ESRF	**Experiment title:** STUDIES OF THE VERWEY PHASE TRANSITION IN MAGNETITE	**Experiment number**: HS3274
Beamline: ID10A	**Date of experiment**: from: 20 June. 2007 to: 28 June 2007	**Date of report**: 26 Febr. 2008
Shifts:	**Local contact(s)**: Dr Federico ZONTONE	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

*Andrzej KOZŁOWSKI, Zbigniew TARNAWSKI, *Wojciech TABIŚ, *Nhu KIM-NGAN TARNAWSKA, *Joachim KUSZ, Zbigniew KĄKOL

This experiment was the continuation of the experiment HS2806, and was aimed to observe the temporal changes of magnetite Fe_3O_4 lattice symmetry occurring at the Verwey transition and to study the fluctuations of the lattice close and at the transition. No such studies were conducted on this material till now.

At the Verwey transition at T_V=125K large latent heat manifests the abrupt change of major physical characteristics; e.g., the crystal symmetry that turns from monoclinic (space group Cc) below T_V to cubic Fd-3m. Despite 60 years of interest the transition is still not entirely understood. In view of that, we have set up the project aimed to simultaneously observe how magnetic susceptibility χ_{AC}, electrical resistivity ρ and the specific heat change exactly at the transition. Due to large latent heat of transition, the time of this observation may be largely extended. In the present experiment we added yet another characteristics, crystal lattice symmetry, that can be observed simultaneously with others mentioned above, while the transition develops.

Our experiment was conducted in ID10A on Troika 1, with the E=7.1 keV radiation. Two samples of stoichiometric magnetite, each ca. 0.5 g, were measured. One was cut parallel to (110) plane (hereafter referred to as "110 sample"), the other parallel to (553) plane ("553 sample"). For each measurement the miniature Pt1000 thermometer (ca. 1X1X0.5 mm^3) was glued on top of the sample, adjacent to the beam, to precisely monitor samples' temperature. Additionally, the special setup for the simultaneous observation of AC susceptibility χ_{AC} was constructed (two pairs of coils, wound together, one of each supplied AC magnetic field (f=125Hz), the other were pick up coils; the subtracted signal was measured by the lockin

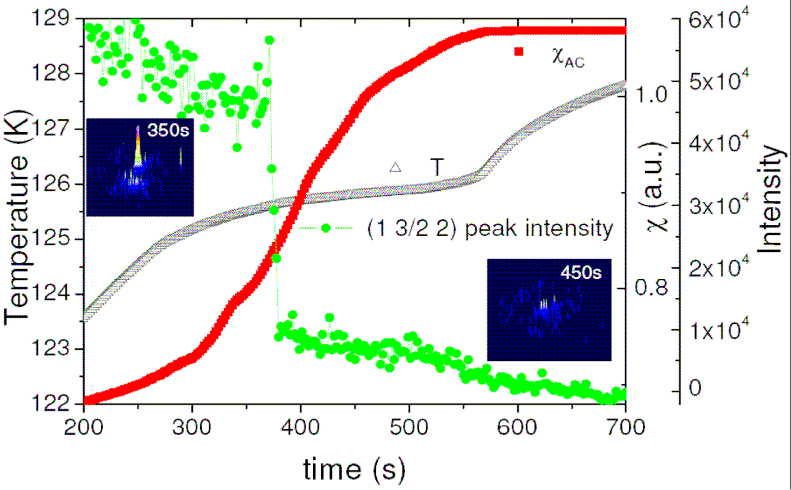

Fig. 1. "553" sample. Temporal changes of the integrated (1 3/2 2) peak intensity (integrated over main part of CCD screen), sample temperature and χ_{AC} across the Verwey transition on heating.The insets show characteristic CCD intensity profiles.

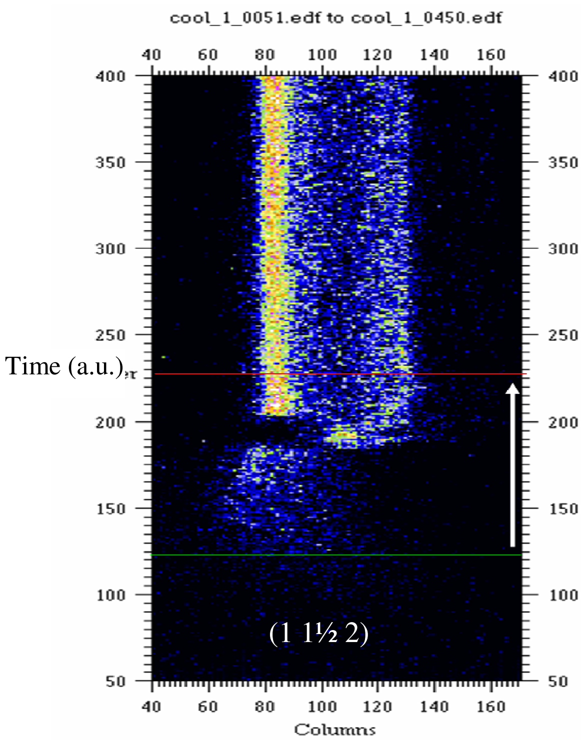

Fig. 2. "553" sample. Temporal changes of the CCD screen representative section for (1 1½ 2) peak on cooling (contrary to Fig. 1). Note that no peak is observed for Time<120 and that the peak changed its center of gravity from, initially, column 80, to 105 (180<Time<200) and, finally, again to 80.

amplifier hooked up to SPEC) and was used for "553" sample measurements. This setup was also treated as the heat barrier between the sample and the cold finger that should allow for the long time of transition observation. For each sample, the three primary high-T phase peaks were found (to calculate the orientation matrix), the superlattice peaks ((2 2 ½) in case of "110" sample and (1 1½ 2) in case of "553" sample) were located and traced through the Verwey transition, first by the point detector and then by CCD camera. In this last case, utilizing the beam partial coherence, we wanted to observe crystal lattice dynamics. The most important results are:

1. The superstructure peak disappears in the first one third part of the transition, as observed on heating (fig. 1, (1 1½ 2) peak of "553" sample), i.e. where both T profile-plateau and a χ_{AC} step still signal the undergoing processes, ultimately leading to a high T phase. Since this result is valid also for cooling (the superlattice peak always appears close to low-T site of a transition), it may reflect the general fact that the Verwey transition is caused by the structural changes that trigger the transformation of other subsystems.

2. The jumps of the (1 1½ 2) peak center of gravity in "553" sample (observed on CCD camera and shown on Fig. 2) are most probably due to structural twins dynamics occurring both at the temperature very close to the transition (Fig. 2), but also ca. 10K below T_V. This last result, observed also for (2 2 ½) peak of "110" sample, was found ca 2 hours after the low T phase was established.

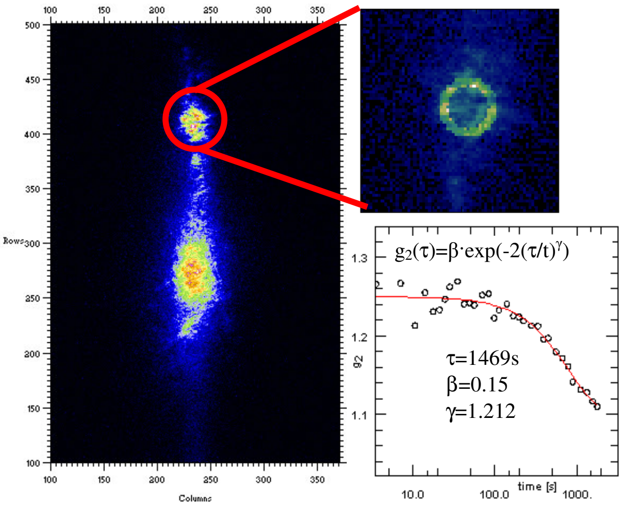

Fig. 3. The streched exponential fit to the one time correlation function calculated for part of the (2 2 ½) superstructure reflection in "110" sample. For this analysis, circular peak shape was assumed.

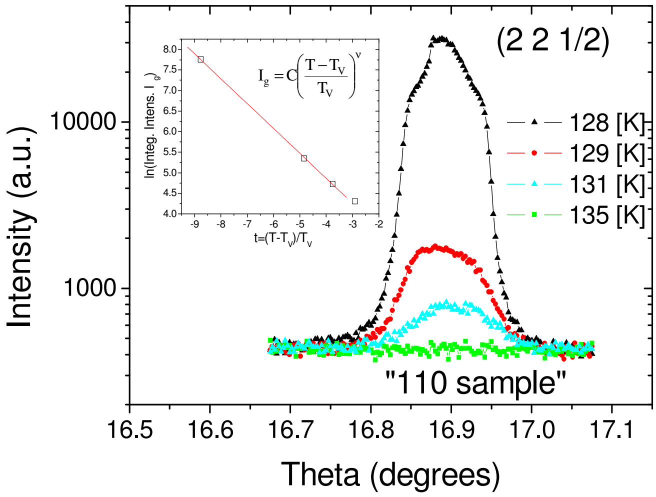

Fig. 4. Theta scan of the superstucture reflection for "110" sample at T just above the Verwey transition. Ln from the "integrated" intensity (surface below the peak) is plotted vs $t=(T-T_V)/T_V$ (where T_V=127.98K) suggests critical behavior

3. Some long lasting speckle dynamics was found for part of (2 2 ½) reflection for "110" sample (Fig.3) with the characteristic time τ declining with increasing Q values (radius of the circle). Here, due to very low peak intensity, the circular peak profile was assumed. Summarizing, we have observed either fast dynamics, as with the phenomenon shown on Fig. 2, or very slow, lasting hundreds of seconds (even longer than that on Fig. 3)

4. Characteristic diffuse scattering was found for "110" sample just above the transition (see Fig. 4), despite the fact that the transition is discontinuous. The critical exponent for the integrated intensity $I_g = C((T-T_V)/T_V)^{\nu}$ relation (see the inset of Fig. 4) was ν=-0.606. This resembles the neutron measurements results in [3] and will be the subject of the next proposal for the beamtime in ESRF.

In our opinion, the HS3274 experiment was successful: almost all from the planned issues were addressed. We have not have, however, enough beamtime to further study the Q scans for "553 sample" as well as we could not use the AC susceptibility setup for "110" sample due to problems with heat transfer between the sample and the cold finger. Finally, the problem of diffuse scattering at $T>T_V$, found for "110" was not even attempted for "553" ((1 1½ 2) dynamics was observed just before the beam time was over): this should be thoroughly studied in a separate experiment. All the problems addressed above are the subject of the still ongoing analysis and discussion.

We highly appreciate the cooperation with ID10A team. Also, the help offered by Peter van der Linden can not be overestimated.

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