ESRF	**Experiment title:** IXS investigation of a possible phonon-softening mechanism driving the Peierls-type phase transition in the complex carbide Sc_3CoC_4	**Experiment number**: HC-4514
Beamline: ID28	**Date of experiment**: from: April 13th, 2021 to: April 19th, 2021	**Date of report**: July 21st, 2021
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Report: In the low-dimensional complex carbide Sc_3CoC_4 the completion of a Peierls-type structural distortion at 82 K is followed by the onset of superconductivity at 4.5 K.[1–4] This combination of two usually incompatible states, *i.e.* a structurally distorted and a superconducting state, sets the compound apart from other, in some cases already widely studied low-dimensional materials like *1T*-$TiSe_2$[5] and $ZrTe_3$.[6] The mechanism underlying the structural phase transition in Sc_3CoC_4 and the onset of low-dimensional superconductivity[7] is not completely settled. Namely, no exhaustive explanation reconciling the appearance of two anomalies in the temperature-dependent electrical resistivity $\rho(T)$ at 150 K and 82 K (Fig. 1a) with the observation of only one distinct structural transition has been given so far. Recent combined experimental and theoretical investigations suggest a relation with an extended soft-phonon mechanism driving the Peierls-type distortion.[8]

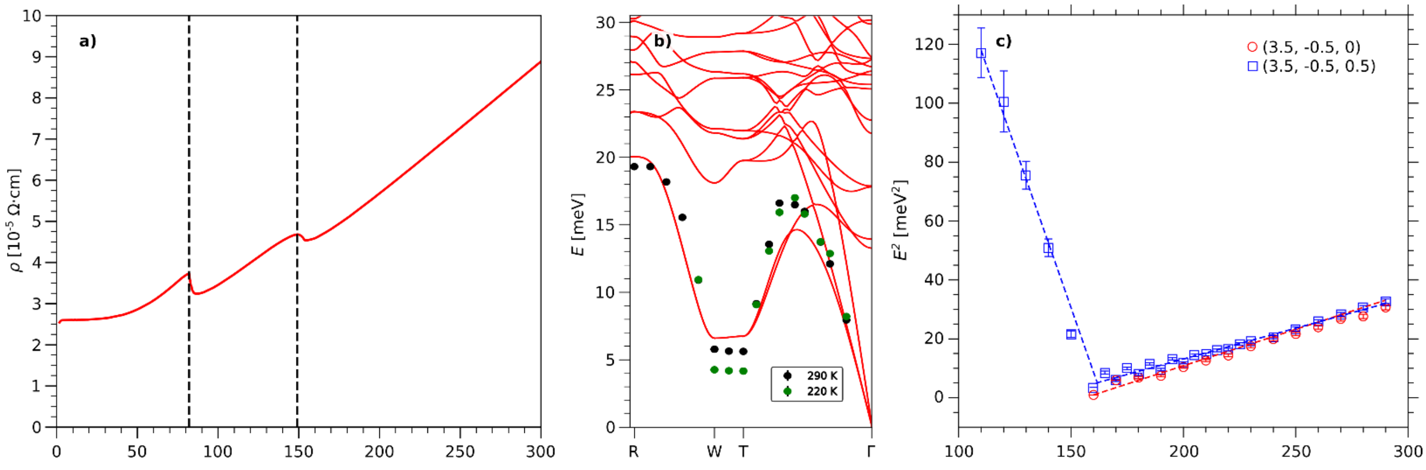

Fig. 1: a) Temperature-dependent electrical resistivity $\rho(T)$; b) calculated (red line) and measured phonon energies at 290 K (black circles) and 220 K (green circles) along the high-symmetry path R-W-T-Γ in the Brillouin zone of HT-Sc_3CoC_4; c) temperature-dependent softening of the entire phonon branch between W (3.5, -0.5, 0.5) and T (3.5, -0.5, 0) and subsequent re-hardening at W.

Samples for the remote experiment were prepared by standard arc-melting procedures. From several samples sent to the beam line one Sc_3CoC_4 single crystal was selected by preliminary diffuse scattering experiments at ID28 side station at various temperatures, confirming high sample quality and presence of the expected significant diffuse scattering. The sample was then transferred to the inelastic main beamline and temperature scans between room-temperature (RT) and 100 K were performed for *q* points around (3.5, -0.5, 0) and

(3.5, -0.5, 0.5). Apertures were set to a beam profile of 20 x 55 µm^2 and the Si (9,9,9) reflection (corresponding to an energy of 17.8 keV) with a data integration time of 40 s per scan was used throughout.

The analysis of the *T*-dependent IXS spectra unambigously proves an unusual phonon-softening process between RT and 150 K affecting both *q*-points. Already at RT significant inelastic scattering contributions at low energies hint at the existence of a soft phonon branch between the W (3.5, -0.5, 0.5) and T point (3.5, -0.5, 0) of the Brillouin zone (Fig. 1b). Inspection of the *T*-dependent phonon energies in Fig. 1b and Fig. 1c then reveals a softening process with a square-root temperature behavior that reaches its energy minimum around 150 K. Here, the soft phonon branch at T disappears due to its back-folding to Γ, whereas the branch at W re-stiffens when cooling below the transition temperature of 150 K (Fig. 1c). Further data collected at *q*-points in the vicinity of the soft W-T branch shows a very good agreement with the results of our DFT phonon-dispersion calculations (Fig. 1b). We may therefore conclude, that the structural phase transition in the title compound is indeed initiated by a phonon softening process which is already ongoing at room-temperature and finishes at about 150 K.

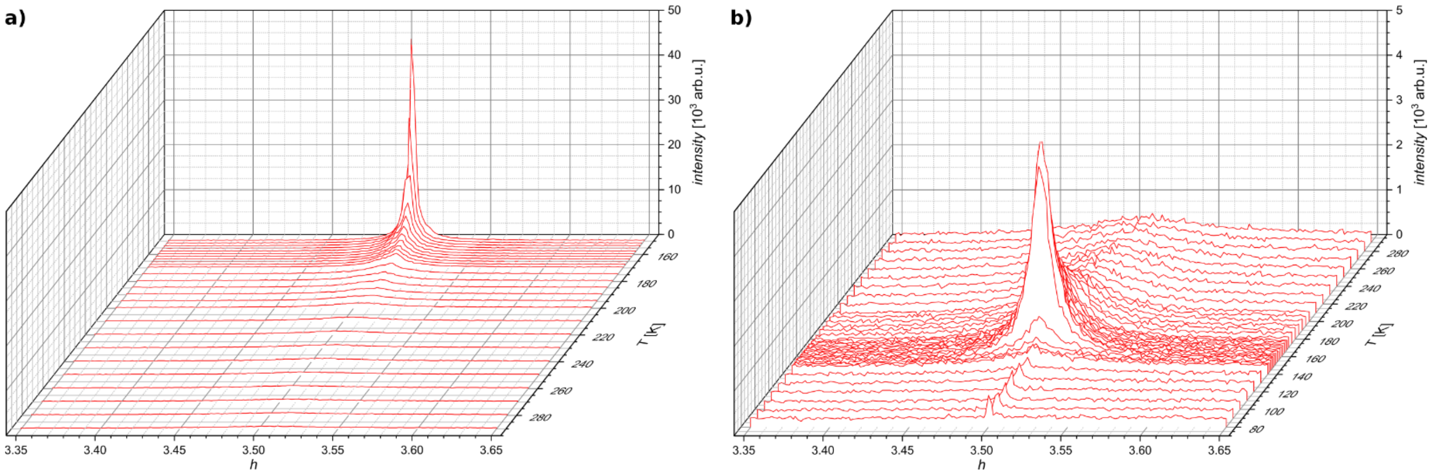

Fig. 2: Temperature-dependent development of diffuse scattering intensity a) at (3.5, 0.5, 0) corresponding to T and b) at (3.5, 0.5, 0.5) corresponding to W. Diffuse data in a) is only shown down to 150 K due to the larger intensity scale of the superstructure reflections emerging below this temperature.

The completion of the phonon softening process already at 150 K leaves the question about the significance of the 82 K anomaly in $\rho(T)$. Therefore, within the alotted beamtime further diffuse scattering experiments were conducted for temperatures from RT down to approx. 80 K. In line with our preliminary in-lab experiments, these experiments showed intense lines of diffuse scattering extending parallel to c^* with a distinctly different temperature-dependent behaviour of the points corresponding to T and W (Fig. 2a and Fig. 2b). While at T the condensation of diffuse intensity into the characteristic superstructure Bragg peaks of the LT phase of the title compound is observed between RT and 80 K, the data obtained at W reveals an unexpected and up to now unknown behaviour. In line with the results of the inelastic scattering experiments, the diffuse intensity increases with decreasing temperature down to 150 K. Around 150 K, a sharp peaking of the diffuse intensity in a lambda-shaped fashion is observed. In contrast to our preliminary in-lab scans, however, diffuse intensity could still be observed at positions between the superstructure reflections below 150 K, *i.e.* also after termination of the phonon-softening process. Comparison with the inelastic scattering data shows that this diffuse intensity is quasi-elastic in nature. It might be speculated that the anomaly observed in $\rho(T)$ at 82 K marks the completion of an ordering process of the structural distortion in the well separated layered building units of Sc_3CoC_4 along the *c* axis.

References:

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