ESRF	**Experiment title:** Late stage phase ordering kinetics of the Ortho-II transition in $YBa_2Cu_3O_{6+x}$	**Experiment number:** HS-26
Beamline: ID11	**Date of Experiment:** **from:** Sept. 23, 96 to: Sept. 29, 96	**Date of Report:** Aug. 25, 1997
Shifts: 15	**Local contact(s) :** P. Rejmankova	Received at ESRF: 2 8 AOUT 1997

Names and affiliations of applicants (*indicates experimentalists):

Paul SCHLEGER* and Helene CASALTA*

Institut Laue-Langevin
BP 156
38042 GRENOBLE Cedex 9

Report:

The cell-doubled ortho-II phase is a predominant structural feature of the high temperature superconductor $YBa_2Cu_3O_{6+x}$. It is charactcrized by short range ordered alternating full and empty Cu-O chains in the basal plane, and associated atomic displacements of surrounding atoms, giving rise to strongly modulated *diffuse* superstructure peaks. The absence of long range order was suggested (PRL 74, 1446, (1995)) to arise from random-field effects or from an intrinsic tendency towards glassy behaviour. To investigate the issue of the ordering kinetics and transition character further, this experiment directly measured the correlation length development upon quenching from the disordered to the ordered ortho-II phase.

A first experiment was conducted in 1995 (HC 259) focussing on the short-time behaviour (t < 8 hrs) where the $\xi \sim \sqrt{t}$ Allen-Cahn growth was clearly demonstrated, but a distinction between logarithmic or stretched-exponential behaviour was not evident. In this follow-up experiment, we focussed on the later stages of growth (t < 42 hrs), which promised to reveal the ordering character.

A 2 mg single crystal of $YBa_2Cu_3O_{6\,5}$ was mounted in a specially designed small oven capable of quenching rates of up to 10 °C/sec. The large Kappa diffractometer was used with 22 keV x-rays focussed both horizontally and vertically. The sample was quenched from 180 °C to various temperatures (100. 110, 118, 125). The short time ordering (10 s < t < 20 min) was determined from measurements of the peak intensity of the (4.5,0,0) superstructure peak. For longer times (1 min < t < 42 hrs), the c-axis correlation length was directly determined from repeated I-scans over the (4.5,0,0) peak fitted to Lorentzian-squared line shapes. Due to the beam instability which caused the beam-spot to wander across the sample over time, diffractometer realignments were necessary roughly every 2 hours to guarantee that the always same part of the crystal was illuminated.

The last shift was used to measure a series of short-time quenches (10 s < t < 20 min) to 100, 125, 130, 140, 145, 150 °C, to complement the data set taken in our first, experiment.

Although we again had difficulty dealing with the beam instability, the quench to 100 °C was sufficiently accurate to show that indeed an approach to equilibrium via a stretched exponential is unlikely (Fig. 1). Despite the kink occurring after roughly 8 hours (coinciding with the first refill), the data clearly favours a logarithmic growth.

Recent papers dealing with the phase ordering kinetics of Ising systems with competing interactions showed the existence of logarithmic growth *in the* absence of disorder (e.g. J.D. Shore et al., PRB 46 11376 (1992); M. Rao and A. Chakrabarti, PRE 52 R13 (1995)). A subsequent Monte-Carlo simulation for the Ortho-II phase in $YBa_2Cu_3O_{6.5}$ (P.J. Kundrotas et al. PRB 54 4357 (1996)) saw indications of logarithmic growth arising from the competing in-chain and cross-chain interactions. These simulations were made using tetragonal symmetry as the initial condition, using an asymmetric next-nearest neighbour model (ASYNNNI) which is inconsistent with experiment. Using Ortho-I symmetry as the initial condition, we find similar behaviour, however. Although the pure ASYNNNI model by no means accurately describes the structural phase diagram and the oxygen ordering thermodynamics in this system, the generic feature that competing interactions lead to logarithmically slow growth is a strong indication that this may be at the origin of the Ortho-II ordering behaviour.

The basic idea is that the strong tendency for Cu-0 chain formation exists already at much higher temperature and gives rise to scale dependent energy barriers against domain wall movement within the Ortho-II phase. Whereas in an impurity picture, the cross-over from Allen-Cahn to logarithmic growth occurs when the correlation volume becomes comparable to the inverse of the impurity concentration, the cross-over in a competing interaction picture seems to happen once the Ortho-II order parameter starts to grow, i.e. when one Ortho-II domain becomes favoured over the other, and all local defects have been annihilated.

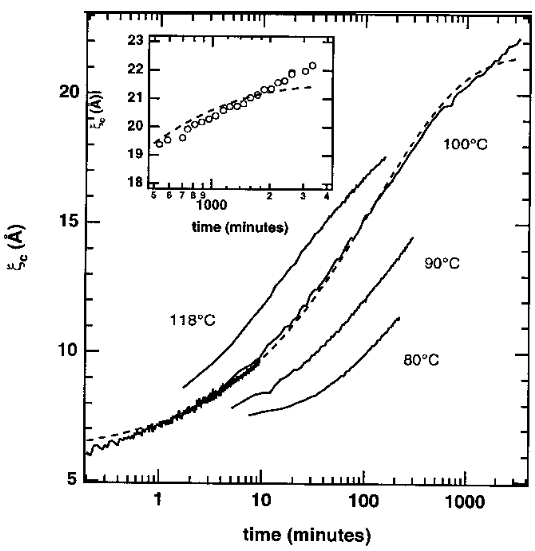

Fig. 1 Time dependence of the c-axis correlation length for various quenches from 180 °C. The insert shows a blow-up of the 100 °C quench at t,he latest times. The dashed line is a fit using a stretched exponential $\exp(\sqrt{t/\tau})$. The insert illustrates the difficulty that this form has in describing the late time data.

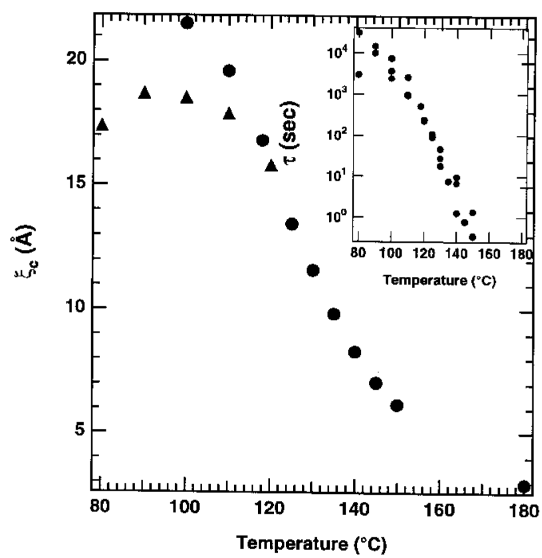

Fig. 2 Temperature dependence of the extrapolated infinite time c-axis as deduced from a stretched exponential fit. The circles are from the data of this experiment and the triangles from our previous experiment. The saturation below 100 °C is due to the inability of the stretched exponential form to fit the late-time data. The insert shows the ordering time constant τ from all our measurements.