

Experimental report

15/09/2023

Proposal: 4-02-629

Council: 4/2023

Title: Phonon dispersion and electron phonon coupling in LiFeAs

Research area: Physics

This proposal is a new proposal

Main proposer: Akshay TEWARI

Experimental team: Navid QURESHI
Markus BRADEN
Akshay TEWARI

Local contacts: Andrea PIOVANO

Samples: LiFeAs

Instrument	Requested days	Allocated days	From	To
IN8	7	7	23/08/2023	30/08/2023

Abstract:

LiFeAs is a remarkable exception in the family of FeAs-based superconductors, as it becomes superconducting without any doping or application of pressure. Furthermore, the usual superconducting mechanism can be excluded, because LiFeAs exhibits a different band structure without nesting. Electron phonon coupling is thus a promising mechanism for this material. We propose to study the phonon dispersion in LiFeAs to search for finger prints of electron phonon coupling.

Experimental Report

Instrument	IN8
Proposal Number	4-02-629
Proposal	Phonon dispersion and electron phonon coupling in LiFeAs
Experimentalist	Akshay Tewari, Markus Braden
Local Contact	Andrea Piovano

The aim of the experiment was to study the phonon dispersion in the Fe-based superconductor LiFeAs and to search for anomalies in the temperature dependencies of the phonon frequencies. LiFeAs [1,2] is rather unique among FeAs-based superconductors (FBSC), because it is superconducting with a high T_c of $\sim 18K$, without doping or application of pressure. LiFeAs reveals no nesting between hole and electron pockets [3-7] which is incompatible with the mostly accepted pairing mechanism for FBSCs [1]. LiFeAs exhibits magnetic excitations around the same q -values as in other FeAs-based superconductors, but these fluctuations are incommensurate and carry less weight when compared to the 122-series with comparable T_c [8]. Further interest arises from the proposed topological nature of superconductivity in this material [9].

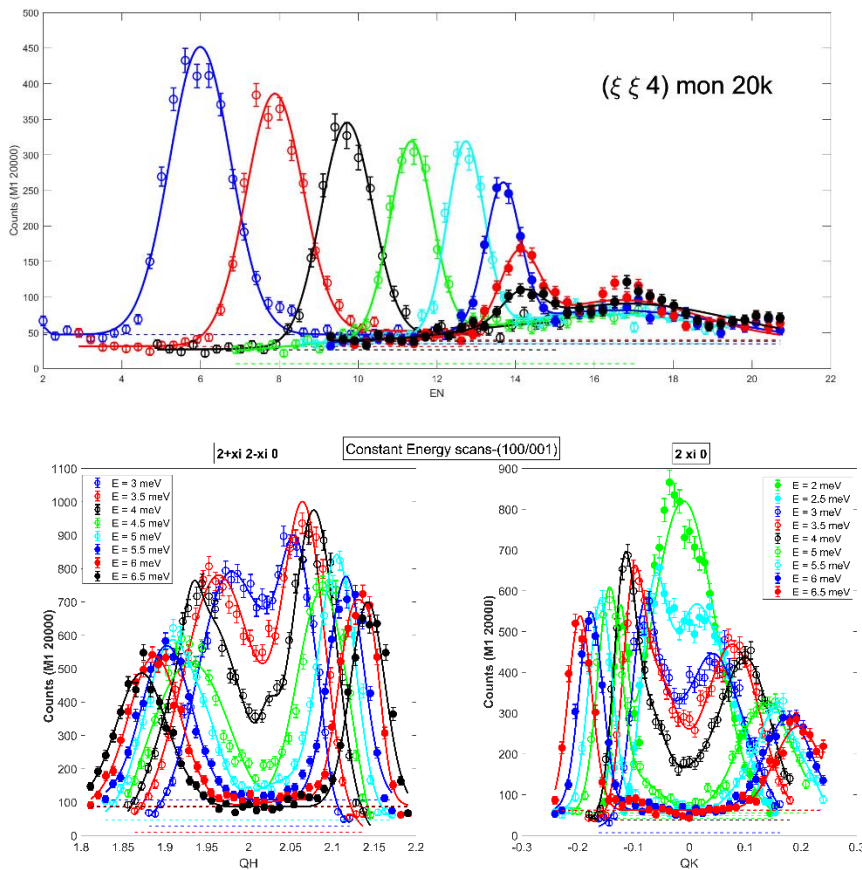


Fig. 1: Upper panel: Constant-Q scans at $(\xi\xi 4)$ with $\xi=0.15\dots 0.5$ normalized to about 30s counting time at a temperature of 1.5K. Lower panel constant-E scans ($(2+\xi 2-\xi 0)$ and $(2 \xi 0)$) across acoustic phonon modes normalized to the same monitor ($T=1.5K$).

We used two crystals grown with non-neutron-absorbing 7Li isotope that were already used in earlier INS experiments. Since LiFeAs is extremely air-sensitive, the entire sample preparation has been performed under Ar atmosphere, and the crystals were transported to the ILL in closed Al cans that finally were mounted in the orange-type cryostat. The crystal used in the first set of experiments was in $[110]/[001]$ orientation, and the second crystal was aligned with its c axis vertical to the scattering plane. Due to the activation of the crystals and due to their air-sensitivity changing the orientation on-

site is impossible. We used a Si (111) monochromator and a PG (002) analyzer, both in focusing modes. A PG filter in front of the analyzer suppressed higher-order contaminations. Most scans were performed with fixed final neutron momentum $k_f=2.662\text{\AA}^{-1}$, but in some cases, we used $k_f=3.84\text{\AA}^{-1}$ to avoid the intrinsic contamination at about 44meV. With the rather large crystals and due to the exceptional performance of IN8 data could be taken very rapidly with counting times of the order of 30 seconds even at larger phonon energies. In total more than 300 scans were recorded with many of them determining several modes. Exemplary scans aiming at acoustic phonons at a temperature of 1.5 K are shown in Fig. 1.

The experiments were guided by calculating the dynamical structure factors with a Born-von-Kármán force-constant model. Even though this model had deficiencies to describe the phonon frequencies, it could predict the Brillouin zones to observe strong phonon signals that can be attributed to single modes. In Fig. 2 we present the phonon dispersion data obtained at various Brillouin zone in the two scattering geometries.

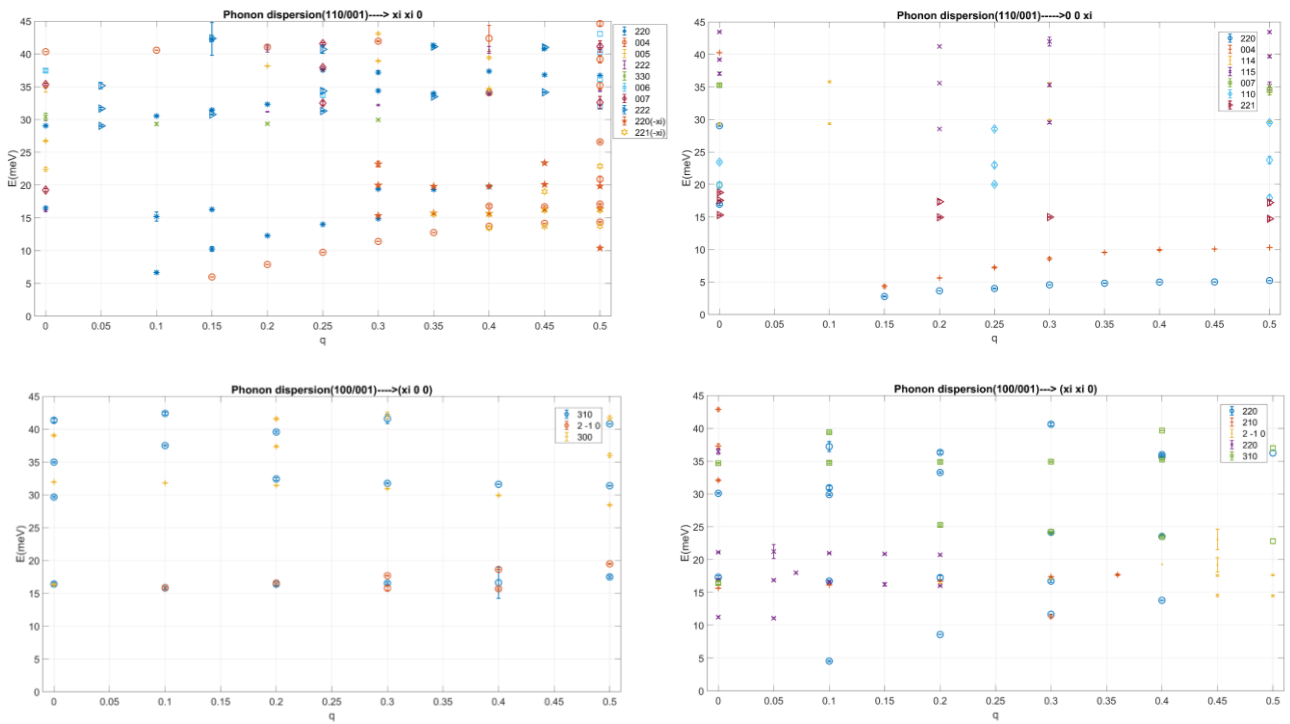


Fig. 2: Dispersion of phonon frequencies obtained by mostly constant-Q scans in various Brillouin zone, which are indicated in the legend. Data from the two scattering geometries are presented in the upper and lower two panels, respectively.

The phonon can be compared to two DFT calculations of phonon frequencies in LiFeAs [12,13]. Overall there is good agreement, but closer inspection reveals significant differences. In ref. [13] there is a pronounced instability in the longitudinal and transverse acoustic modes at the Z point (0 0 0.5) where phonon frequencies almost fall back to zero. Such behaviour is excluded by our data that do not indicate any zone-boundary instability, and it is also not seen in the calculation by Huang et al. [12]. The latter calculation, however, finds all phonon frequencies at energies below ~ 40 meV in contradiction with our measurements. Concerning these high energy modes our data better agree with the calculation by Jishi et al. [13]. Clearly improved DFT calculations are needed to compare with the experimental phonon dispersion, in order to identify phonon modes that sense an impact of some additional coupling. The experiment already finds several modes broadened compared to the resolution and the majority of modes, which can point to such coupling, see also Fig. 3. In particular

around 15 to 20meV several phonons appear to be broadened, which agrees with the energy range for strong boson coupling observed in ARPES [3,14].

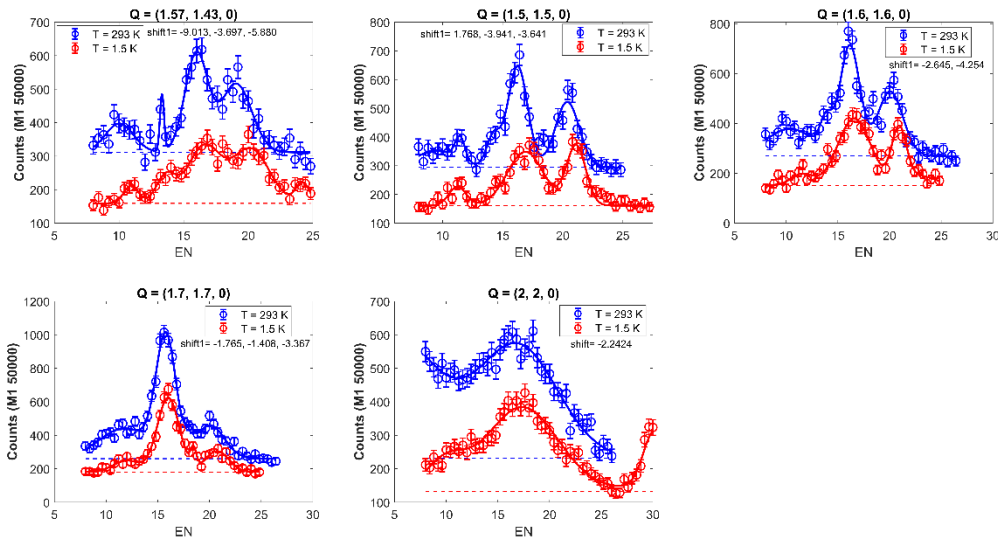


Fig. 3: Temperature dependence of selected phonon modes. Blue symbols denote room temperature data and red symbols data at 1.5 K. The relative energy shift in % is given in the insets. Some modes exhibit a very strong shift, and some modes are considerably broader than the resolution and the *normal* phonon width in LiFeAs.

For both scattering geometries, we studied selected phonon modes also at room temperature to search for further fingerprints of additional coupling. Strong relative energy shifts, $(E_{1.5K} - E_{295K}) / 0.5 * (E_{1.5K} + E_{295K})$, of above 6% were observed for some modes, but all modes harden upon cooling. In particular longitudinal modes with the propagation vector close to $(0.5, 0.5, 0)$ exhibit strong shifts, which can point to an electronic effect, because the strong magnetic excitations are also observed in this part of the Brillouin zone [8]. We also studied the transversal acoustic phonons that can couple to the nematic instability, for which one can expect some anomalous softening at low temperature, but in agreement with a brief earlier phonon study [11] and with a recent elastoresistance measurement, no evidence for such an effects was observed.

In summary the experiment with two crystal orientations allowed us to fix the phonon frequencies of most modes along the $(\xi 0 0)$ $(\xi \xi 0)$ and $(0 0 \xi)$ paths in the tetragonal Brillouin zone. In addition, some modes are heavily broadened or exhibit a strong temperature dependency that exceeds what can be explained in a purely structural approach, in particular concerning the in-plane polarized modes.

[1] P.J. Hirschfeld, C. R. Physique **17**, 197 (2016) [2] Tapp et al., PRB **78**, 060505 (2008). Wang, Sol.St.Com. **148**, 538 (2008). Pitcher et al., Chem. Com. 5918 (2008). [3] S. Borisenko et al., Phys. Rev. Lett. **105**, 067002 (2010), A. A. Kordyuk et al., PRB **83**, 134514 (2011) [4] H. Eschrig and K. Koepernik, Phys. Rev. B **80**, 104503 (2009) [5] A. Lankau et al., Phys. Rev. B. **82**, 184518 (2010) [6] S. H. Baek et al., J. Phys.: Condens. Matter **25** 162204 (2014) [7] Brydon et al., Phys. Rev. B **83**, 060501 (2011). [8] N. Qureshi et al., Phys. Rev. Lett. **108**, 117001 (2012); Phys. Rev. B **90**, 144503 (2014) [9] L. Kong et al., nat. Comm. **12**, 4146 (2021); P. Zhang et al., Nature Physics **15**, 41 (2019) [10] M. Wissmann et al., Phys. Rev. B **106**, 054508 (2022) [11] A. Merritt et al., J. Nov. Supercon. **29**, 3049 (2016) [12] G.Q. Huang et al., Phys. Rev. B **82**, 014511 (2010) [13] R. A. Jishi and H. M. Alyahaei, Adv. In Cond. Mat. Phys. 804343 (2010) [14] G. Li et al., Chin. Phys. B **29**, 107402 (2020) [15] R. P. Day et al., Phys. Rev. B **105**, 155142 (2022).