Experimental report

11/11/2021

Proposal: 5-23-709 **Council:** 4/2018

Title: Symmetry-lowering in the nematic phase of FeSe superconductor

Research area: Physics

This proposal is a new proposal

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Samples: FeSe

Instrument	Requested days	Allocated days	From	To
D2B	2	2	19/06/2019	21/06/2019
D4	4	4	19/02/2020	23/02/2020

Abstract:

The aim of this proposal is to perform a combined Rietveld and pair distribution function (PDF) analysis of neutron diffraction data collected at low temperatures on the structurally simple prototype Fe-based superconductor FeSe. Unconventional superconductivity is well known to emerge in the vicinity of symmetry-breaking phase transitions. In FeSe, a precursor state with a pseudogap has been identified based on several anomalies in the electronic properties above the superconducting transition at Tc l 8 K. Basing on our preliminary results obtained at the ID22 beamline of the ESRF, here, we would like to investigate a structural component associated with this precursor state using a combined Rietveld and pair distribution function (PDF) analysis of powder neutron diffraction data.

Symmetry lowering in the nematic phase of FeSe superconductor

The structurally simplest Fe-based superconductor FeSe is an unusual material with superconducting transition temperature T_c = 8 K, which can be enhanced up to 37 K by application of pressure. It undergoes a nematic phase transition at T_s = 90 K, at which the crystal structure changes from tetragonal to orthorhombic symmetry. In addition, the compound displays anomalies in the electronic properties at $T^* \approx 75$ K and $T^{**} \approx 30$-20 K [1]. We recently performed a combined Rietveld and pair distribution function (PDF) analysis of neutron diffraction data collected at low temperatures and investigated the local orthorhombic strain associated with different electronic anomalies in FeSe.

For the reciprocal-space structure analysis (Rietveld refinement) the data were collected on the high resolution powder diffractometer D2B (λ = 1.596 Å) in the temperature range 3–90 K in the heating cycle. For real-space PDF analysis, the diffraction patterns were collected at the D4c instrument (λ = 0.4959 Å) at temperatures 150 and 7 K upon cooling and in the temperature range 3-150 K plus 298 K, upon heating.

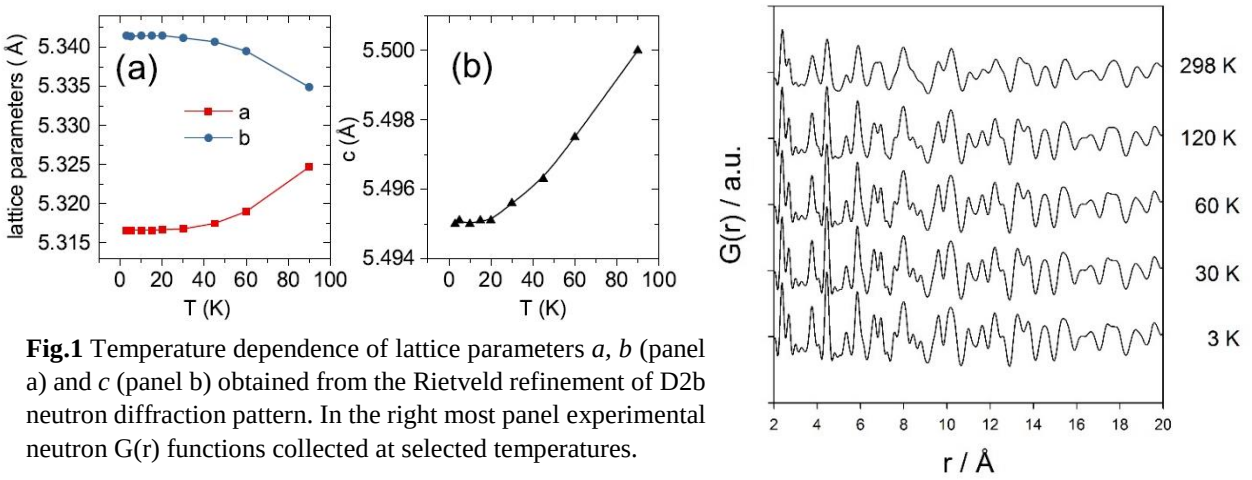

Fig.1 Temperature dependence of lattice parameters *a, b* (panel a) and *c* (panel b) obtained from the Rietveld refinement of D2b neutron diffraction pattern. In the right most panel experimental neutron G(r) functions collected at selected temperatures.

In Fig. 1(a) and 1(b), the lattice parameters obtained by Rietveld refinements of D2B data are displayed. The diffraction data were refined using the orthorhombic *Cmma* model.
The right most panel of Fig. 1 reports selected reduced pair distribution function curves G(*r*) computed using the data collected at D4c (Q_{max} = 23.6 Å^{-1}).

In Fig. 2(a), the ratio of lattice parameters *b/a* obtained from the reciprocal-space Rietveld analysis and PDF G(*r*) analysis for different *r* ranges are plotted. We should also note that in the 2-5 Å range, the G(*r*) samples mainly samples distances between atoms in the same FeSe plane while for larger *r* ranges, inter-plane contacts have increasing weight in the G(*r*). This result suggests a larger crystallographic coherence of the orthorhombic distortion in the *ab* plane compared to the direction of the *c* axis. Compared to the average values, the *b/a* ratio increases for refinements focusing on the short-range parts of the *G(r)* functions, which suggests that the orthorhombic strain is locally enhanced. For 2-7 and 2-5 Å ranges, the *b/a* ratio reaches a plateau above 75 K and up to 150 K when the *r* interval is the shortest. Note that 75 K corresponds to the temperature T^*, which is considered as the onset temperature of spin fluctuations [2]. For the *r* ranges 2-7 Å and 2-5 Å a kink can be seen around 20 K, which coincides with the electronic

anomaly observed at T^{**}. Figs. 2 (b)-(e) display the anisotropic displacement parameter U(Fe) and U(Se) values refined in different r ranges. To avoid correlations among the parameters, we assumed $U_{11} = U_{22}$ for both Fe and Se in the $G(r)$ refinements.

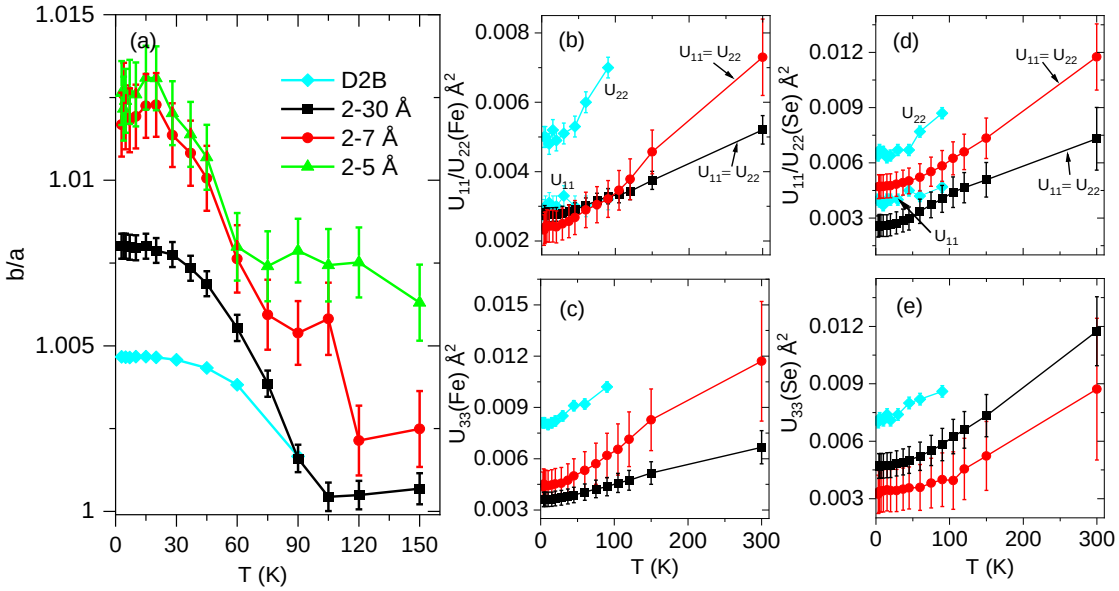

Fig. 2 Comparison of parameters obtained from the neutron diffraction data analysis. (a) Ratios of lattice parameter b/a, (b)-(e) anisotropic displacement parameters U_{11}, U_{22} and U_{33}, obtained from both reciprocal space refinements of D2B measurements and real-space Rietveld analysis of D4c $G(r)$ functions for different r ranges. In the $G(r)$ refinements, we fixed $U_{11} = U_{22}$ for both Fe and Se. Legend in (a) applies to all panels. The error bars for the D2B data shown in (a) are smaller than the symbol size.

In Fig. 3, atomic distances obtained from the analysis of local structure $G(r)$ and the corresponding distance obtained from the Rietveld analysis are plotted. For the distances considered, it can be noted that, (i) the shortest Fe–Se distance in the local structure is slightly shorter than the corresponding distance in the average structure, (ii) the shorter Fe–Fe distance in the local structure is always larger than that in the average structure, and (iii) the Fe–Se distance is ≈4.45 Å in both local and average structures.

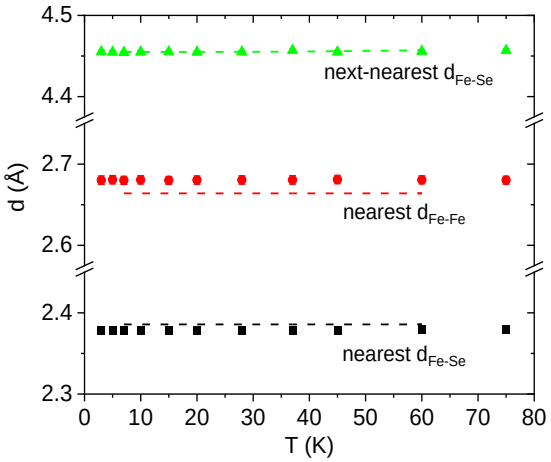

Fig.3 Neutron diffraction $G(r)$ peak positions in the temperature range 3 - 75 K compared with the corresponding distances obtained from the reciprocal space Rietveld refinement of the D2B data. The error bars are smaller than the symbol size.

Although the nematic phase transition occurs in FeSe at T_s = 90 K, at which the average crystal structure changes from tetragonal to orthorhombic, the short-range orthorhombic distortion exists already at 300 K and extends over a length scale of 10-30 Å [3]. We considered both *Cmma* and *P4/nmm* models for the full temperature range studied here. For the neutron diffraction data taken at 298 K, the *Cmma* model provided lower residual values than the P4/nmm for *r* values as small as 5 Å. Our results are thus in full agreement with the earlier PDF work in Ref. [3].

Our studies did not show any further lowering of crystal symmetry than *Cmma* in the nematic state and the anomalies at temperatures *T** and *T*** are found to be purely of electronic origin. The local symmetry remains *Cmma* also in the superconducting state down to at least 3 K. The local orthorhombic symmetry spanning three orders of magnitude in temperature is yet another trait of FeSe. A manuscript based on this work is under preparation.

References

[1] S. Rößler *et al.*, Phys. Rev. B **92**, 060505(R) (2015).
[2] V. Grinenko *et al.*, Phys. Rev. B **97**, 201102(R) (2018).
[3] R. J. Koch *et al.*, Phys. Rev. B **100**, 020501(R) (2019).