Experimental report

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Council: 10/2022

Title: Probing the k-dispersion of the thermal fluctuation spectrum near theantiferromagnetic phase transitionof NiO

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Main proposer: **Andrea GAUZZI**

Experimental team: Andrea GAUZZI

Local contacts: Iurii KIBALIN
Bachir OULADDIAF
Nebil AYAPE KATCHO

Samples: NiO

Instrument	Requested days	Allocated days	From	To
D10	7	3	29/08/2023	04/09/2023
ORIENTEXPRESS	1	1	28/08/2023	29/08/2023

Abstract:

In the present experiment, we aim at measuring directly the thermal fluctuation spectrum of the order parameter in the vicinity of a second-order phase transition by using single-crystal neutron diffraction. As a model system, we propose to study the antiferromagnetic (AFM) transition of NiO, owing to the simplicity of its nuclear and magnetic structures. The above spectrum will be measured by measuring precisely the q-dependence of the diffuse scattering intensity across the AFM Bragg peak at (1/2 1/2 1/2) along the [100], [110] and [100] directions of the cubic symmetry and at different temperatures in the thermal fluctuation region. The knowledge of the spectrum shall enable us to unveil the exact functional form of the anisotropic exchange interaction and thus to verify the deviation of the effective Hamiltonian driving the transition from simplified model Hamiltonians like Ising, Heisenberg and 3DXY models.

Experimental report #5-53-313: Probing the k-dispersion of the thermal fluctuation spectrum near the antiferromagnetic phase transition of NiO

Scientific objectives and expected results. The aim of the experiment was a direct measurement of the thermal fluctuation spectrum of paramagnons above the antiferromagnetic transition (AFM) of NiO using single-crystal neutron diffraction. The motivation underlying the experiment is the existence of an intimate link between the thermodynamic fluctuation spectrum of the order parameter in the vicinity of a second order phase transition and the effective Hamiltonian of the transition. Thus, the experimental determination of the spectrum is expected to unveil the effective interaction driving the transition, which may elucidate the microscopic mechanism in complex systems where several interactions compete. As a model system, in the present experiment we have studied NiO, owing to the simplicity of its nuclear and magnetic structures. In practical terms, our plan was to probe the thermal fluctuation spectrum of paramagnons by measuring precisely the **k**-dependence of the diffuse scattering (DS) intensity across the AFM Bragg peak at (1/2 1/2 1/2) along the (100), (110) and (100) directions of the cubic symmetry and at different temperatures in the fluctuation region above T_N-530 K. The knowledge of the spectrum shall enable us to unveil the exact functional form of the anisotropic exchange interaction and thus to verify the deviation of the effective Hamiltonian of the transition from simplified model Hamiltonians, such as Ising, Heisenberg and 3dXY models.

Sample measured. Using the Orient Express Laue diffractometer, we first screened the quality of three single crystals. Based on the results of these measurements, we selected a commercial platelet-like single crystal of 3 mm $\times$ 3 mm size and 1 mm thickness oriented toward the (100) direction, which made the identification of the most intense (200) nuclear peak easier using the 2D detector.

Measurements performed. The first day was devoted to the mounting of the sample and of the furnace, its alignement and the determination of the UB matrix for the 2D detector. We then set the sample temperature at 500 K, well below the AFM ordering temperature, $T_N = 530$ K, in order to check the $(\frac{\bar{1}}{2},\frac{\bar{1}}{2},\frac{\bar{1}}{2})$ magnetic Bragg peak and to establish the acquisition time required to obtain a satisfactory statistics. We then studied the evolution of the intensity of the peak with temperature by warming up the sample up to 540 K, where the intensity became comparable with that of the background for acquisition times of 2'/point. At this stage, we were ready to launch a first series of q-scans along the (111) symmetry direction at various temperatures across T_N, namely 524, 526, 528, 532, 534 and 536 K. During the night, a communication problem occurred between the temperature controller and the NOMAD remote control system. As a result, the command to change the setpoint temperature was not received when the temperature was 528 K. In the morning, we had therefore to resume the measurement. In the second evening, we launched a second series of scans along the remaining symmetry directions in order to probe the anisotropy of the DS intensity. Again, the same communication error occurred during the night, which required to resume again the measurement. The following days, we repeated some scans above T_N in order to improve the statistics. Indeed, as mentioned below, already the first data set exhibits an indication of oscillatory behaviour of the DS intensity, which is unexpected. In the late morning of the fourth day, we decided to move to the analyser configuration with the aim of collecting similar q-scans as before with minimised background noise and free from quasi-elastic contributions. After having recalculated the UB matrix, the new series of q-scans using the analyser was then launched in the evening to last until the end of the available time. In the following morning, we decided to open the collimator in order to enhance the signal-to-noise ratio. Because of time limitation and the longer acquisition time required by the analyser, we could measure only four scans at 524, 528, 532, and 536 K and repeat the last two scans twice in order to further improve the statistics. In the remaining hours available, we measured a few points of a last q-scan along the (111) direction at 540 K.

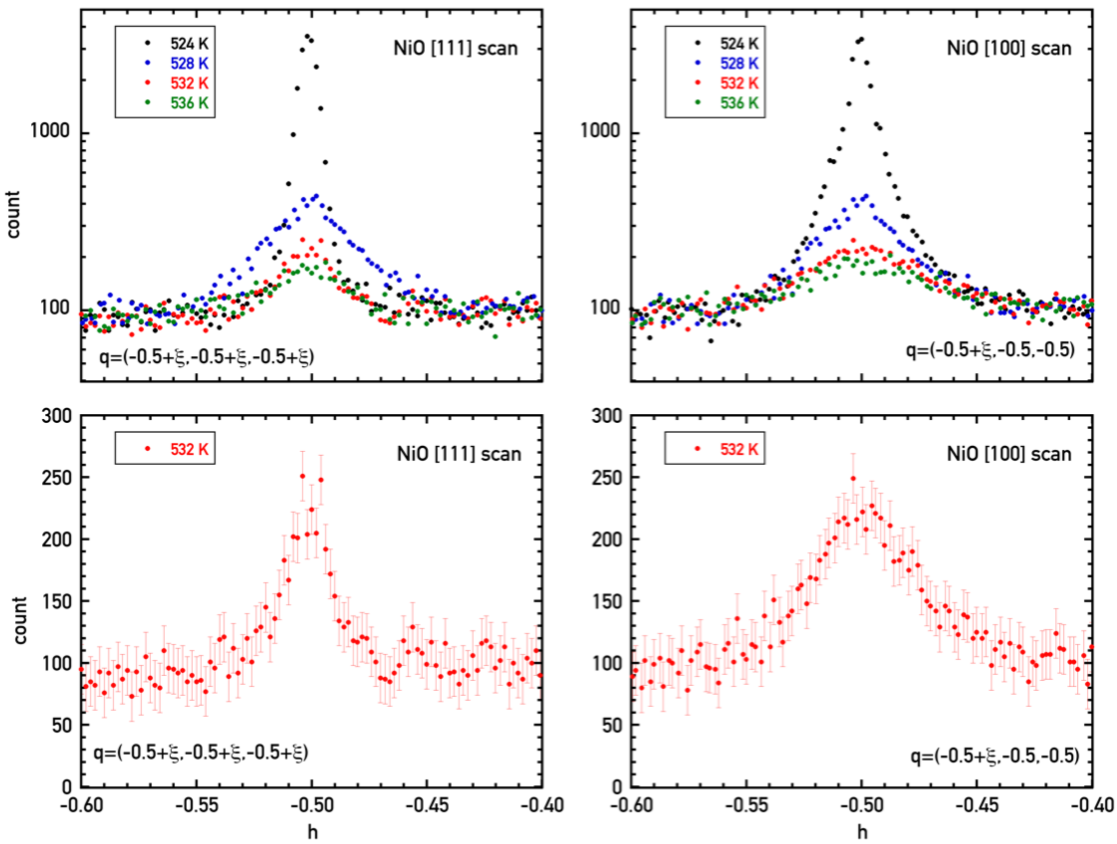

Figure 1: Representative q-scans of the diffuse scattering of NiO across the $(\frac{\bar{1}}{2},\frac{\bar{1}}{2},\frac{\bar{1}}{2})$ magnetic Bragg peak along the (111) (left) and (100) (right) symmetry directions. Top panel: temperature evolution across $T_N = 530$ K. Bottom: detail of the scans slightly above T_N, at 532 K.

Results and preliminary analysis. Owing to the short time passed after the end of the experiment, we limit ourselves to show a few data plots and to outline a preliminary analysis of the results. In Fig. 1, we plot representative scans measured using the analyser taken at the four temperatures measured, 524, 528, 532, and 536 K, along the (111) and (100) directions and also a detail of the scans at 532 K, the first point measured above T_N=530 K. As expected, one first notes a rapid decrease of the peak intensity but also a marked anisotropy of the peak shape and width and a sizable increase of the peak width above T_N. Second, the scan at 532 K along the (111) directions exhibits a clear indication of oscillating behaviour, which is very surprising for it suggests an unconventional fluctuation spectrum of the paramagnons arising from nonuniform spin-spin correlations in the nm scale.

Perspectives and future experiments. We consider that the results obtained in this first diffraction experiment - especially the data taken using the analyser - are very promising for the data quality is sufficiently good to determine rather precisely the momentum dependence of the zero-energy thermodynamic fluctuation spectrum of paramagnons. To do so, we should first know the analyser transfer function in order to deconvolute the peaks from the instrumental broadening. As already envisaged at the moment where the proposal was submitted, a continuation of the experiment using inelastic neutron scattering on the same D10+ instrument is also necessary to determine the full energy- and momentum dependence of the fluctuation spectrum. This is particularly important in light of the unexpected observation of oscillating behaviour of the DS intensity, which prompts us to carry out more elastic and inelastic measurements with improved statistics in order to confirm this observation and, in the positive, to determine the wave vector and temperature dependence of the oscillations.