Linking phonons in $SrFe_2As_2$ to magnetic fluctuations

The discovery of superconductivity in the iron pnictide materials [1] provided a new class of high-temperature superconductor that could be compared with the more-well-known cuprates. Initial investigations suggested that, like the cuprates, the high T_c of the pnictide materials could not be explained by conventional phonon-mediated pairing. None-the-less, the measured phonon dispersion in the pnictides was in surprisingly poor agreement with *ab initio* density functional (DFT) based phonon calculations. This failure appeared in several ways (see e.g. [2]) including the tendency of the non-magnetic calculations to get the wrong energies for many of the phonon modes for the samples above T_N and a tendency for calculations including magnetic order to indicate a much stronger splitting than was observed below T_N (see also Fig. 3). In fact, no phonon splitting at finite momentum transfer had been observed prior to the present work, despite calculations suggesting such splitting should be several times larger than instrumental resolution. Thus, while the phonon structure of the pnictides appeared conventional in that there were no broad modes, no large softening, and no strong temperature dependence, the dispersion was also anomalous in that it was surprisingly different than calculations.

Experimentally, careful investigation of phonons in magnetically ordered pnictides has been hampered by the twinning of materials below T_N. Investigations of twinned materials showed that the few-meV splitting predicted by calculation was certainly *not* present, but finer details might have been obscured by the presence of twin structure. Therefore, we undertook [3] to make single-domain samples of one iron-pnictide, $SrFe_2As_2$, as shown in Fig. 1: a small amount of pressure was used to preferentially select the shorter lattice constant at the structural transition (a slight distortion of the material from tetragonal to orthorhombic occurs at T_N = 200 K). This led to single domain samples below T_N, as was confirmed by X-ray diffraction. Phonon measurements at **BL35XU** and **BL43LXU** [4] then showed that while the splitting induced by the magnetic-order was smaller than calculated, it was clearly visible, as seen in Fig. 1(c,d).

The surprising phonon results, small splitting below T_N, and anomalous dispersion above T_N, can be interpreted self consistently, and quantitatively, by allowing for the presence of magnetic fluctuations. In particular, if one considers [3] the response of an atom in a harmonic potential with an effective force constant fluctuating randomly between two values, one finds the frequency response has two clear lines when the mean dwell time is longer than the inverse splitting of modes, but then collapses into a single line at the average value when the mean dwell time becomes small, as seen in Fig. 2. The measured phonon response is then qualitatively explained as follows: above T_N, the magnetic fluctuations are fast enough that all lines that are collapsed, and one sees what is essentially the average response; meanwhile below T_N some, but not all, of the fluctuations, have stabilized and the splitting appears.

The above conceptual picture was quantitatively explored [3] by separating, and then scaling, the

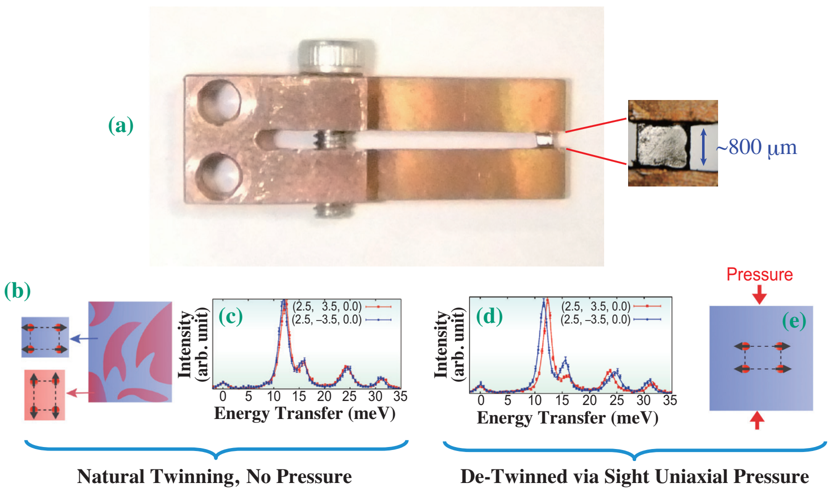

Fig. 1. Detwinning of a $SrFe_2As_2$ single crystal. The top panel, **(a)**, shows the sample glued in a holder used to exert slight pressure on the sample. The bottom panel shows the phonon response for a crystal without pressure **(c)** and detwinned **(d)**. Panels **(b)** and **(e)** schematically indicate the domain structure. For (c) and (d), the sample was held at 140K < T_N = 200 K.

anisotropic part of the force constant matrices derived from a magnetically ordered *ab initio* calculation. This led to greatly improved agreement between the calculations and measured results. In particular, below T_N, the anisotropic part of the force constant matrices had to be reduced by about a factor of 3 (see Fig. 3(a)), which, interestingly, is about the same factor as is needed to reduce the magnetic moment from the calculated to the observed value (see also discussion in [5]), and may be linked to fluctuations persisting below the onset of long-range order at T_N, in specific electronic orbitals [6]. Meanwhile, above T_N, where there is no magnetic order, selecting only the isotropic part of the force constants from the *magnetically ordered* calculation (e.g. setting the scale factor to zero) provides a much better model than the non-magnetic calculation (Fig. 3(b)).

This work solved several of the mysteries that had beset investigation of phonons in the iron pnictides. While we discuss only one material, $SrFe_2As_2$, in the "122" class of pnictides, similar disagreements between measurements and calculations exist also in other 122 materials, as well as the 1111 materials (such as PrFeAsO). Thus, it is probably reasonable to extend this picture, and calculational method, also to those materials. More generally, this is one example of how phonon response may be used to probe magneto-elastic coupling. Given better resolution (or different time scales) it may even be possible to directly probe the fluctuation time scale (e.g., the middle panels of Fig. 2) through phonon linewidth measurements. The investigation of magneto-elastic coupling is one of the goals of work at BL43LXU.

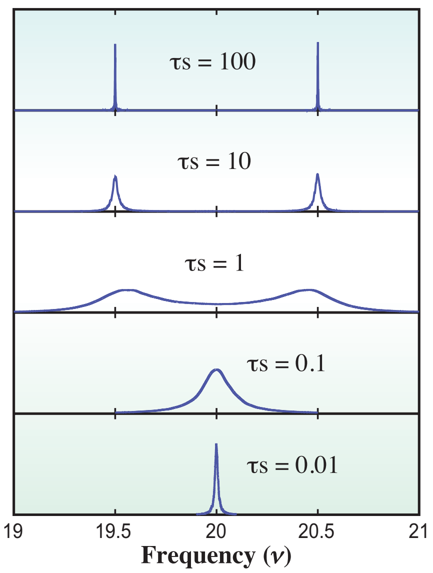

Fig. 2. Simple model for the effect of magnetic fluctuations on the phonon response. The calculations are for harmonic response of a mass m on a spring with a constant k that randomly fluctuates between two values, $k_1 < k_2$ where $s=(\sqrt{k_2}/m - \sqrt{k_1}/m)/2\pi = 1$. If the fluctuation occurs with a negative exponential distribution and has mean dwell time τ then the frequency response changes from two relatively well-defined lines for $\tau s \gg 1$ to a single line for $\tau s \ll 1$. The plot shows the frequency in units of splitting, s. (The average frequency of the phonon mode has no impact in this model [3]).

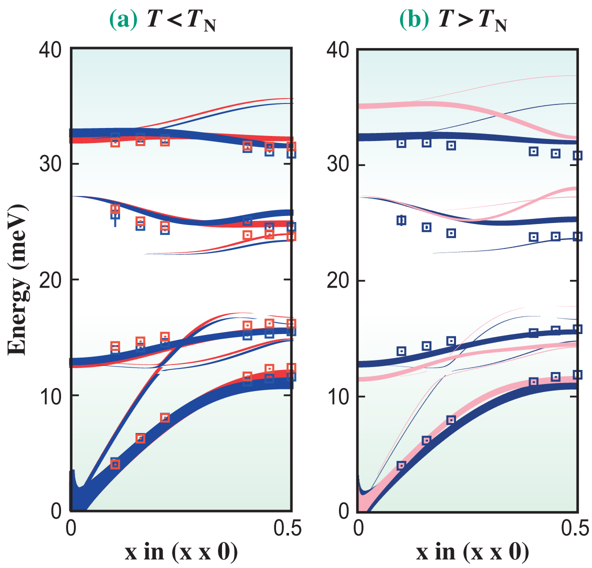

Fig. 3. Improved models incorporating reduced anisotropy to account for magnetic fluctuations. **(a)** shows the response below T_N with the anisotropy reduced by a factor of 3 while **(b)** shows the measurements above T_N compared to non-magnetic calculations (light pink) and the isotropic part of the magnetic response (light blue). The horizontal axis is the momentum transfer, the deviation from the (3 3 0) or (3 $\bar{3}$ 0) Bragg point in reciprocal lattice units (see also [3]). For the data points, the energy splitting below T_N (e.g., that in Fig. 1(d)) is visible as the slight separation between the red and blue data points in 3(a) which then merge in 3(b).

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