

Experimental report

17/02/2020

Proposal: 7-02-188

Council: 4/2019

Title: Phonon softening at the incommensurate phase transition of the chargedensity wave compound TbTe₃

Research area: Physics

This proposal is a resubmission of 7-02-179

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Samples: TbTe₃

Instrument	Requested days	Allocated days	From	To
IN22	7	0		
THALES	7	5	28/01/2020	03/02/2020
ORIENTEXPRESS	2	2	31/01/2020	02/02/2020

Abstract:

The possibility for charge density waves (CDW) coexisting with superconductivity in the high T_c cuprates has sparked a renewed interest in CDW formation mechanism and dynamics. A long standing fundamental controversy that goes back to the early days is the mechanism by which the CDW state is created, either weak or strong electron-phonon coupling (WEPC or SEPC). Experiments show evidences of SEPC in a many CDW systems and those shown in the quasi 2D rare-earth tritelluride TbTe₃ are amongst the most convincing ones. Among other features the Kohn anomaly in the phonon spectrum do not behave as expected in the WEPC theory. Previous inelastic X-ray scattering (IXS) experiments reported a large phonon softening at the approach of the phase transition temperature, T_C=330K. However our IXS experiments do not show clear-cut evidences of the occurrence of a phonon softening. Indeed the data can rather be explained by an increasing of the phonon damping at the approach of T_C and the occurrence of a central peak. In this experiment we want to measure the phonon softening with 5-10 times better energy resolution than IXS in order to disentangle this problem.

Experimental report 7-02-188 TbTe₃ sur Thales:

Aim of the experiment:

The possibility for charge density waves (CDW) coexisting with superconductivity in the high T_c cuprates has sparked a renewed interest in CDW formation mechanism and dynamics. A long-standing fundamental controversy that goes back to the early days is the mechanism by which the CDW state is created, either weak or strong electron-phonon coupling (WEPC or SEPC). Experiments show evidences of SEPC in a many CDW systems and those shown in the quasi 2D rare-earth tritelluride TbTe₃ are amongst the most convincing ones. Among other features the Kohn anomaly in the phonon spectrum do not behave as expected in the WEPC theory. Previous inelastic X-ray scattering (IXS) experiments reported a large phonon softening at the approach of the phase transition temperature, $T_C=330\text{K}$. However previous IXS experiments from our group did not show clear-cut evidences of the occurrence of a phonon softening. Indeed, the data can rather be explained by an increasing of the phonon damping at the approach of T_C and the occurrence of a central peak.

In this proposal we wanted to fully characterize the evolution of the soft mode at the approach of T_C , taking advantage of the good energy resolution of cold three-axis spectrometer. We were especially interested in the occurrence of the central peak and the dynamics below T_C , in particular the decoupling of the soft mode into a phason and amplitudon mode.

Experimental details:

Single crystals of TbTe₃ grow in platelet shape with the crystallographic b^* axis perpendicular to the platelet. We used a 120 mg single crystal, oriented with the $[3,3,1]$ and $[0,1,0]$ directions in the scattering plane, in order to access the CDW ordering wave vectors observed in X-ray diffraction experiments [1,2]. The use of the goniometers on the instrument with the 3D option of NOMAD gave access to out of scattering plane components in a restricted Q range limited by the angular range of the goniometers with mounted cryostat. The sample was measured in the temperature range $340\text{ K} > T > 2\text{ K}$. We used a Si(111) monochromator and PG(002) with k_f fixed of $k_f=3\text{ \AA}^{-1}$, giving access to a dynamical range within $|Q|<5\text{ \AA}^{-1}$. In the second part of the experiment we performed additional scans at low Q in a standard configuration with $k_f=1.5\text{ \AA}^{-1}$, without Be filter in order to avoid collisions between the strongly inclined cryofurnace and the filter. The orientation of the crystal has been verified with several in-plane Bragg peak and the out-of scattering plane peaks $\{112\}$.

Results:

CDW order, structural peaks:

Structural peaks, related to the incommensurate CDW order, appear at propagation vectors $\mathbf{k}_{CDW}=(0,0,0.3)$. We systematically measured positions at room temperature, previously observed by x-ray diffraction experiments and known to have large scattering intensity. Fig. 1a shows the temperature dependence of the $(3,7,0.3)$ peak. Although supposedly the strongest among the measured peaks, the measured intensity was of the order of 10^1 n/s . No lattice shifts have been observed within the selected temperature range. The cryofurnace turned out to be slightly unstable in the needed temperature range, leading to large error bars on the integrated intensity and full width half maxima (FWHM) of the observed peak. Despite the

noisy data, a clear (linear) temperature dependence can be observed, with a disappearance of the static signal above $T=335$ K, in agreement with the results published in the literature [1-2]. Measurements related to the lattice dynamics turned out to be impossible due to the lack of intensity by roughly 10^3 orders of magnitude, assuming a similar behavior of the structure factor for phasons, as for standard acoustic phonons. Test measurements have consequently not shown any signature of excitations related to low energy lattice dynamics.

CDW related to magnetic order:

Recent literature reports about a possible coupling of CDW to the magnetic system [2]. Although CDW order is believed to take place in the Te-Te layers, these periodic modulations might influence the electronic states of the 4f-multiplet splitting of Tb^{3+} and introduce non-spherical charge distribution varying from site to site in the adjacent Tb-Te layers. Consequently, CDW order has also been observed related to the magnetic structure below an ordering temperature of about $T_N=5.7$ K. While the position seems to be related to the CDW, the intensity is related to the magnetic ordering and disappeared above T_N reported in the literature [3]. Figure 2 shows scans along the L direction close to the magnetic Bragg position $Q_M = (1,1,0.5)$. Magnetic intensity is observed at the position $L = 0.2$ r.l.u., corresponding to CDW induced magnetic peak at the position $Q_{CDW,M}=(1,1,0.2)=Q_M-k_{CDW}$. The intensity disappears above $T \sim 6$ K.

In the remaining time, we attempted to measure some inelastic signals, as shown in Figure 3. The energy scan performed at $Q_{CDW,M}$ shows intense sharp signals of presumably four dispersing excitation levels. A detailed study of the excitation levels at different temperatures

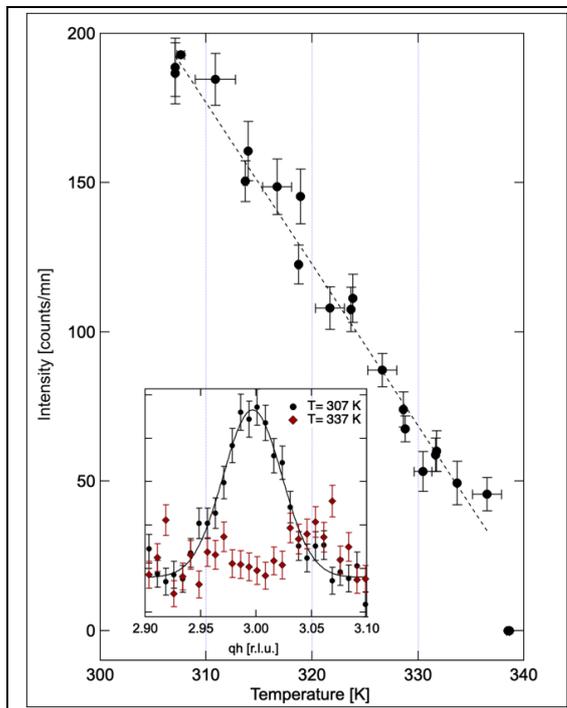


Fig.1: Temperature dependence of the CDW structural peak (3,7,0.3).

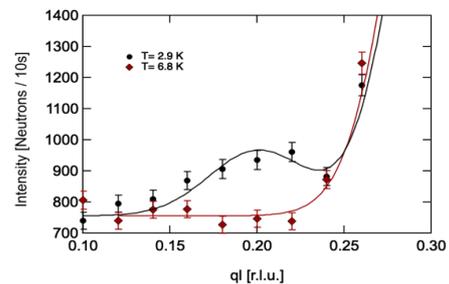


Fig.2: Magnetic intensity observed around the position $Q_{CDW,M}=(1,1,0.2)$ below (black circles) and above (red diamonds) the order temperature $T_N=5.7$ K.

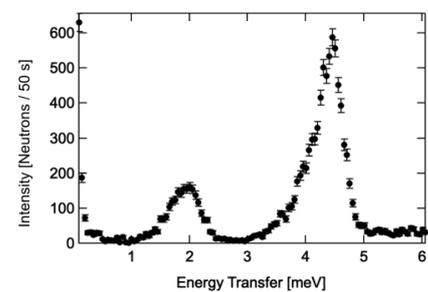


Fig.3: Energy scan around at the position $Q_{CDW,M}$ at $T=2.8$ K.

is envisaged.

- [1] N. Ru, et al., Phys. Rev. 77, 035114 (2008)
- [2] W.S. Lee, et al., Phys. Rev. B 85, 155142 (2012)
- [3] F. Pfuner, et al., J. Phys. : Condens Matter 24 (2012) 036001