# Experimental report

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| **Proposal:** 7-02-224 | **Council:** 4/2023 |

**Title:** Soft phonon mode in BaNi2(As1-xPx)2

**Research area:** Materials

**This proposal is a new proposal**

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**Experimental team:** Frank WEBER
Tom Laurin LACMANN

**Local contacts:** Paul STEFFENS

**Samples:** NbSe2
BaNi2As2
BaNi2(As0.929P0.071)2

| Instrument | Requested days | Allocated days | From | To | |
|---|---|---|---|---|---|
| ORIENTEXPRESS | 1 | 1 | 24/05/2023 | 25/05/2023 | |
| THALES | 6 | 6 | 22/11/2023 04/04/2024 | 23/11/2023 09/04/2024 | |

**Abstract:**

BaNi2As2, a candidate system for charge-driven electronic nematicity, features an incommensurate charge-density-wave (CDW) order which can be suppressed by 10% of P-doping in BaNi2(As1-xPx)2. Recently, inelastic x-ray scattering (IXS) identified the corresponding CDW soft phonon mode in BaNi2As2. Contrary to expectations, it seems that the soft mode already acquires zero energy up to 30K above $T_{CDW}$ = 146K, though the temperature where the soft mode indeed reaches zero energy could not be identified unambiguously. Here, we propose to measure the T-dependence of the CDW soft mode in BaNi2(As1-xPx)2 with x = 0 and x = 0.07 ($T_{CDW}$ = 85K). Most importantly, the Gaussian-like INS energy resolution will enable a clear distinction of the Lorentzian-like (nearly) overdamped soft mode from the elastic line ¿ impossible in IXS due to the Lorentzian-like resolution function. Results for x=0 will clarify the soft mode behavior and measurements for x = 0.07 will shed light on the soft mode when $T_{CDW}$ is suppressed by a factor of two and better separated from a triclinic phase transition following at lower temperatures.

We performed a study of the soft phonon mode in BaNi$_2$As$_2$ at **Q** = (0.72,2,-1) along with measurements of the dispersion along the line **Q** = (H,2,-1), 0.5 ≤ H ≤ 1.0. Measurements were done at T = 144K – 300K.

% Motivation

The idea for the experiment was based on recent publications[1,2] studying the soft phonon mode of the charge-density-wave (CDW) phase transition in BaNi$_2$As$_2$. In particular, Song et al.[2] reported that the soft phonon mode is fully damped already at T = 180 K, which is about 30 K above the reported CDW transition temperature[3]. Previous work on the soft phonon mode was done via inelastic x-ray scattering (IXS). The aim of the current experiment was to employ the much better energy resolution of inelastic neutron scattering.

% experimental setup

The experiment was carried out on the ThALES triple-axis spectrometer because the velocity selector allows to use a random final energy. In our case, we wanted a low final energy $E_f$ (to have a good resolution). However, we had to use $E_f$ = 10 meV because the scattering triangle could not be closed with a smaller $E_f$. Still, this resulted in a considerable improvement of the energy resolution compared to standard $E_f$ = 14.7 meV, which is typically used on thermal-neutron TAS. We did not use any filter in the scattered beam. Finally, we were able to remove the velocity selector, too, since it turned out that no spurious scattering or additional background were present. The allowed us to measure up to (for ThALES looking at the cold neutron source) relatively high incident energies of up to $E_i$ = 19.75 meV, wich corresponds to an energy transfer of 9.75 meV in Figure 1(a).

The sample was a 250-mg piece grown by Amir Haghighirad in the end of March 2024 (batch: AAH 184). It was mounted in a standard Orange cryostat and measurements were done at temperatures T = 144K – 300K.

% Results

Apart from the good energy resolution (FWHM = 0.4 meV at zero energy transfer), INS offers another advantage, which is the Gaussian-like resolution function. This should make it easier to distinguish between the resolution-limited elastic line and the soft phonon mode which can be represented by a damped harmonic oscillator (DHO) function.
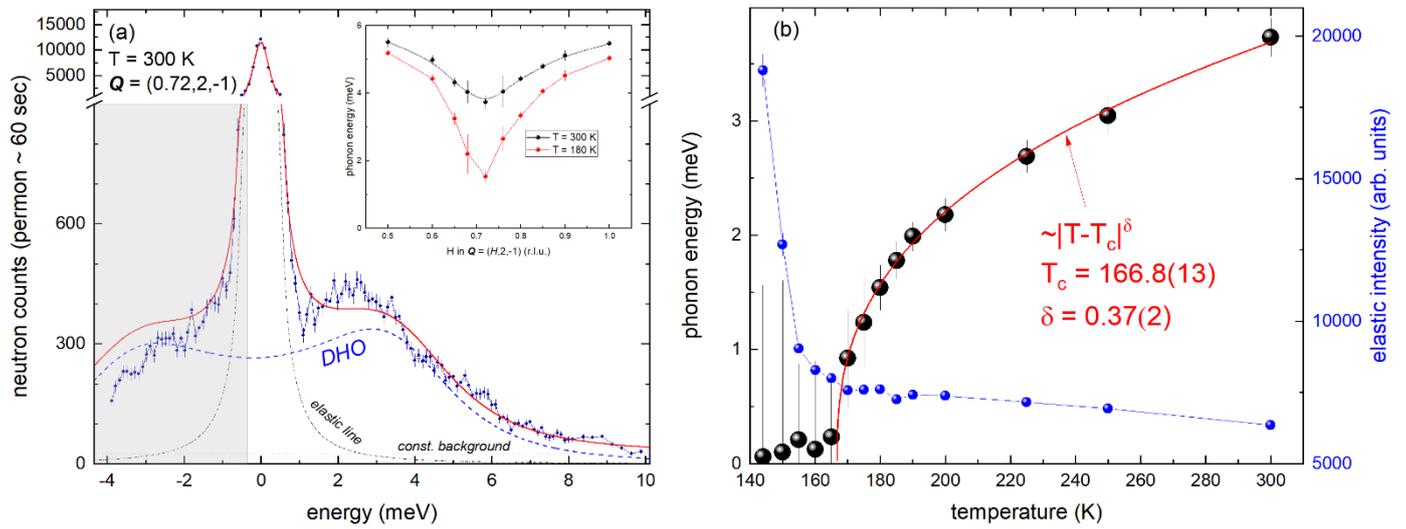
**Fig. 2. Raw data and preliminary analysis.** (a) Raw data from inelastic neutron scattering at $Q$ = (0.72,2,-1) and T = 300 K. The data were approximated for energy transfers of E $\geq$ -0.3 meV (outside the grey box) with a pseudo-Voigt function for the elastic line and a damped harmonic oscillator (DHO) for the soft phonon mode. The approximated functions were plotted over the whole energy range (see more detailed discussion in the text). The inset shows the dispersion of the soft phonon mode along $Q$ = ($H$, 2, -1), $0.5 \leq H \leq 1$, for T = 180 K and 300 K. (b) Energy of the soft phonon mode (large spheres, left-hand scale) and the elastic intensity as function of temperature (small spheres, right-hand scale). The red line denotes a power law fit of the form $|T-T_c|^\delta$ with the fit parameters given in the panel.

Figure 1(a) shows a preliminary analysis of the data taken at the CDW wave vector, $q_{CDW}$ = (0.28,0,0), in the Brillouin zone adjacent to $\tau$ = (1,2,-1), i.e., $Q$ = (0.72,2,1). **The here presented analysis is preliminary because it is not yet convoluted with the experimental resolution and therefore could only be done considering scattering at positive energy transfers.** The resolution convolution is necessary as can be seen in Figure 1(a). The DHO function approximated for the positive energy transfers, i.e., energy-loss neutron scattering, does not describe well the observed intensities at the negative energy transfers. The main reason is that the resolution in the latter case is better since the incident energy is smaller than for energy-loss scattering. This work is in progress.

Nevertheless, the preliminary results show anomalous phonon dispersion in agreement with published data and the softening of the phonon on cooling. A power-law fit to the softened but finite phonon energies for T $\geq$ 170 K yields $T_c$ = 167 K and an exponent $\delta$ = 0.37. It seems that the intensity of the resolution-limited elastic scattering is already increasing below 160 K. Thus, there seems to be a temperature region characterized by a fully-softened phonon mode without an static superlattice peak. However, the range of this tempearture region is smaller than previously reported.

1    Souliou, S. M., Lacmann, T., Heid, R., Meingast, C., Frachet, M., Paolasini, L., Haghighirad, A. A., Merz, M., Bosak, A. & Le Tacon, M. Soft-Phonon and Charge-Density-Wave Formation in Nematic BaNi$_2$As$_2$. *Physical Review Letters* **129**, 247602, doi:10.1103/PhysRevLett.129.247602 (2022).

2    Song, Y., Wu, S., Chen, X., He, Y., Uchiyama, H., Li, B., Cao, S., Guo, J., Cao, G. & Birgeneau, R. Phonon softening and slowing-down of charge density wave fluctuations in BaNi$_2$As$_2$. *Physical Review B* **107**, L041113, doi:10.1103/PhysRevB.107.L041113 (2023).

3    Meingast, C., Shukla, A., Wang, L., Heid, R., Hardy, F., Frachet, M., Willa, K., Lacmann, T., Le Tacon, M., Merz, M., Haghighirad, A.-A. & Wolf, T. Charge density wave transitions, soft phonon, and possible electronic nematicity in BaNi$_2$(As$_{1-x}$P$_x$)$_2$. *Physical Review B* **106**, 144507, doi:10.1103/PhysRevB.106.144507 (2022).