ESRF	**Experiment title:** **Coherent X-ray diffraction study of the dynamics of long wavelength phason fluctuations in quasicrystals**	**Experiment number**: HS1772
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Shifts:	**Local contact(s)**: F. Yakhou	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

*M. de Boissieu, *S. Francoual, *F. Livet, *F. Bley, LTPCM, INPG, UMR CNRS 5614, BP 75 38402 St Martin d'Hères Cedex, France

*R. Caudron, LEM, Onera-CNRS, BP 72 92320 Chatillon, France

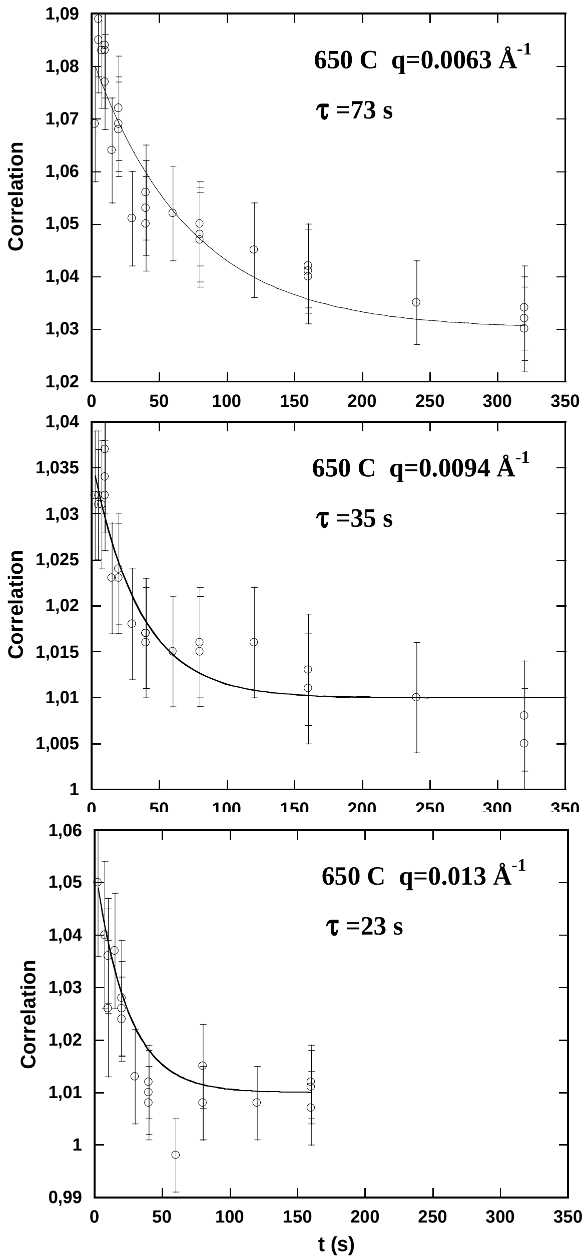
Report:

Quasicrystals are long range ordered material which lack translational invariance. They can now be obtained as large single grain with a high structural quality. Because of the quasiperiodic long range order, new modes, called phason, are believed to exist in quasicrystals. Unlike phason modes in incommensuratly modulated phases these modes are collective diffusive modes and do not propagate. This is because at the atomic scale long wavelength fluctuations result in atomic jumps between two positions having almost the same local environment (see [1] for an introduction).

Long wavelength phason fluctuations give rise to diffuse scattering located close to the Bragg reflections in very much the same way that phonon gives rise to thermal diffuse scattering (TDS). Such a diffuse scattering has been observed in the i-AlPdMn phase and can be interpreted as resulting from phason fluctuations [2]. Its temperature evolution indicates that these fluctuations are pre-transitional fluctuations which are frozen in at room temperature [3].

Theoretically, since these modes are diffusive, a mode with a wavevector q is expected to decay in time exponentially with a characteristic time decay varying as q^{-2}. Since long wavelength phasons are thermally activated we might expect to find a temperature (above 400°C) at which slow fluctuations occur and should be measurable by coherent X-ray diffraction. In a previous experiment we have shown the feasibility of such an experiment: clear speckle patterns have been observed in the diffuse scattering at room temperature [4].

We used the same experimental set-up as in ref [4] to produce a coherent beam on the ID20 beam line: a set of slits (60x60 μm^2) located at 3 m from the sample acts as the effective source. The coherence of the beam is defined by a 10 μm pinhole just before the furnace. The sample was placed in an evacuated furnace, under a secondary vacuum. The speckle pattern was recorded by a Princeton Instrument directly illuminated CCD camera, located at 1.8 m from the sample. Data have been analysed by a droplet algorithm [5].



However the coherence of the beam, measured with a standard silica aerogel, was significantly smaller than previously observed and measurement in the quasicrystal have been restricted to the low angle 5-fold reflection with indices 7/11 (q=1.8 Å^{-1})

The i-AlPdMn sample was previously annealed at 780C for 8 days under UHV: this led to the formation of large terraces with a characteristic size of 50 µm. This sample preparation has been crucial for the surface stability during the ESRF experiment. For each temperature, the sample position has been optimised so as to bring the footprint of the beam inside one terrace in order to avoid spurious interferences fringes.

Above 500C, a significant time dependence of the speckle pattern has been observed. Reliable data could be obtained at 600 and 650C, with 3 different positions of the 2D detector. The correlation function as a function of time is shown in the figure. Data have been fitted by exponential time decay, as expected for a diffusive process, from which τ, the time decay, is extracted. Results of the fit are shown as a solid lines on the figures. As can be seen, τ decreases as q increases. The number of measurement is small, but the obtained data are compatible with a diffusive law, i.e. $\tau = D_{phason}\, q^{-2}$, with $D_{phason} = 310^{-17}\ m^2s^{-1}$ at 650C .

Similar results have been obtained at 600C, with a much slower dynamics, since D_{phason} is about 10 time larger. This indicates that phason fluctuations dynamics is thermally activated.

These results are compatible with what is known for atomic diffusion in quasicrystals. Mn atoms are slow diffuser with $D_{Mn} = 10^{-14}\ m^2s^{-1}$ at 65OC [6]. Taking this value, and a phason wavelength of 1000 Å (q=0.0063 Å^{-1}) the mean distance <l> that the Mn atoms will span during the time τ= 73s, is of the order 8700 Å, i.e. 9 time the phason wavelength.

In conclusion, dynamics of phason fluctuations have been observed by coherent X-ray scattering. As predicted theoretically it is a diffusive process.

References

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