ESRF	**Experiment title: Studies of the Verwey phase transition dynamics in magnetite**	**Experiment number**: HS2806
Beamline: ID10A	**Date of experiment**: from: 23 Nov. 2005 to: 29 Nov. 2005	**Date of report**: 10 Feb. 2006
Shifts: 18	**Local contact(s)**: Dr Andrei FLUERASU	*Received at ESRF:*

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The Verwey transition in magnetite at ca. T_V=125K is mainly known due to the rise of resistivity ρ by two orders of magnitude while cooling below T_V. The transition is also reflected in magnetic AC susceptibility χ_{AC} and the crystal symmetry that changes from cubic to monoclinic below T_V. In spite of long studies, the processes undergoing at the transition are not clear [1]. In view of these we have set up the project aimed to observe how χ_{AC}, ρ, specific heat and the structure change at the transition.

We have already shown [2] that while the sample temperature stayed constant ("utilizing" the effect of the latent heat), ρ and χ_{AC} gradually changed with time. Since the changes of the electronic pattern we are observing by χ_{AC} [3] and ρ are linked to the lattice, the direct observation of the proliferation of the new symmetry phase at the transition (e.g. by the observation of (4 4 ½) [4] superlattice reflections) was the next logical experimental step; this experiment constituted the main aim of our project at ESRF. The second goal was to record the speckle pattern around superlattice reflections; this should tell us how the structure fluctuates at the transition region.

Our experiment was conducted in ID10A on Troika 1, with the E=8 keV radiation. We have used point detector; the CCD detector was planned for XPCS. We have measured two single crystal stoichiometric magnetite samples: the first one, the same where measurements reported in [2] were conducted, was cut approximately parallel to (553) plane and had the miniature Pt1000 thermometer glued to the surface exposed to radiation. The sample was then glued to the cold finger of the cryostat via the thin glass plate to diminish the heat transfer (in order to maintain the constant sample temperature at the transition).

Although the well defined peaks ((333), (222), (440)) were found at room temperature, the inevitable movement of the sample after pumping out the cryostat caused the change of sample orientation. As a result, we could only observe temperature variation of the cubic phase peak shoulder (Fig.1). We have precisely scanned the transition region (with ca. 5 mK T steps) and the corresponding T dependence of peak position is shown on Fig. 2. The inset shows the temperature profile while the sample was continuously heated across the transition. Since the temperature stabilization was not reached (the blue line shows that much better stabilization is possible), it is apparent that the thermal insulation between sample and the cold finger was not sufficient. Although the data are very noisy, Fig. 2 suggests that the structural changes proceed gradually across the transition (bracketed by vertical lines), concomitant with ρ and χ_{AC} behavior[2].

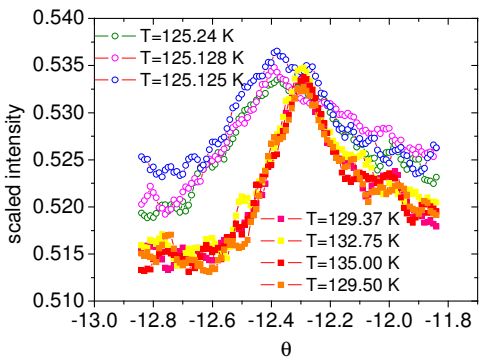

Fig. 1. The general change at the transition of the primary peak shoulder. Only the representative scans from the total 100 are presented

The second sample was cut parallel to (110) plane and was glued to the cold finger with in plane [110] direction parallel to the horizontal scattering plane.

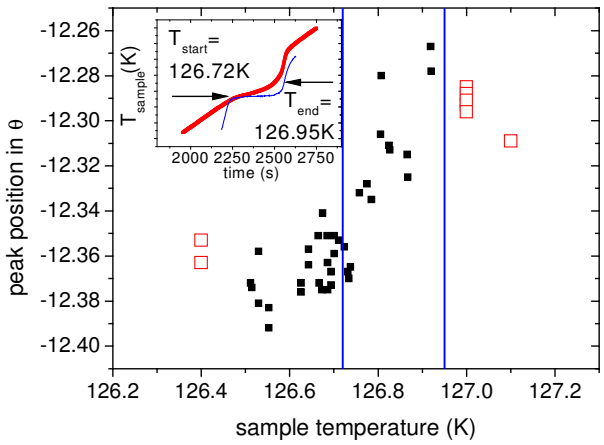

Fig. 2. Temperature dependence of the primary peak shoulder position in the transition region (bracketed by vertical lines at T_{start} and T_{end} estimated from the temperature profile shown in red in the inset). Red point indicate typical positions of the peak at low and high T (not in scale). The blue line in the inset is the T prifile of the same sample for better heat isolation [2].

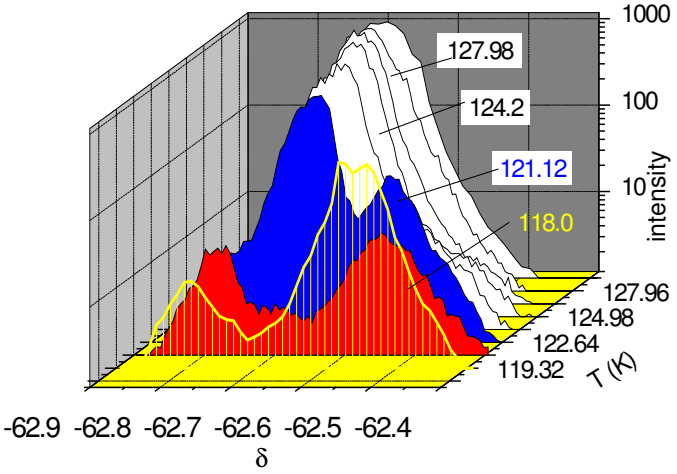

Fig. 3. Temperature variation of (440) peak shape (as a function of counter position δ) for the second sample. Coloured profiles indicate the transition region. Note, that sample position (angles θ, χ and φ) were optimized for right subpeak.

Since the additional thermometer was not installed, the temperature values from Fig. 3 refer to cold finger thermometer and the real sample temperature is 2-3 K above that reported. We have found the (400) an (440) reflections and we have traced (440) down to the Verwey transition temperature where it started to split and move in δ,θ,χ and φ space (see the Fig. 3). We then traced one of the resultant peaks and tried to optimize sample/counter positions for this particular reflection until the temperature was set to 2K below the transition temperature. Due to lack of time, no attempts were made to find superlattice reflection (4 4 ½). In the last thirty minutes we have, however, made introductory measurements of photon correlation with coherent radiation; the correlation function at T just below T_V for the second sample is shown on Fig. 4. Although the precise analysis was not possible (there was no time to fully optimise the apparatus for coherent scattering observation; the temperature was not stable enough and the peak centre was not known to measure the length scales of the fluctuations) the data clearly show that lattice dynamics, with the characteristic time τ of tens of seconds, can be observed in Fe_3O_4 by this technique.

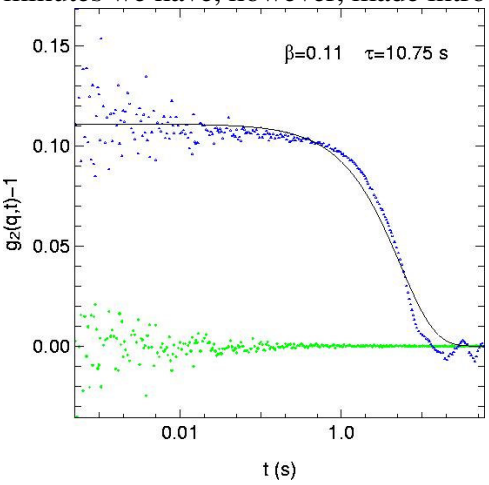

Fig. 4 Second order correlation function g_2 for the second sample (and the monitor readings; green). The solid line is a simple exponential fit $g_2(t,q)=\beta\exp(-2t/\tau)$.

There is no doubt that our experiment felled below our expectation. We are, however, now sure that this time was needed to get to know all the details and subtleties to ensure the full success of the future experiment (we are going to submit the proposal for the continuation of the HS2806 experiment), in particular, in the observation and quantitative analysis of lattice dynamics. Specifically, few points are now clear:

a) the sample must be fully oriented and the surface must be mirror polished; b) the heat transfer from the cold finger to the sample should be further diminished to allow for the constant temperature at the transition; c) only peaks that don't "move" at the transition (e.g. (4 4 ½)) should be observed, preferably with CCD detector; continuous optimization of sample position for the peak like (440) cause peak parameters not realistically reflecting symmetry changes at the transition.

We highly appreciate the cooperation with ESRF team. Also, two parts of the cryostat instrumentation were prepared on site prior to the experiment and our results clearly demonstrate the ingenuity of the cryostat prepared by Peter van der Linden group.

References

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