ESRF

Experiment title:

**Mechanism of the Vervey transition in magnetite**

Experiment number:

**HS-3754**

**Beamline:**

**Date of experiment:**
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**Shifts:**
15

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**Report:** A recent group theoretic analysis of the Verwey transition in magnetite has identified a combination of order parameters of symmetry $X_3$ and $\Delta_5$ as compatible with the change of lattice from the high temperature cubic phase to the monoclinic structure below the transition $T_V$ = 122 K [1]. Phonon calculations and test experiments showed that the lowest $X_3$ (optical) and $\Delta_5$ as well as $X_4$ (acoustic transverse branch) scatter strongly at or near the $X$-points (445) and (554), respectively. Figure 1 shows spectra at (554) of the X4 and at (445) of the $X_3$ with a slight contribution of $X_4$ at energy resolution of 1.8 meV.

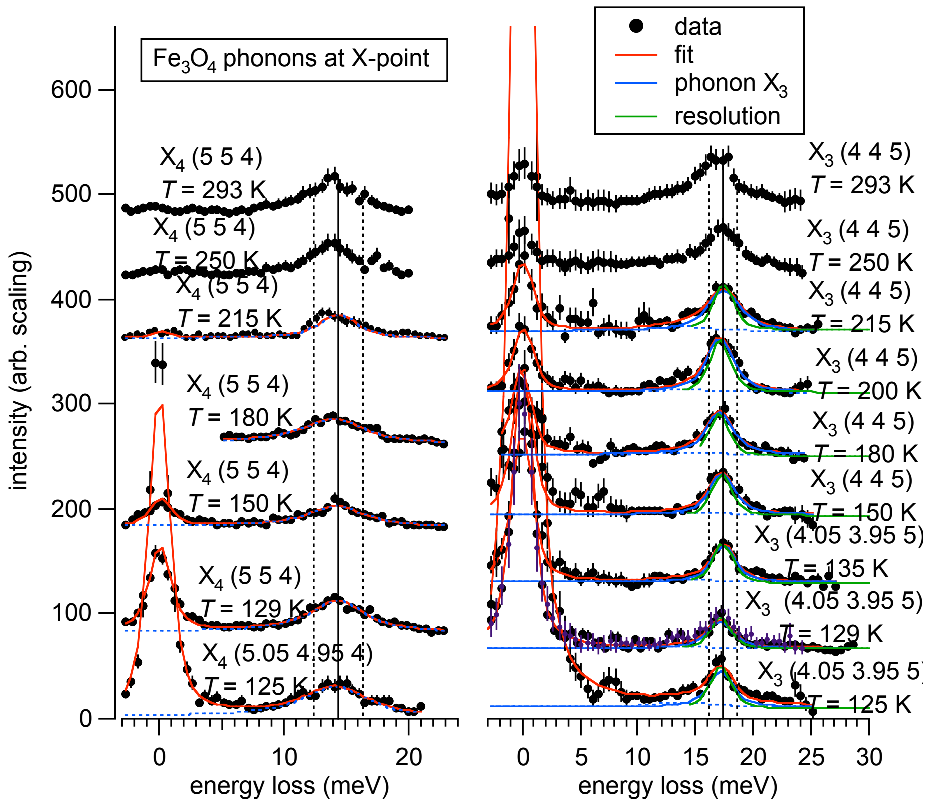


**Fig. 1:** Inelastic x-ray scattering scattering spectra at various temperatures above the Verwey transition $T_V$ = 122 K at (554) showing the $X_4$ phonon and (554) showing the $X_3$ phonon and a slight contribution from $X_4$. The (11 11 11) mode of the beamline with 1.8 meV resolution was chosen due to the unavoidable overlap of the two phonons in the spectra.

The raw data in Fig.1 reveal on inspection (i) the phonon $X_4$ is serverely broadened at all temperatures, (ii) the $X_3$ phonon has a nearly resolution limited width, although at high temperatures a slight broadening is visible, (iii) the elastic intensity (zero energy peak) rises strongly on approaching the transition. The rise of elastic intensity necessitates to measure the lowest temperature points not at but near the fully high symmetric positions, as indicated in Fig.1. Below the transition a shift of the peaks of $X_3$ (softening) and $X_4$ (hardening) is seen. The change of lattice into the monoclinic phase, however, makes it impossible to assign this shift to an electronic or lattice symmetry effect and to discuss its origin.

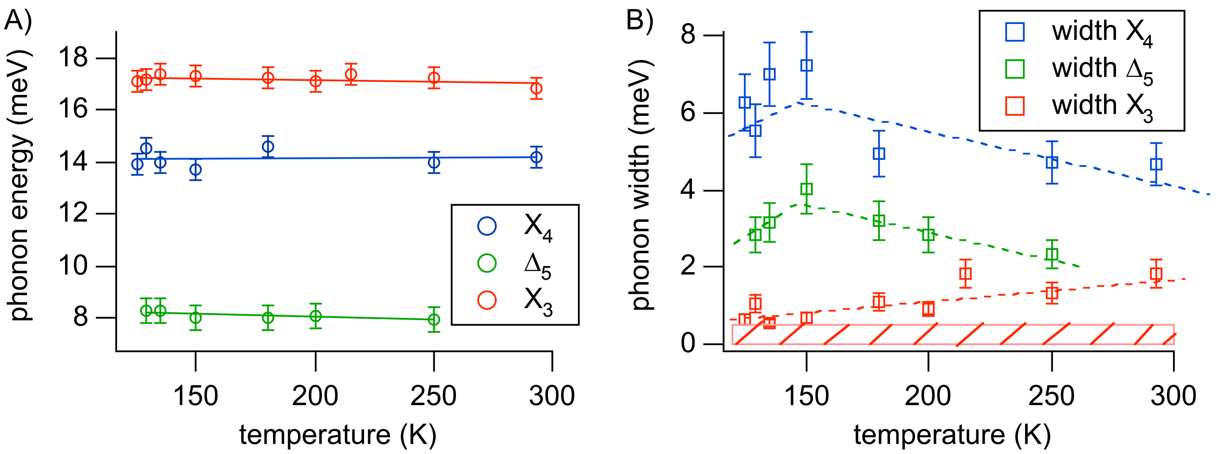


**Fig. 2:** Results of a peak fitting analysis of the phonons $X_4$, $X_3$, and $\Delta_5$ as a function of temperature above $T_V$ = 122 K: A) peak positions and B) peak widths. The shaded area close to zero peak width indicates the uncertainty region, where a resolution limited model fits as well as a model with broadening of the phonon. Dashed lines serve as a guide to the eye.

Figure 2 shows the results of a peak fitting analysis using Lorentzian peaks to represent the phonons convoluted with the measured resolution function. The temperature dependence of the phonon energy shows a slight hardening of $\Delta_5$ at lower temperature that may be compatible with the increased elasticity in the contracted lattice. $X_3$ and $X_4$ show no significant temperature dependence of the energy. The width of both $X_4$ and $\Delta_5$ belonging to the same branch is surprisingly high at all temperatures. A slight increase with lower temperatures is visible down to 150 K, where the phonons sharpen up again. The rather sharp $X_3$ phonon on the other hand reduces its width almost to the resolvable limit on approaching the transition. This contrasting behaviour of $X_3$ and $X_4$, which are compatible and incompatible with the symmetry of the phase transition indicates a particular role of $X_3$. More interestingly the reversal of the broadening trend of $X_4$ and $\Delta_5$ at lower temperature indicates a special regime in the region of 10 – 25 K above the transition temperature $T_V$.

In this temperature range a recent resonant x-ray diffraction study found indications of the formation of charge and orbital order, while the mean crystal lattice is still in its cubic high temperature phase. The phonons, as our result shows, react to the electronic order in their lifetime as expressed in the width.

References


[1] P. Piekarz, K. Parlinski, and A.M. Ole, Phys. Rev. Lett. 97, 156402 (2006) and Phys. Rev. B 76, 165124 (2007).

[2] J. E. Lorenzo, C. Mazzoli, N. Jaouen, C. Detlefs, D. Mannix, S. Grenier, Y. Joly, and C. Marin, Phys. Rev. Lett. 101, 226401 (2008).