, Berkeley, California 94720, USA
7Materials Science Department, University of California, Berkeley, California 94720, USA

We report neutron scattering experiments probing the influence of uniaxial strain on both the magnetic and structural order parameters in the parent iron pnictide compound, BaFe$_2$As$_2$. Our data show that modest strain fields along the in-plane orthorhombic b-axis can affect significant changes in phase behavior simultaneous to the removal of structural twinning effects. As a result, we demonstrate in BaFe$_2$As$_2$ samples detwinned via uniaxial strain that the in-plane C$_4$ symmetry is broken by both the structural lattice distortion and long-range spin ordering at temperatures far above the nominal (strain-free), phase transition temperatures. Surprising changes in the magnetic order parameter of this system under relatively small strain fields also suggest the inherent presence of magnetic domains fluctuating above the strain-free ordering temperature in this material.

PACS numbers: 74.70.Xa, 74.62.Fj, 75.50.Ee, 75.40.Cx

A number of recent experiments have exploited uniaxial strain as a means of structurally detwining bilayer iron pnictide compounds in order to explore the anisotropies inherent in the electronic ground states of these materials [1-4]. A key result of many of these studies in both undoped and doped bilayer iron pnictides has been the report of dramatic anisotropies in the in-plane charge transport that surprisingly persist to temperatures far above the strain-free tetragonal to orthorhombic phase transition in these materials ($T_S$) [1]. This in-plane transport anisotropy develops between the orthorhombic $a$- and $b$-axes and is indicative of broken $C_4$ symmetry in the tetragonal phase induced via the applied, detwinning, strain field. The surprising evolution of this anisotropy as a function of doping has led to a novel picture involving an additional energy scale in the iron pnictide phase diagram possessing $C_2$ symmetry associated with nematic order [0].

Within the detwinned parent AFe$_2$As$_2$ (A = Ca, Ba, Sr, ...) (A-122) compounds, the only systems to date that exhibit this anisotropic in-plane behavior above $T_S$ are BaFe$_2$As$_2$ [1] and EuFe$_2$As$_2$ [5], both of which possess more continuous structural and magnetic phase transitions relative to other 122 variants [7,8]. This peculiarity suggests that the nominal (strain-free) phase behavior in these systems—and in particular BaFe$_2$As$_2$—may be particularly susceptible to the influence of mechanical strain relative to other 122 counterparts whose intrinsically stronger first order phase behaviors preclude anisotropy resolvable above their strain-free $T_S$ [2,9].

In Ba-122, where the phase behavior is seemingly the most continuous, an anomalous lattice softening has been observed in resonant ultrasound experiments in proximity of $T_S$ [17,18] thus suggesting a large susceptibility to the influence of applied strain fields. This fact combined with the growing number of studies utilizing uniaxial strain as a means of detwining Ba-122 and related 122-compounds [6] suggests that it is particularly important to directly explore the influence of the resulting strain fields on both its magnetic and structural phase transitions.

In this letter, we report neutron scattering measurements probing the effects of uniaxial, compressive, strain on both the magnetic and structural phase transitions within an iron pnictide parent compound, BaFe$_2$As$_2$. Neutrons allow for the bulk magnetic and structural response of the sample to be surveyed simultaneously as the planar strain field is enhanced, and phase behavior in the system can therefore be directly observed as the strain fields necessary to detwin a given crystal are applied. Similar to other iron pnictide parent materials [10-12] Ba-122 develops an ordered antiferromagnetic phase ($T_{AF}$) with $Q_{AF} = (\pi, 0, \pi)$ below $T_S$; however, within Ba-122 and other parent 122 systems these two transitions develop nearly simultaneously [13,15]. The presence of strong spin lattice coupling in these 122 parent pnictides [8,13,16] along with the potential influence of a tricritical point [15] in their phase diagrams render sharp phase transition onsets corresponding to either strongly first order [10] or nearly second order [8] phase transitions whose nature varies with the A-site ion.

Our data show that as compressive uniaxial strain fields are applied along the orthorhombic b-axis, the nominally sharp structural phase transition shifts upward and broadens in temperature accompanied by a surprising parallel increase in the onset temperature of long-range
antiferromagnetic order. Our results suggest that the strong in-plane anisotropies observed above the strain-free $T_S$ in detwinned Ba-122 can be primarily accounted for by a direct shift and renormalization of the magnetic and structural order parameters under applied strain.

BaFe$_2$As$_2$ single crystals were grown via standard, self-flux techniques [19]. Once grown, the samples were cut along the orthorhombic $a$- or $b$-axes and mounted within a small pressure clamp (Fig. 1 (a)). A belleville washer was used to monitor applied force across the sample. Samples were aligned within the [H 0, L] scattering plane using the Fmmm orthorhombic unit cell. Neutron experiments were performed on the HB-1A triple axis spectrometer at the High Flux Isotope Reactor, Oak Ridge National Lab and on the N5 triple-axis spectrometer at the Canadian Neutron Beam Centre, Chalk River Laboratories. Experiments on N5 were performed with a pyrolytic graphite (PG) monochromator ($E_i = 14.5$ meV) and analyzer with a PG filter placed after the sample and collimations of $30' - 60' - \text{sample} - 33' - 144'$. The HB-1A setup consisted of a double-bounce PG monochromator ($E_i = 14.64$ meV), PG analyzer with a PG filter before the sample collimations of $48' - 48' - \text{sample} - 40' - 68'$.

Data probing the evolution of the structural order parameter as pressure is increased along the [0, 1, 0]-orthorhombic axis are plotted in Figure 2. The metric by which the structural order parameter can be resolved necessarily evolves as the pressure is increased and the crystal is progressively detwinned. Under zero strain, the formation of twins below $T_S$ in principle allows both the $a$- and $b$-axes to be resolved within the same scattering plane and the structural order parameter $\delta = \frac{a - b}{a + b}$ to be determined; however the momentum resolution of our neutron experiments only resolves a broadening of the peak line shape below $T_S$ as the orthorhombic $a$- and $b$-axes distort away from each other in twinned samples. Due to this and in order to obtain an unbiased fit to the structural peaks as various strain fields are applied, we instead fit all peaks as single Gaussians whose width changes as the peak splits and twin domains develop.

This single Gaussian treatment of the structural peaks is appropriate in the case of a detwinned crystal; however it is only an approximation for two closely spaced Gaussian peaks in fits to data from twinned crystals. To verify that this single peak approximation is adequate in the twinned limit, we remodeled previously reported data (using similar collimations and the same spectrometer) using only a single Gaussian fit rather than two symmetrically displaced Gaussian peaks and plotted the results in Figs. 1 (b) and (c). Refitting the data gives an identical onset temperature of $T_{AF} = 136$ K to that previously reported [8] demonstrating the validity of this approximation for determining transition temperatures.

Turning now to the current BaFe$_2$As$_2$ data, the peak widths and resulting lattice parameters determined by the Gaussian peak fits to the $(2, 0, 0)$ nuclear Bragg peak are plotted as a function of temperature for three strain fields in Figs. 2 (a) and (b) respectively. The $(2, 0, 0)$
peak width broadens at $T_{AF} = 135.7$ K in the strain-free case consistent with the expected $T_S$ [5]; however as pressure along the [0, 1, 0] axis is increased progressively to 0.7 MPa the broadening observed at $T_S$ vanishes. This is explicitly shown in Figs. 2 (c) and (d) where the (2, 0, 0) peak is shown above and below $T_S$ in both 0 MPa and 0.7 MPa respectively. Within the resolution of our measurements, there is no resolvable width change in peak line shape once 0.7 MPa is applied suggesting that the crystal has been substantially detwinned.

In order to resolve the onset of the structural phase transition as the crystal is progressively detwinned, the longer in-plane lattice parameter $a$ is plotted as a function of temperature in Fig. 2 (b) for the three applied pressures. Here, the shift in the average in-plane lattice parameter for the strain-free crystal simply reflects the asymmetric splitting between $a$- and $b$-axes as the structural phase transition sets in; however under 0.7 MPa of pressure, it is immediately apparent that the onset of $T_S$ has shifted upward in temperature to $T_S \approx 147$ K. Fig. 2 (b) explicitly demonstrates that as pressure is increased, the onset temperature for $T_S$ shifts systematically upward as strain fields approaching the detwinning threshold are approached.

Now turning to the response of spin ordering to the applied uniaxial strain field, the intensity of the $Q = (1, 0, 3)$ magnetic peak is plotted in Fig. 3 (a). Similar to the response of the structural phase transition, the onset of magnetic order systematically increases as strain is increased across the sample. Specifically, an enhanced tail of magnetic scattering appears that deviates from the previously modeled power law behavior [5]. Our previous measurements in strain-free BaFe$_2$As$_2$ have already demonstrated that this tail of scattering is present across a similar range of temperatures (albeit at much lower intensities [5]) and suggest that the application of strain enhances the volume fraction of the ordered moment formation across this higher temperature scale. The spin order that develops in this tail of scattering shown in Fig. 3 (c) is long-range with a minimum spin-spin correlation length at 142 K of $\xi = 208 \pm 19 \AA$—identical within error to data collected at 3K in this sample. The diameter of the correlation length was found via $\xi = 2\sqrt{2\ln(2)}(w)^{-1}$ where $w$ is the fit Gaussian peak width in $\AA^{-1}$.

In Fig. 3 (a), at $T = 3$ K the relative scattering intensity from magnetic moments contributing to the (1, 0, 3) reflection (determined by normalizing the (1, 0, 3) to the (0, 0, 4) nuclear peak intensity) increases upon the application of increased strain. This is simply due to the progressive reduction of the volume fraction of the sample with magnetic domains whose moments are oriented out of the scattering plane. We note however that if we assume no twinning under 0.7 MPa the ordered moment at 3 K is only $0.87\mu_B$ relative to $1.04\mu_B$ measured in the twinned, zero strain, state—suggesting that the sample remains partially twinned ($\approx 43\%$) at 0.7 MPa pressure.

The tail of scattering above $T_{AF}$ in Fig. 3 (c) at present does not seem to originate from a simple picture of critical fluctuations above a second order phase transition given the lack of a resolvable divergence in the 3D spin-spin correlation length; rather the enhanced magnetic scattering seems to stabilize over large domains within the sample as strain is increased. Nevertheless, critical scattering can not be entirely precluded so we have instead identified three energy scales: (1) an AF ordering temperature $T_{AF}$ determined with a simple power law $M^2 = A(1 - T/T_{AF})^{2\beta}$ using the range of temperatures 120 K $\leq T \leq 136$ K, (2) an onset temperature, $T_{AF, onset}$, where three dimensional magnetic scattering is first resolvable, and (3) the onset of the structural phase transition $T_S$. These three energy scales are plotted as a function of increasing pressure in Fig. 3 (b).

The structural phase transition shifts upward approximately 11 K upon applying modest uniaxial pressure of only $\approx 0.7$ MPa. Increases in both $T_{AF}$ and $T_S$ manifest with strain fields below the detwinning threshold; how-

![Figure 3](image-url)
ever, upon applying enough strain to substantially detwin the sample, both $T_{AF, onset}$ and $T_S$ increase dramatically. Our results are therefore germane to studies probing intrinsic anisotropies within BaFe$_2$As$_2$ under uniaxial strain \cite{1}. Rather than simply allowing the resolution of intrinsically anisotropic electronic fluctuations that break the $C_4$ symmetry of the tetragonal phase, strain fields along the $[0, 1, 0]$-axis of Ba-122 generate a shift in the nominal, strain-free, phase behavior. From our current data however, it is not clear which shifted phase primarily drives the observed transport anisotropies in strained Ba-122. The surprising rise in the onset of AF order with increasing uniaxial strain does not simply serve to mitigate the influence of assumptions \cite{2}, we observe a progressive increase in the onset temperatures of both $T_S$ and $T_{AF}$ with increasing uniaxial strain along the in-plane b-axis. Once the crystal has been substantially detwinned, both the onset of the magnetic and structural phase transition temperatures have shifted upward dramatically and their phase behavior has broadened into a smeared, second-order-like transition. Our results suggest that the in-plane charge transport anisotropy reported above the strain-free $T_S$ in mechanically detwinned BaFe$_2$As$_2$ \cite{3} likely stems from either or both of the renormalized magnetic and structural phase transitions under enhanced strain fields.

Note added: Upon preparing this manuscript for submission, a related work was posted probing the structural phase transition’s response to uniaxial strain in BaFe$_2$As$_2$ \cite{4}. Their result reporting an upward shift in $T_S$ upon the application of strain is consistent with the results presented here.

This work was supported by NSF Award DMR-1056625 (S.W.) and DOE de-sc0002554 (Z.W.). This work was partly performed at ORNL’s HFIR, sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, U.S. DOE and partly supported by the DOE, BES, Materials Sciences and Engineering Division (A.S.). The work at LBNL was supported by DOE DE-AC02-05CH11231 (R. B.).

References:

[1] J-H. Chu et al., Science. 329, 824 (2010).
[2] M. A. Tanatar et al., Phys. Rev. B 81, 184508 (2010).
[3] Hsu-Hui Kuo et al., Phys. Rev. B 84, 054540 (2011).
[4] T. Liang et al., J. Phys. Chem. Solids 72, 418 (2010).
[5] J. J. Ying et al., Phys. Rev. Lett. 107, 067001 (2011).
[6] I. R. Fisher L. Degiorgi, and Z. X. Shen, Rep. Prog. Phys. 74, 124506 (2011).
[7] M. Tegel et al., J. Phys.: Condens. Matter 20, 452201 (2008).
[8] Stephen D. Wilson et al., Phys. Rev. B 79, 184519 (2009).
[9] E. C. Blomberg et al., Phys. Rev. B 83, 134505 (2011).
[10] C. de la Cruz et al., Nature 453, 899 (2008).
[11] Q. Huang et al., Phys. Rev. Lett. 101, 257003 (2008).
[12] S. Li et al., Phys. Rev. B 80, 020504(R) (2009).
[13] A. Jesche et al., Phys. Rev. B 78, 180504(R) (2008).
[14] Marianne Rotter et al., Phys. Rev. B 78, 020503(R) (2008).
[15] M. G. Kim et al., Phys. Rev. B 83, 134522 (2011).
[16] A. I. Goldman et al., Phys. Rev. B 78, 100506(R) (2008).
[17] Masahito Yoshizawa et al., arXiv:1111.0366
[18] Rafeal M. Fernandes et al., Phys. Rev. Lett. 105, 157003 (2010).
[19] A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, D. J. Singh, D. Mandrus, Phys. Rev. Lett. 101, 117004 (2008).
[20] I. I. Mazin and M. D. Johannes Nature Physics 5, 141 (2009).
[21] E. C. Blomberg et al., arXiv:1111.0997v1
[22] S. Ishida et al., Phys. Rev. B 84, 184514 (2011).

\*stephen.wilson@bc.edu

Stephen D. Wilson et al., Phys. Rev. B 79, 184519 (2009).