



	Experiment title: Freezing of low energy electronic excitations at the Verwey transition of magnetite	Experiment number: HC2404
Beamline: ID32	Date of experiment: from: 6 July 2016 to: 12 July 2016	Date of report: 9 September 2016
Shifts: 18	Local contact(s): Flora Yakhou-Harris	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): Dr. Christian Schüßler-Langeheine*, Dr. Piter Miedema*, Prof. Alexander Föhlisch, Helmholtz-Zentrum Berlin, Germany, Dr. Justine Schlappa*, European XFEL GmbH, Hamburg, Germany, Dr. Martin Beye*, DESY, Hamburg, Germany		

Report:

(Preliminary) The experiment addressed the energetics of electronic excitations when freezing into static electronic order. Analogously to phonon mode softening upon formation of structural order, electronic excitations should soften upon formation of electronic order. As an example we chose the famous Verwey transition that occurs in magnetite (Fe_3O_4) around 122.8 K. This transition is known to involve changes in structural order parameters as well as orbital and potentially charge order.

With the advanced RIXS-instrumentation available at ID32 at ESRF, we chiefly aimed at studying a characteristic 250 meV loss feature in the vicinity of the $(00^{1/2})$ orbital ordering wave vector characteristic for the low-temperature phase (notation refers to the cubic high-temperature phase). Analogously to the situation for phonons, we only expect a mode softening around the order vector of the newly formed phase. The 250-meV feature occurs in the Fe L_3 -edge RIXS spectrum and has tentatively been assigned to an intra- t_{2g} orbital excitation, while other interpretations have been brought forward as well. With the overall resolution set to about 65 meV we were able to observe additional features. In particular a feature around 70 meV energy loss was just visible as a shoulder to the elastic line. Its character is still to be identified.

In the low-temperature phase and with the spectrometer centered on the orbital order Bragg peak, the detector saturated completely through elastic scattering. To mitigate this problem and to be able to measure the weak energy loss features, we moved to the direct vicinity of the Bragg peak at about $q=(0.02,0,0.5)$. We were thus able to discern inelastic features from the Bragg peak without too much reduction of the softening to be expected.

In this geometry when ramping the temperature up and down across the Verwey transition, we consistently observe an energetic shift of the loss feature by about 20 meV towards lower energies (see a sample dataset and preliminary analysis in Fig. 1; a more careful data analysis is ongoing). The energetic shift of the feature is found to occur exactly at the temperature, where the elastic scattering has already lost most of its intensity and only critical fluctuations of the order contribute to the elastic scattering. This is exactly the temperature region where a softening of this excitation mode was expected.

For comparison we also recorded spectra at $(1/8 0 1/2)$ far away from the orbital order peak. At this position in momentum space the same loss features can be found, but no energy shift was observed (Fig. 1).

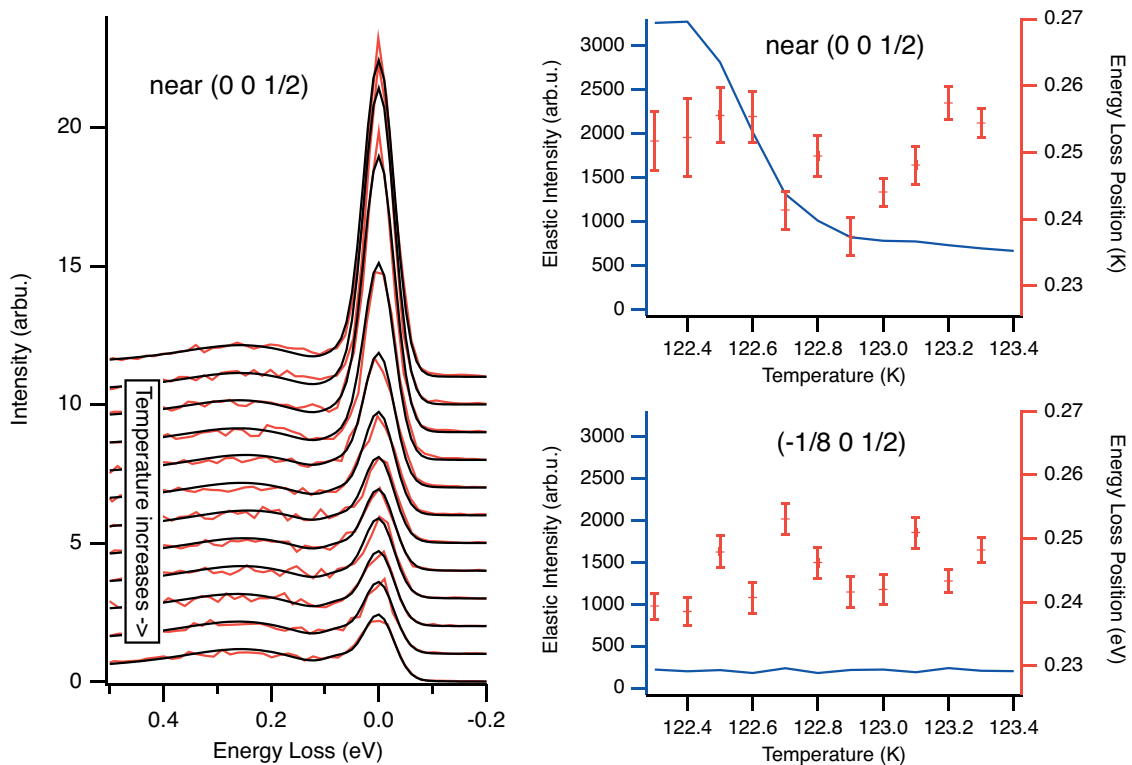


Figure 1: Left: Series of spectra measured close to the orbital order peak position; the loss in elastic intensity upon heating through the Verwey transition is clearly observed. Right top: energy of the characteristic loss feature (red) and elastic peak intensity (blue) extracted from the spectra. Right bottom: results from a data set recorded far away from the orbital order peak.