Experimental report

25/01/2024

Proposal:	**CRG-3006**	**Council:**	4/2023
Title:	Magnetic structure and excitationsin long-range oxygen ordered $Pr_2NiO_{4.25}$		
Research area:	Physics		

This proposal is a new proposal

Main proposer:	**Werner PAULUS**
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Local contacts:	Wolfgang F SCHMIDT

Samples: $Pr_2NiO_{4.25}$
La_2CoO_{4+d} (Sample from TEST-3205)

Instrument	Requested days	Allocated days	From	To
IN12	7	8	19/06/2023	27/06/2023

Abstract:

We want to compare magnetic ordering and excitations in oxygen doped $Pr_2NiO_{4.25}$ with the isoelectronic $Pr_{1.5}Sr_{0.5}NiO_4$ phase. The oxygen doped phase shows translational periodicities of more than 100 Å, while $Pr_{1.5}Sr_{0.5}NiO_4$ adopts the 5.5 x 5.5 x 12.4 Å3 K_2NiF_4 type unit cell. While the magnetic ordering is essentially 2D for Sr-doped phases, O-doping results into a significantly stronger coupling along the c-axis, related to the presence of interstitial oxygen, which is supposed to be part of the magnetic exchange pathway. The stronger magnetic exchange interactions in case of O-doping is also indicated by a doubling of the c-axis below 100K, while remaining invariant for the Sr-doped phase. We focus to explore dispersions perpendicular to the 2D layers, i.e. along [001], which has been essentially neglected so far. We want to study the hierarchy of different degrees of freedom as oxygen together with electronic ordering phenomena, i.e. magnetic and charge ordering

Magnetic structure and excitations in long-range oxygen ordered Pr2NiO4.25 (CRG-3006)

(**proposer:** W. Paulus, M. Ceretti, M. Meven, A. Piovano, W. Schmidt, **local contact:** W. Schmidt)

The presence of charge and spin ordering phenomena in cation-doped nickelates result in a complex structural and electronic phase diagram. $Pr_2NiO_{4+\delta}$ ($PNO_{4+\delta}$) combines large oxygen doping (δ_{max}=0.25 ≡ 0.5 holes) with high oxygen ion conductivity down to ambient temperature, which also allows for oxygen (re-)ordering even at RT. In this context, long-range oxygen order adds an additional degree of freedom on top of charge-, spin, and orbital ordering. Compounds doped with interstitial oxygen show long range modulations in the scattering pattern. The interstitial oxygens affect structural and electronic interlayer coupling including a significantly stronger coupling along the *c* axis. This makes them a valuable tool to better understand correlations between the structural and electronic phase diagram. In this part of our study, we want to look into magnetic ordering and excitations in oxygen doped $PNO_{4.23}$. In spite of its structural neighbourhood to $PNO_{4.25}$ we found recently, that for this composition already a slight thermal treatment introduces a change of the local structure of the sample towards the $PNO_{4.25}$ setting arrangement without introducing additional oxygen, that directly and strongly affects the magnetic order.

Measurements. We used both the unpolarized and polarized setups of IN12 in order to look for details of the magnetic structure and excitations of $PNO_{4.23}$. Fig. 1.a) shows the inelastic scans of the non-annealed $PNO_{4.23}$ performed on IN12 in the unpolarized mode of IN12 along H (left) and along L (right) at about 1.5 K. A repetition of the Q-scans along H for the annealed sample did not reveal noticeable changes (Fig. 1.b)).

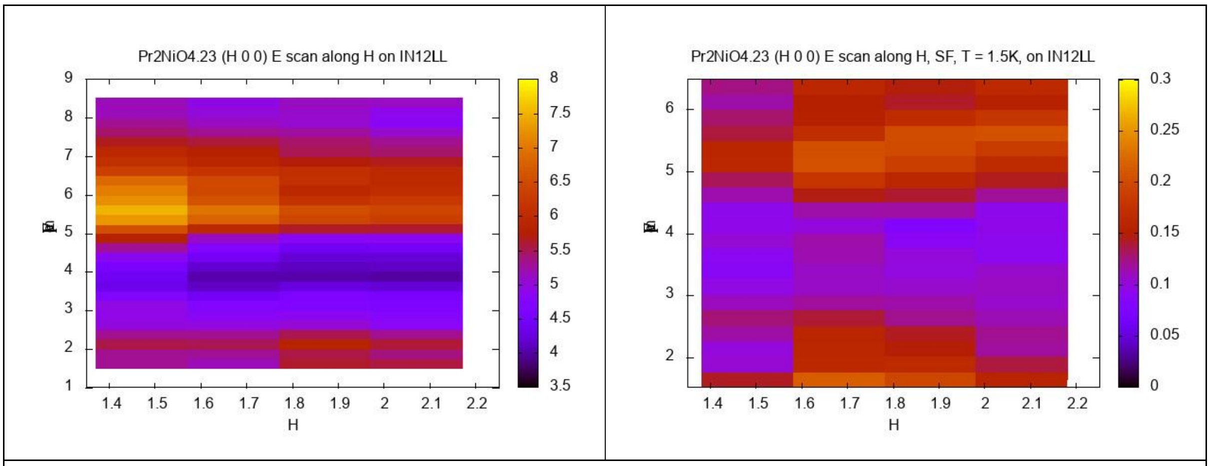

Fig. 1: (a): Q-scans of the non annealed $Pr_2NiO_{4.23}$ (left) and (b) of the annealed $Pr_2NiO_{4.23}$ (right, spin flip mode, the non spin flip mode did not reveal any significant intensities)

Furthermore, we performed elastic scans on selected reflections along H and along L respectively at different temperatures on the sample to tickle the aforementioned potential influence from thermal treatment.

In a first step, we took data of the non-annealed state (10 K steps, unpolarized neutrons). The incommensurate distribution of reflections and the transition temperatures around 160 K and around 40 K are very close to the ones known for $PNO_{4.25}$ (Fig. 2). Especially, the non-annealed $PNO_{4.23}$ sample confirms the doubling of the *c* axis in terms of magnetic order found for $PNO_{4.25}$ (experimental report 4-01-1800).

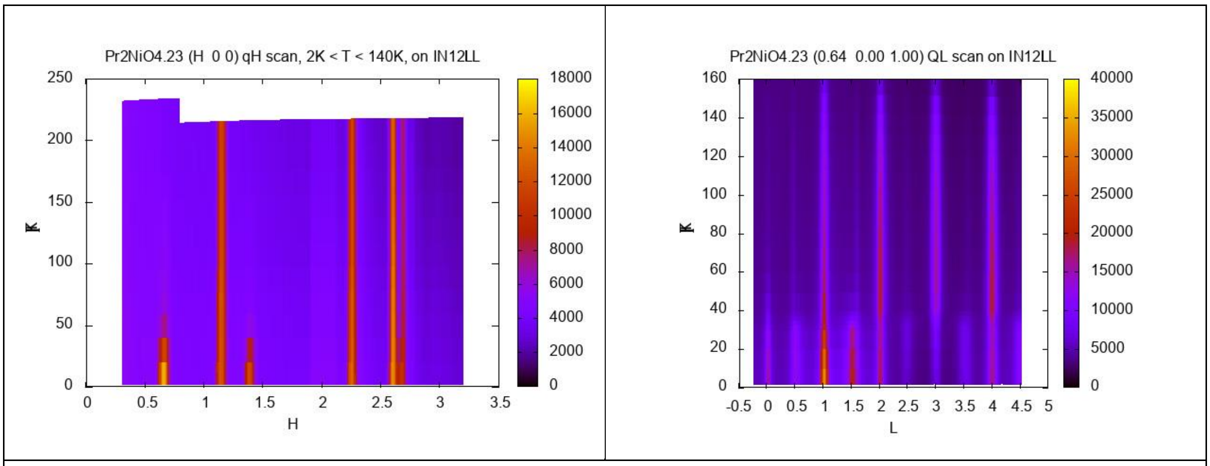

Fig. 2: Elastic H scan (left) and L scan (right) of non-annealed $Pr_2NiO_{4.23}$

In a second step, we took data along the same directions in the annealed state (40 K steps due to limited time, polarized neutrons) in order to separate unambiguously the nuclear and magnetic contributions, especially along the L direction and compared them with the data from the non-annealed sample. The elastic scans along H do not reveal any significant differences (Fig. 3, left). But the L scans show a completely different picture where the magnetic doubling of the *c* axis (L= n + ½, n = 0, 1, 2, etc.) vanishes almost completely already at T=1.5 K. Weak remains of the magnetic L = ½ contribution from the non-annealed state might be attributed to a non perfect transformation of the large sample.

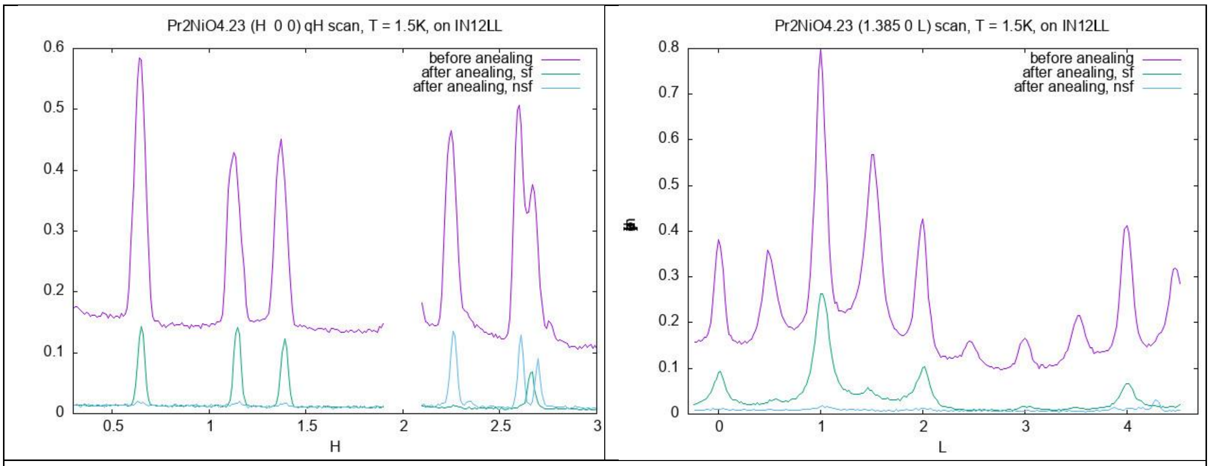

Fig. 3: Elastic scans along H and L of the both the non-annealed and annealed $Pr_2NiO_{4.23}$ sample at 1.5 K (intensities in arbitrary units).

The complete evaluation of the data at IN12 has not yet been completed. Nevertheless, The preliminary results shown here confirm a dramatic change in the magnetic structure, which raises new questions about its cause and which we believe should be addressed in further studies.