Uniaxial pressure effect on structural and magnetic phase transitions in NaFeAs and its comparison with as-grown and annealed BaFe$_2$As$_2$

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We use neutron scattering to study the effect of uniaxial pressure on the tetragonal-to-orthorhombic structural ($T_s$) and paramagnetic-to-antiferromagnetic ($T_N$) phase transitions in NaFeAs and compare the outcome with similar measurements on as-grown and annealed BaFe$_2$As$_2$. In previous work on as-grown BaFe$_2$As$_2$, uniaxial pressure necessary to detwin the sample was found to induce a significant increase in zero pressure $T_N$ and $T_s$. However, we find that similar uniaxial pressure used to detwin NaFeAs and annealed BaFe$_2$As$_2$ has a very small effect on their $T_N$ and $T_s$. Since transport measurements on these samples still reveal resistivity anisotropy above $T_N$ and $T_s$, we conclude that such anisotropy cannot be due to uniaxial strain-induced $T_N$ and $T_s$ shifts, but must arise from intrinsic electronic anisotropy in these materials.

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I. INTRODUCTION

The parent compounds of iron pnictide superconductors such as NaFeAs and BaFe$_2$As$_2$ exhibit a tetragonal-to-orthorhombic lattice distortion at temperature $T_s$ and paramagnetic-to-antiferromagnetic phase transition at $T_N$ ($\leq T_s$), forming a low-temperature collinear antiferromagnetic (AF) state with ordering wave vector along the [±1,0] directions of the orthorhombic lattice [Figs. 1(a) and 1(b)].$^{1-7}$ Because of the twinning effect in the orthorhombic AF state, AF Bragg peaks from the twinned domains in Fig. 1(c) should occur at [±1,0] and [0, ±1] positions in reciprocal space [Fig. 1(d)].$^8$ To probe the possible electronic anisotropic state (the electronic nematic phase) that breaks the $C_4$ rotational symmetry of the paramagnetic tetragonal phase in iron pnictides,$^9$ one needs to prepare single-domain samples by applying a uniaxial pressure (strain) along one axis of the orthorhombic lattice.$^{10,11}$ Indeed, transport measurements on uniaxial pressure detwinned samples of NaFeAs (Ref. 12) and BaFe$_2$As$_2$ (Ref. 13) reveal clear resistivity anisotropy above the zero pressure $T_N$ and $T_s$ that has been interpreted as arising from the spin nematic phase$^{14-16}$ or orbital ordering$^{17-23}$ in the paramagnetic tetragonal state. However, recent neutron scattering experiments on as-grown BaFe$_2$As$_2$ find that a uniaxial pressure necessary to detwin the sample can also induce a significant ($\sim$10 K) upward shift in $T_N$ and $T_s$,$^{24}$ suggesting that the observed resistivity anisotropy above the stress-free $T_N$ and $T_s$ in detwinned samples$^{12,13}$ may actually occur in the AF ordered orthorhombic state below the strain-induced $T_N$ and $T_s$. Furthermore, the resistivity anisotropy above $T_N$ and $T_s$ in as-grown BaFe$_2$As$_2$ and electron-doped BaFe$_{2-x}$Co$_x$As$_2$ becomes much smaller in annealed samples,$^{25,26}$ suggesting that the observed resistivity anisotropy in the tetragonal phase is not intrinsic to these materials but arises from the anisotropic impurity scattering of Co atoms in the FeAs layer.$^{26,27}$

In this paper, we use neutron scattering to study the uniaxial pressure effect on magnetic and structural phase transitions in NaFeAs (Ref. 4) and as-grown and annealed BaFe$_2$As$_2$.$^{20}$ While our measurements on as-grown BaFe$_2$As$_2$ confirm the earlier work that the uniaxial pressure necessary to detwin the crystal also causes significant increases in $T_N$ and $T_s$,$^{24}$ we find that similar uniaxial pressure has a very small effect on the magnetic and structural phase transitions in NaFeAs and annealed BaFe$_2$As$_2$. Since transport measurements on identical NaFeAs and annealed BaFe$_2$As$_2$ show clear resistivity anisotropy at temperatures well above the $T_N$ and $T_s$ under uniaxial pressure, we conclude that the resistivity anisotropy seen in detwinned NaFeAs and annealed BaFe$_2$As$_2$ in the paramagnetic tetragonal phase must be intrinsic properties of these materials. These results suggest the presence of an electronic nematic state in the paramagnetic tetragonal phase unrelated to the Co-impurity scattering in electron-doped BaFe$_{2-x}$Co$_x$As$_2$ family of materials.$^{26,27}$

II. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the schematic lattice and magnetic structures of NaFeAs and BaFe$_2$As$_2$, respectively.$^8$ On cooling from the high-temperature tetragonal state, NaFeAs exhibits a tetragonal-to-orthorhombic structural transition at $T_s \approx 58$ K and then a paramagnetic-to-antiferromagnetic transition at $T_N \approx 45$ K.$^7$ For comparison, $T_s$ and $T_N$ in BaFe$_2$As$_2$ occur almost simultaneously below about 138 K.$^{6,7}$ In the absence of uniaxial pressure, the low-temperature magnetic and crystal structures have equally populated twinned domains with mixed AF orthorhombic states as shown in Fig. 1(c). Figure 1(d) shows the $[H,K]$ plane of the reciprocal space where the AF and crystalline lattice Bragg peaks for a twinned sample are seen at (±1,0)/0, ±1) and (±2,0)/0, ±2) positions, respectively. Upon applying uniaxial pressure along the orthorhombic $a_0/b_0$ direction,$^13$ a single domain with sufficient large size can be achieved [Fig. 1(e)], the resulting...
self-flux method. The samples were cut to squared shapes along the \(a_o/b_o\) directions of the orthorhombic structure and fit into the aluminum-based detwining devices for both transport and neutron scattering experiments [Fig. 1(g)]. Figure 1(b) shows the comparison of transport measurements for both twinned and detwinned NaFeAs. Consistent with earlier measurements,\(^5\) we find clear resistivity anisotropy below about \(T^* \approx 70\) K. Our neutron scattering experiments were carried out using the BT-7 triple-axis spectrometer at NIST Center for Neutron Research (NCNR) and HB-1A at High-Flux-Isotope-Reactor (HFIR), Oak Ridge National Laboratory. For HB-1A measurements on BaFe\(_2\)As\(_2\), the collimations are 48°-48°-sample-40°-68°. The magnetic measurements for NaFeAs were carried out on BT-7 with open-50°-sample-50°-20° with \(E_f = 14.7\) meV. To separate the (2,0,0)/(0,2,0) nuclear Bragg peaks in the orthorhombic state of a twinned sample [Fig. 1(d)], we used tight collimation of 10°-10°-sample-10°-25° on BT-7 with \(E_f = 14.7\) meV. For NaFeAs, the lattice parameters are \(a_o = 5.589, b_o = 5.569,\) and \(c = 6.991\) Å.\(^4\) BaFe\(_2\)As\(_2\) has lattice parameters \(a_o \approx b_o \approx 5.595\) Å and \(c = 12.92\) Å.\(^6\) The wave vector \(Q\) in three-dimensional reciprocal space in Å\(^{-1}\) is defined as \(Q = H \hat{a}^*_o + K \hat{b}^*_o + L \hat{c}^*\), where \(H, K,\) and \(L\) are Miller indices and \(\hat{a}^*_o = \hat{a}_o \pi/a_o, \hat{b}^*_o = \hat{b}_o \pi/b_o, \hat{c}^* = 2\pi/c\) are reciprocal lattice units (rlu). We aligned the crystals in the \([H,0,0] \times [0,0,L]\) scattering plane, where AF Bragg peaks occur at \((\pm 1,0,L)\) with \(L = \pm 0.5, \pm 1.5, \ldots\) for NaFeAs (Ref. 4) and \(L = \pm 1, \pm 3, \ldots\) for BaFe\(_2\)As\(_2\) (Ref. 6). If the twinned domains are equally populated in the zero pressure state, AF Bragg peak intensity at \((\pm 1,0,L)\) should be the same as that at \((0, \pm 1,L)\). On the other hand, if uniaxial pressure completely detwins the sample, the magnetic scattering intensity at \((\pm 1,0,L)\) in the detwinned state should increase by a factor of 2 compared with the twinned state.

The detwining device we used is shown in Fig. 1(g). By knowing the compressibility of the spring and the area of the sample, we can estimate the applied uniaxial pressure. For our measurements, we always apply the pressure at room temperature. Since we are using springs with known force constants to apply uniaxial pressure, and thermal contractions of the sample and the aluminum holder are much smaller than the compression of the spring, applied pressure will not vary significantly with temperature. For NaFeAs, we have \(P_0 = 0, P_1 \approx 7\) MPa, and \(P_2 \approx 15\) MPa. The applied uniaxial pressures are \(P_1 \approx 7\) and \(P_2 \approx 6\) MPa for the as-grown and annealed BaFe\(_2\)As\(_2\) crystals, respectively. To determine the effect of uniaxial pressure on NaFeAs, we measure the temperature dependence of the \((1,0,1.5)\) magnetic and \((2,0,0)/(0,2,0)\) nuclear Bragg peaks. Figure 2(a) shows the temperature dependence of the magnetic \((1,0,1.5)\) peak intensity normalized to the \((2,0,0)/(0,2,0)\) nuclear peak. At zero pressure, we see a clear magnetic intensity increase below \(T_N = 45\) K. On increasing to \(P_1\) and then to \(P_2\), we see that the magnetic scattering intensity almost doubles, suggesting that the uniaxial pressure has indeed detwinned the sample. However, the Néel temperatures of the system remain unchanged at \(T_N = 45\) K within the errors of our measurements. The normalized magnetic order parameter in Fig. 2(b) shows almost identical behavior for \(P_0\), \(P_1\), and \(P_2\), thus confirming that the uniaxial pressure needed to

FIG. 1. (Color online) The lattice and magnetic structures of (a) NaFeAs and (b) BaFe\(_2\)As\(_2\). While BaFe\(_2\)As\(_2\) has the orthorhombic lattice and magnetic unit cells, NaFeAs consists of two orthorhombic chemical unit cells stacked along the c axis. The real-space schematics of a (c) twinned crystal and a (e) detwinned crystal, the two sets of domains have the same population for the twinned crystal, whereas one set of domains dominates in the detwinned crystal. In reciprocal space, the magnetic and structural peaks corresponding to the two sets of domains have equal intensities for a (d) twinned crystal, while for a (f) detwinned crystal the dominant set of domains is enhanced, while peaks corresponding to the minority set of domains have diminished intensities. Green spheres represent the magnetic \((1,0,L)\) peak and its equivalent points, red spheres represent the structural \((2,0,0)\) peak and its equivalent points. The blue arrows in (e) and (f) represent applied uniaxial pressure. (g) Schematic of the pressure device used in this work. Springs of known force constants and area of the sample edge were used to estimate the applied pressures. (h) Resistivity of twinned and detwinned NaFeAs. The dashed lines represent \(T_x\) determined from neutron scattering and \(T^*\) the onset temperature of resistivity anisotropy; the shaded region represents the temperature range of \(T^*\) under different pressures found from neutron scattering results.

AF Bragg peaks now occurring predominantly at \((\pm 1,0)\) positions [Fig. 1(f)].

We prepared high-quality single crystals of NaFeAs, as well as as-grown and annealed BaFe\(_2\)As\(_2\) crystals, using the
detwin NaFeAs has no measurable impact on $T_N$. Figures 2(c) and 2(d) show wave-vector scans along the $[0,1,0.5]$ and $[1.0,L]$ directions at $P_0$, $P_1$, and $P_2$. Consistent with the order parameter data in Fig. 2(a), the effect of uniaxial pressure is to increase the intensity of the AF Bragg peak $(1,0,1.5)$. To probe the effect of uniaxial pressure on the tetragonal-to-orthorhombic lattice distortion temperature $T_s$, we studied the temperature dependence of the lattice orthorhombicity on the $(2,0,0)/(0,2,0)$ nuclear Bragg peaks using tight collimations. If the sample is ideally detwinned, the FWHM of $(2,0,0)/(0,2,0)$ should not increase. But, because the sample is still partially twinned, the FWHM of $(2,0,0)/(0,2,0)$ shows a clear increase below $T_s = 58$ K for $P_0$, $P_1$, and $P_2$, and thus suggests that the applied uniaxial pressure also has only a small impact on $T_s$ [Fig. 2(e)]. Figure 2(f) shows the temperature dependence of the lattice orthorhombicity $\delta = (a_o - b_o)/(a_o + b_o)$ at zero pressure and its comparison with the centers of $\theta$-$2\theta$ scans. We can see a small (~4 K) increase in $T_s$ when NaFeAs is detwinned [Fig. 2(f)]. Since the lattice orthorhombicity is very small, we can not separate the $(2,0,0)/(0,2,0)$ nuclear Bragg peaks and use them to confirm the the population of each domain.

Having established that the uniaxial pressure needed to detwin NaFeAs has only a small impact on $T_N$ and $T_s$, we...
FIG. 3. (Color online) Elastic neutron scattering measurements on as-grown and annealed BaFe$_2$As$_2$ under ambient conditions ($P_0$) and with applied uniaxial pressure ($P_1$ is $\sim$7 MPa for the as-grown sample and $\sim$6 MPa for the annealed sample). The background-subtracted and normalized magnetic order parameters measured at (1,0,3) for (a) as-grown and (b) annealed BaFe$_2$As$_2$ under ambient conditions and with applied uniaxial pressure. (c) and (d) show expanded plots of the magnetic order parameter near the magnetic transition temperature. The arrows indicate temperatures at which the intensity reaches 1% of the intensity at 2 K. Full-width at half-maximum (FWHM) of $\theta-2\theta$ scans at (2,0,0)/(0,2,0) for the (e) as-grown sample and the (f) annealed sample fit with single Gaussians as a function of temperature.

investigate the effect of uniaxial pressure on $T_N$ and $T_s$ in as-grown and annealed BaFe$_2$As$_2$. From previous work on as-grown BaFe$_2$As$_2$, we know that the uniaxial pressure necessary to detwin the sample will also increase the onset of $T_N$ and $T_s$ by $\sim$12 K.$^{24}$ On the other hand, transport measurements on as-grown and annealed BaFe$_2$As$_2$ suggest that the large resistivity anisotropy in detwinned as-grown samples is due to disorder in these materials and annealing significantly reduces the resistivity anisotropy.$^{26}$ To determine how uniaxial pressure affects as-grown and annealed BaFe$_2$As$_2$, we prepared annealed samples by sealing the as-grown samples in an evacuated tube and then staying at 900°C for 50 h. Our neutron scattering measurements on $T_N$ and $T_s$ were carried out on HB-1A. Figures 3(a) and 3(c) compare the low-temperature normalized AF (1,0,3) Bragg peak intensities at $P_0 = 0$ and $P_1 = 7$ MPa for the as-grown BaFe$_2$As$_2$. While the overall magnetic intensity behaves similarly with and without uniaxial pressure, we see a clear increase in $T_N$ from $\sim$139 K at $P_0 = 0$ to $\sim$141 K at $P_1 = 7$ MPa. Therefore, the uniaxial strain-induced increase in $T_N$ is smaller than that of the earlier work.$^{24}$ This may be due to the fact that the sample used in Ref. 24 has the $T_N = 136$ K, somewhat smaller than the $T_N = 139$ K used in our experiment. For the annealed BaFe$_2$As$_2$, similar measurements showed almost identical magnetic order parameters [Figs. 3(b) and 3(d)] and a smaller shift in $T_N$ from $\sim$140 K at $P_0 = 0$ to $\sim$141 K at $P_1 = 6$ MPa. Figure 3(e) plots the temperature dependence of the FWHM of the nuclear (2,0,0)/(0,2,0) Bragg peak for the as-grown BaFe$_2$As$_2$. At $P_0 = 0$, the peak width increases abruptly below $T_s \approx 140$ K, reflecting the fact that a twinned orthorhombic crystal has slightly different lattice parameters for (2,0,0) and (0,2,0). $P_1 = 7$ MPa uniaxial pressure clearly increases the onset $T_N$ as shown in Fig. 3(c), while the $T_s$ under pressure only increases marginally to $T_s \approx 141$ K [Fig. 3(e)].
Similarly, we find that the uniaxial pressure of $P_1 = 6$ MPa on annealed BaFe$_2$As$_2$ only increases $T_\text{f}$ from $\sim 140$ to $\sim 143$ K [Fig. 3(f)].

From the experimental data discussed above, it is clear that the uniaxial pressure necessary to detwin NaFeAs and annealed BaFe$_2$As$_2$ has limited impact on $T_N$ and $T_\text{f}$. Theoretically, it has been argued that a small uniaxial strain of magnitude $\Delta \alpha$ should induce an increase in the magnetic ordering temperature $\Delta T_N = |\Delta \alpha|^{1/3}$ if $T_\text{f} = T_N$, and

$$\Delta T_N = (T_\text{f} - T_N)^{-\gamma} |\Delta \alpha|$$

if $T_\text{f} > T_N$, where the susceptibility exponent $\gamma = 2 + O(1/N)$ (with $N = 3$ corresponding to the physically relevant Heisenberg case). The structural transition temperature is also expected to increase on a scale of $\Delta T_S \sim |\Delta \alpha|^\alpha$, where for $N \rightarrow \infty$, $\alpha = 1 + O(1/N)$. Comparing with the nearly similar stochastic and magnetic phase transitions in BaFe$_2$As$_2$, the structural and magnetic phase transitions in NaFeAs are separated by $\Delta T_N = (T_\text{f} - T_N)^{-\gamma} |\Delta \alpha| = 169$ in NaFeAs should be much smaller than that of BaFe$_2$As$_2$. As a consequence, the shift of the Néel temperature $\Delta T_N = (T_\text{f} - T_N)^{-\gamma} |\Delta \alpha| \approx |\Delta \alpha|/169$ in NaFeAs should be much smaller than that ($\Delta T_N = |\Delta \alpha|^{1/3}$) in BaFe$_2$As$_2$, while the changes in structural transition temperatures ($\Delta T_S$) should be similar for both materials. Indeed, while uniaxial strain seems to have some small effect on magnetic and structural phase transitions of these materials, while transport measurements on identical materials reveal clear resistivity anisotropy above $T_\text{f}$.

In summary, we have shown that the uniaxial pressure needed to detwin NaFeAs and annealed BaFe$_2$As$_2$ has a very small effect on magnetic and structural phase transitions of these materials, while transport measurements on identical materials reveal clear resistivity anisotropy above $T_\text{f}$. We conclude then that the resistivity anisotropy is an intrinsic property in the uniaxial-strained paramagnetic tetragonal phase of NaFeAs and BaFe$_2$As$_2$.

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