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STRUCTURAL CHANGES AT THE VERWEY TRANSITION IN Fe_3O_4

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Keywords: metal-insulator phase transition; magnetite; photon correlation spectroscopy, crystal strucrure

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The aim of the experiment (HS3274, June 2007, ESRF, ID10A) was 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 using X ray photon correlation (XPCS) technique.

At the Verwey transition at T_V = 125 K, a 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*-3*m*. Despite 60 years of interest the transition is still not entirely understood. In view of that, we have set up the project [1, 2] 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.

Two samples of stoichiometric magnetite ("110 sample" and "553 sample"), each ca. 0.5 g, were measured with the *E*=7.1 keV radiation and the superlattice peaks' (2 2 ½) and (1 1½ 2) dynamics was observed by CCD camera with the partially coherent beam. Sample temperature was monitored by the miniature Pt thermometer glued on the sample. AC susceptibility χ_{AC} was measured simultaneously by the setup within the sample holder.

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.

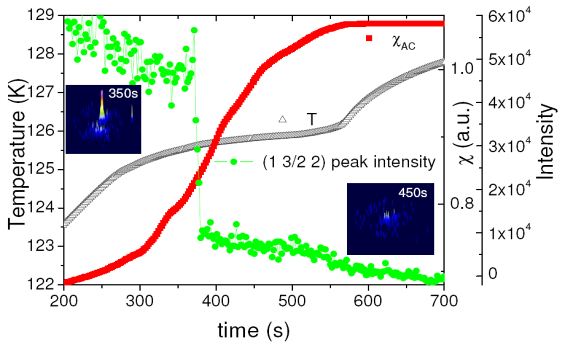

Figure 1. "553" sample. Temporal changes of the integrated (1 3/2 2) peak intensity (integrated over main part of CCD screen), sample temperature and χ_{AC} across the Verwey transition on heating.The insets show characteristic CCD intensity profiles.

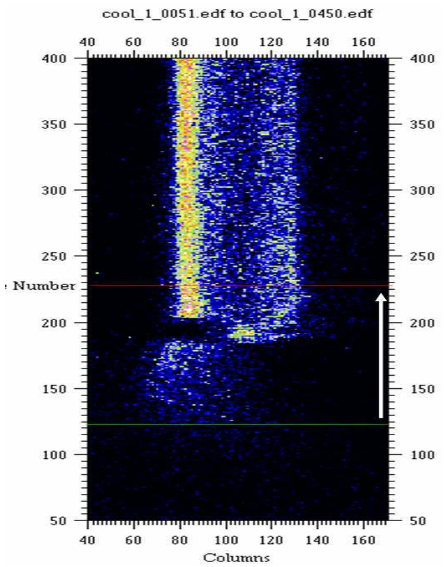

Figure 2. "553" sample. Temporal changes of the CCD screen representative section for (1 1½ 2) peak on cooling (contrary to Fig. 1). Note that no peak is observed for Time<120 and that the peak changed its center of gravity from, initially, column 80, to 105 (180<Time<200) and, finally, again to 80.

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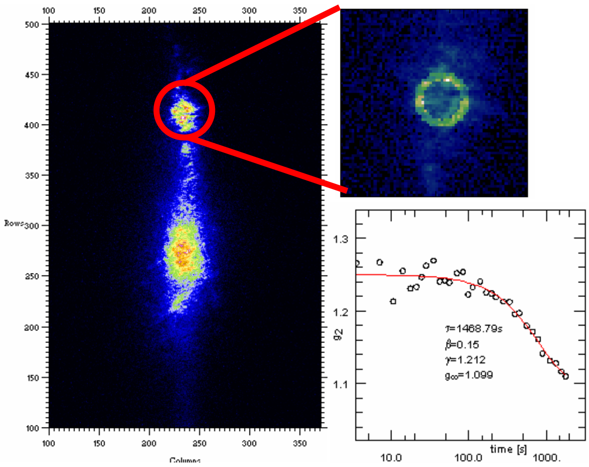

Figure 3. The streched exponential fit to the one time correlation function calculated for part of the (2 2 ½) superstructure reflection in "110" sample. For this analysis, circular peak shape was assumed.

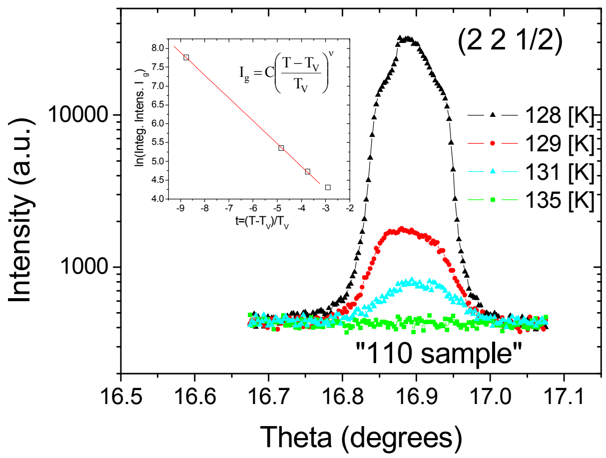

Figure 4. Theta scan of the superstucture reflection for "110" sample at T just above the Verwey transition. Ln from the "integrated" intensity (surface below the peak) is plotted vs $t = (T\text{-}T_V)/T_V$ (where T_V = 127.98 K) suggests critical behavior.

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.
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.
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 $I_g = C((T\text{-}T_V)/T_V)^V$ 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.

Summarizing, both fast dynamics, as with the phenomenon shown on Fig. 2, or very slow, lasting hundreds of seconds (even longer than that on Fig. 3) have been observed by our XPCS experiment. These studies and the data analysis will be continued.

Acknowledgements: Support of the Polish Ministry of Education and Science Grant No. 1P03 B01530 is acknowledged.

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