

Experimental report

28/06/2023

Proposal: 4-03-1759

Council: 10/2022

Title: Temperature dependence of the spin-phonon coupling in AFM transition-metal mono-oxide

Research area: Physics

This proposal is a new proposal

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Samples: NiO

Instrument	Requested days	Allocated days	From	To
ORIENTEXPRESS	1	1	20/05/2023	21/05/2023
IN8	8	7	22/05/2023	30/05/2023

Abstract:

We propose to investigate the dynamics of the spin-phonon coupling in NiO focusing on the crossing points of phonon and magnon dispersion. The relatively simple structure allows a precise modelling of both phonons and magnons via ab-initio (phonons) and model calculations (magnons), respectively, which show good agreement with data from a recent time-of-flight experiment performed on ARCS, ORNL. The TOF data identify the wave vectors, where the magnons and phonons are simultaneously visible. We will study the properties of the dispersing branches as a function of temperature across TN.

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Temperature dependence of the spin-phonon coupling in AFM transition-metal mono-oxide

Motivation

We proposed to study the lattice and magnetic degrees of freedom and their interplay in transition-metal mono-oxide NiO. NiO is considered a prototypical antiferromagnetic insulator [$T_N \approx 523$ K], and was intensively investigated due to its simple structure, spin dynamics but also strong electronic correlations. The relatively simple structure (one chemical unit per cubic unit cell) allows a precise modelling of both phonons and magnons via *ab-initio* and model calculations, respectively. This modeling provides the vital guideline for the proposed TAS experiment at IN8, in which we investigated the wave vectors, where the magnon and phonons dispersions are simultaneously visible, as a function of temperature across T_N .

Technical details

The high neutron flux on IN8 provides the ideal setup to acquire a detailed view of phonon and magnon dispersions. To obtain a reasonably good resolution at the relatively high energy transfers, $10 \text{ meV} \leq E \leq 80 \text{ meV}$, we used double-focusing cooper (Cu 200) monochromator and Cu200 analyzer with the final energy at the analyzer fixed to 14.7 meV. Measurements were performed at $T = 300$ K, and in smaller steps across $T_N = 523$ K up to 650 K. To achieve this temperature range, we used the furnace (ambient to 1100 C°). The reactor power was reduced to 48 MW.

Experiment details

A high-quality single crystal of NiO was grown by floating-zone method in the Department of Physics at the University of Oxford, courtesy of D. Prabhakaran (space group $Fm\bar{3}m$, $a = 4.178 \text{ \AA}$). The crystal is about 4 cm long (diameter of 9 mm) and weighs 5 g. The preliminary work with TOF confirmed the high quality.

On IN8 we performed the measurements in the range from 10 meV to 80 meV to investigate the phonon and magnon dispersion at the L -point (cubic notation) $Q_1 = (2.5, 2.5, 2.5)$, $Q_2 = (2.5, 2.5, 1.5)$, $Q_3 = (1.5, 1.5, 3.5)$ along the T_N . In addition, we have measured $Q_4 = (1, 1, 3.5)$, $Q_5 = (2.25, 2.25, 2.25)$, and, finally, $Q_6 = (1, 1, 2)$ as a reference point.

Later, we measured several wave vectors along $[111]$ direction, such as $Q = (H, H, H)$ from $H = 2.5$ to $H = 3.0$ at 297, 525 and 900 K to map the phonon dispersion. The steep magnon dispersion was mapped by constant energy scans at 0 and 50 meV.

Observed results

We have performed preliminary data analysis. Here we present some of the results. Fig. 1 shows the magnon peak as a function of temperature. The observed jump on the data is related to the plexi glass, which was taken out at higher temperatures due to the signal weakening. The data were fitted to the Gauss curve and the parameters were extracted. This data set allows us to experimentally identify the transition temperature giving us $T_N = 525$ K.

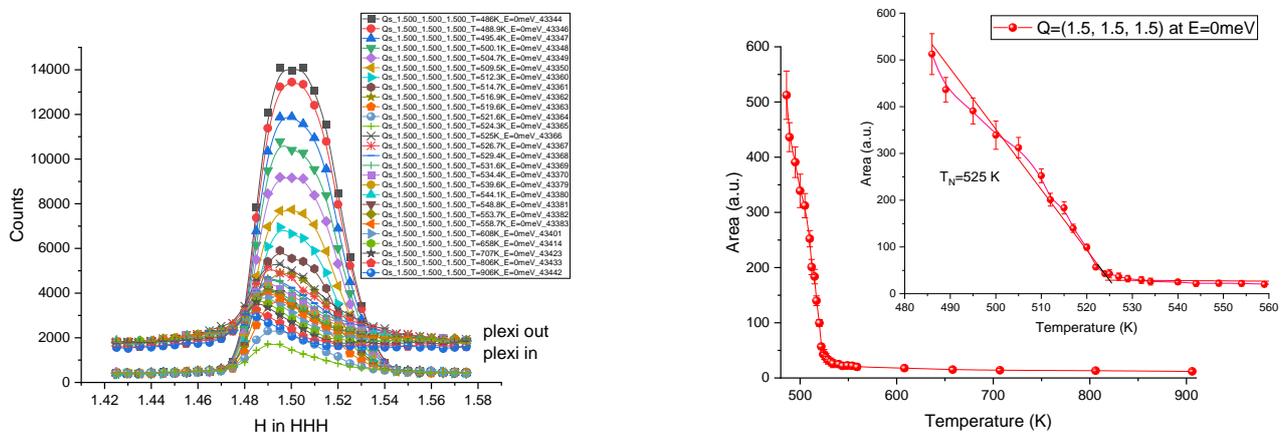


Fig.1. (left) Observed neutron scattering intensities of the magnon at various temperatures through the transition temperature T_N (the jump in the data is related to plexi being in or out). (right) The result of the fitted raw data presented as an area of the magnon peak as a function of temperature. We observe the largest change in the behavior at $T_N = 525$ K.

Investigation of phonon dispersion at $Q = (2.5, 2.5, 2.5)$ is shown in Fig. 2, where the raw data at room temperature is plotted. We can see that the background of the raw curve is relatively high. This is because at this Q point, we are exactly at the magnetic peak. Following the phonon energies, we extracted the temperature dependence of two phonon modes. We observe the softening of both phonon modes. However, there is a slight difference between 40 and 67 meV phonon modes: the high energy phonon softens stronger and then stabilizes. Also, the linewidths of the 67 meV mode are increasing dramatically with temperature.

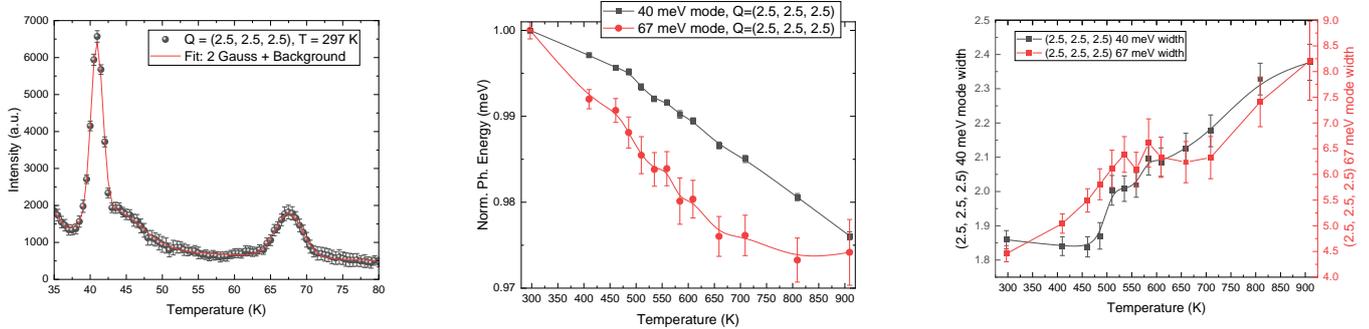


Fig. 2. (left) Raw data at $Q = (2.5, 2.5, 2.5)$ taken at room temperature. We observe two phonon modes: 40 and 67 meV. (middle) These two phonon modes as a function of temperature. (right) The linewidths of the phonon energies.

Additional analysis is required. Observed phonon dispersions will be compared to *ab-initio* lattice dynamical calculations performed in theory group of our institute. Detailed temperatures dependencies across the magnetic phase transition will allow us to verify the correlation between the observed effects and the presence of magnons and magnetic order in NiO.