Experimental report

25/01/2024

Proposal: 4-01-1800 **Council:** 4/2023

Title: Magnetic structure and excitationsin long-range oxygen ordered Pr2NiO4.25

Research area: Materials

This proposal is a new proposal

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Samples: Pr2NiO4.25
Pr1.5Sr0.5NiO4

Instrument	Requested days	Allocated days	From	To
IN22	7	0		
IN12	7	0		
IN20	7	7	20/11/2023	27/11/2023
ORIENTEXPRESS	1	1	16/11/2023	17/11/2023

Abstract:

We want to compare magnetic ordering and excitations in oxygen doped Pr2NiO4.25 with the isoelectronic Pr1.5Sr0.5NiO4 phase. The oxygen doped phase shows translational periodicities of more than 100 Å, while Pr1.5Sr0.5NiO4 adopts the 5.5 x 5.5 x 12.4 Å3 K2NiF4 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

4-01-1800. Magnetic structure and excitations in long-range oxygen ordered Pr2NiO4.25

The presence of charge and spin ordering phenomena in cation-doped nickelates, i.e. La2-xSrxNiO4, yields to a complex phase diagram, structurally and electronically [1]. Depending on the hole-doping concentration, the charges on the TM-sites tend to segregate into stripes for low hole doping, whereas 2D-checkerboard ordering is observed for higher doping concentrations. Pr2NiO4+d presents an interesting case, as it combines large oxygen doping (dmax.=0.25 º 0.5 holes) with high oxygen ion conductivity already at ambient temperature [2-6]. This is also the reason why oxygen ordering can proceed still down to RT. Hole doping achieved by oxygen insertion has been shown to be electronically fully equivalent to cation substitution, but long-range oxygen order is added here on top of charge-, spin, and orbital ordering, as an additional degree of freedom. In this study, we want to compare magnetic ordering and excitations in oxygen doped Pr2NiO4.25 with the isoelectronic Pr1.5Sr0.5NiO4 phase. Thereby the oxygen doped phase shows translational periodicities more than 100 Å [6-7], while Pr1.5Sr0.5NiO4 adopts the small 5.5 x 5.5 x 12.4 Å3 K2NiF4 type unit cell. Exploring the influence of the interstitial oxygen atoms to enhance structural and electronic interlayer coupling is important to better understand correlations between the structural and electronic phase diagram. 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.

Measurements. We want to compare changes in the magnetic structure and excitations for the $Pr_2NiO_{4.25}$ with respect to magnetic dispersions along [001].

The (h00) elastic scan in Figure 1a show the presence of several incommensurate peaks associated to the presence of over-stoichiometric interstitial oxygen. The scan along l direction though the 1.33 incommensurate peak at low T (1.5K) in Figure 1b evidence peaks at both integer and half integer (black arrows) positions. Increasing the temperature above Tn bring to a gradual suppression of mostof the intensity revealing their predominantly magnetic origin. In particular half-integer intensity is completely suppressed at 300K. Stronger magnetic exchange interactions in case of O-doping are then associated with the doubling of the c-axis (while remains invariant for the Sr-doped phase). The doubling of the magnetic c-axis sets in below 120 K. One main aspect here is to understand up to which degree, hierarchical ordering phenomena are correlated. In this regard we want to study how long-range oxygen ordering allows to direct/induce electronic ordering phenomena, i.e. the spin structure and related dynamics. The T dependence of the magnetic intensity at 1.33 0 1 point presented in Figure 2b show a complex behaviour with several changes of slope connected to different magnetic effect. In particular changes seen at about 30 and 50 K might be associated with a reorientation of spins, while the suppression of the magnetic signal gradually disappear in between 50 and 210K. The spin dynamics associated to this magnetic order is presented in Figure 2c in its low energy region. The propagation of a dispersive spin wave is evident.

Analysis of the whole set of data is under way and will bring more insight to the spin wave dispersion in l direction.

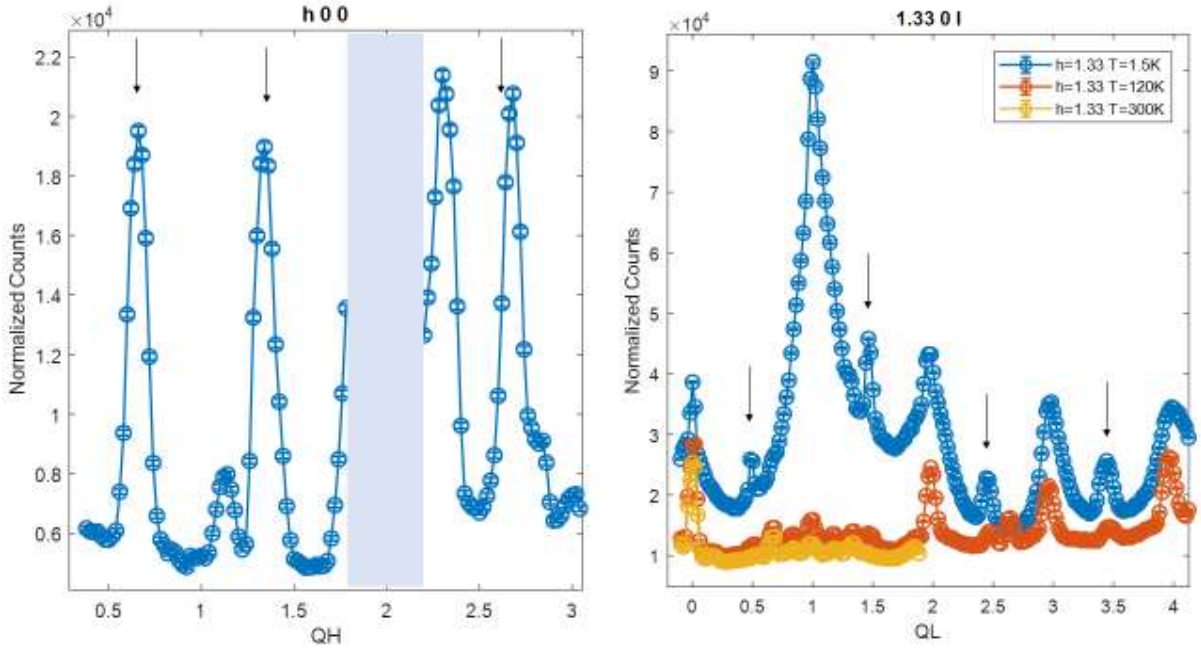

Figure 1. a) elastic scan along (h 0 0) direction showing the incommensurate peaks (black arrows). B) elastic scans in function of temperature along (1.33 0 l) showing the magnetic origin of most of the peaks, including peaks at half integer positions (black arrows) showing the doubling of the magnetic unit-cell.

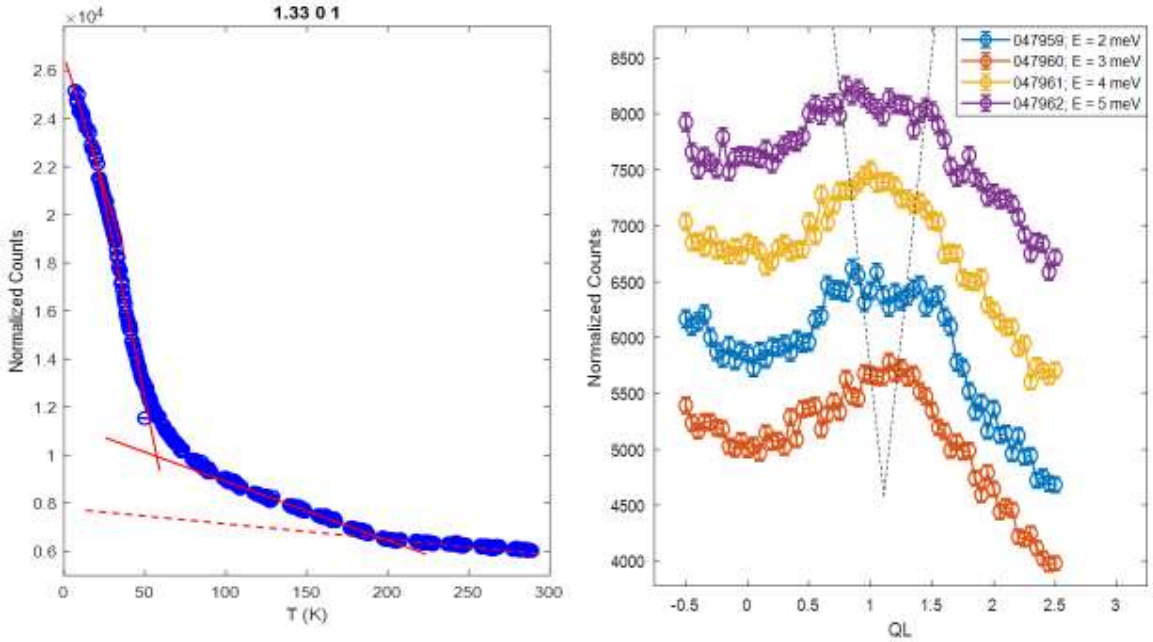

Figure 2. a) elastic intensity of magnetic Bragg peak 1.33 0 1 in function of temperature. Different transitions could be separated at 30K, 50K and 210K where finally magnetic contribution disappear. b) Inelastic scans along l around 1.33 0 1 point showing an evident spin wave dispersion.