ESRF	**Characterization SiO_2 phase transitions by Bader's topological analysis of the experimental electron density obtained by MEM treatment of in situ single crystal X--ray diffraction structure factors**	**Experiment number**: ch-2069
Beamline: ID11	**Date of experiment**: from: 14 dec 2005 to: 19 dec 2005	**Date of report**: 31 march 2006
Shifts:	**Local contact(s)**: Dr. Chiara Capelli	*Received at ESRF:*

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Report:

The transition on a single crystal from α-quartz to β-quartz at room pressure and T ≈ 573 °C has been monitored by means of topological analysis in the Bader's theory framework of the experimental electron density reconstructed via Maximum entropy Method (MEM). The experimental data have been obtained in-situ using the X-Ray diffractometer at ID11 beamline (Bruker Axis CCD area detector). In particular, three data collection have been accomplished, i.e. at room T, T=565 °C and T=585°C. X-ray data have been treated by means of SAINT (integration) and SADABS (corrections) programs within the software package of the ID11 diffractometer. Full-matrix least-squares refinements of the experimental data have been made by the program JANA2000 (Petricek et al, 2005). Refinements have been made on |Fo| (≈ 1000 unique reflection with $\sin\theta/\lambda$ up to 1.4 $Å^{-1}$), taking into account of harmonic ADP's and monopolar multipole expansions, giving an average R = 2%. The relatively low quality of the data sets is due to the heating apparatus, involving a quartz-glass tube containing the crystal (average size ≈ 0.1 mm). The structure factors, put in absolute scale and corrected by isotropic extinction parameters, have been used in the MEM analysis program BAYMEM (Palatinus et al., 2005- University of Praga). MEM was made starting from a prior information obtained by reconstruction of a theoretical pro-crystal map obtained by Fourier summation of a very large number of calculated structure factors obtained from the refinement results. Because of the imperfection of the data, experimental electron densities are affected by the presence of some ripple in the low-density regions.

However, the comparison with the theoretical electron densities obtained via Hartree-Fock quantum mechanical calculations (program CRYSTAL98, University of Turin) lead to recognize in a safe way that the bond critical points [(3,-1) as indicated in Bader, 1992 - Atoms in Molecules] of the structure (wihich involve 2 interaction Si-O and 1 interaction O-O in the low-temperature sample- Fig. 1) evolve to a more complex setup of the chemical bonds, involving at high temperature (T=565°C) the novel formation of several O-O interaction (Fig. 2). Note that the thermal motion of the high temperature electron density map yields to higher values of the electron density at the bond critical points with respect to the static quantum mechanical simulation, showing which is the effect in the electron distribution due to the instantaneous rearrangement of the electron density in a real crystal in an excited state.
A complete topological analysis both in term of study of the other kind of critical points and in term of integration on the atomic basin is still in developement

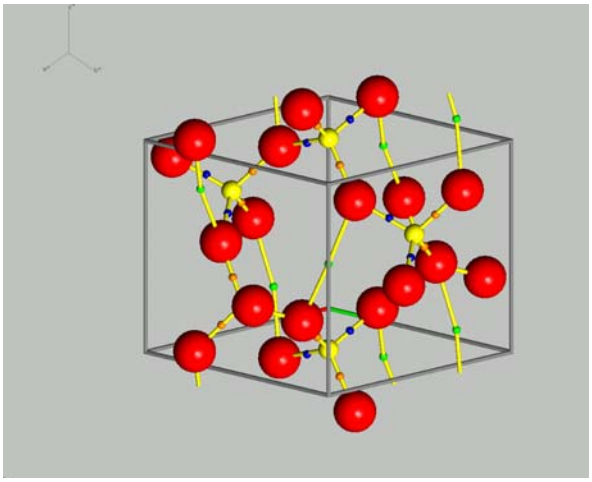

Fig. 1 - MEM structure of quartz at room T. Yellow spheres = Si; Red spheres = O; blue and orange little spheres = (3,-1) critical points along Si-O bonds; green little spheres = (3,-1) critical points along O-O bonds;

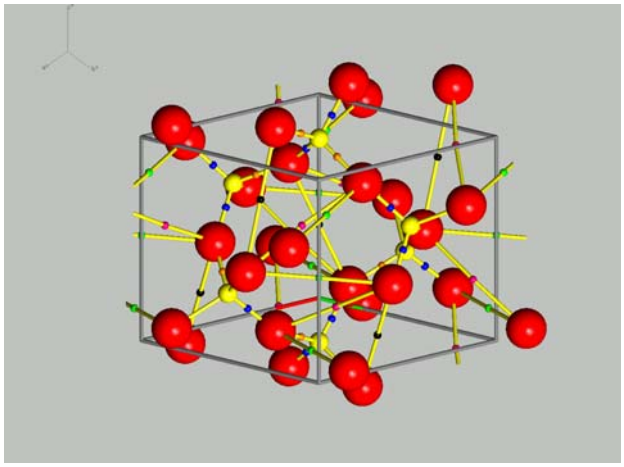

Fig. 2 - MEM structure of quartz at room T. Yellow spheres = Si; Red spheres = O; blue and orange little spheres = (3,-1) critical points along Si-O bonds; green, magenta and black little spheres = (3,-1) critical points along O-O bonds;