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Report:

Quasicrystals (QCs) are intermetallic alloys with a particular type of aperiodic order, displaying the absence of periodicity with the existence of a perfect long-range order. In a very efficient high-dimensional description of aperiodic structures, the overall atomic arrangement in the physical space (E//) can be viewed , for icosahedral (i-)QC phases, as a 3-D section of a 6-D cubic lattice discretely decorated with 3-D atomic surfaces defined in a complementary space $E_{\perp}$. Considering the symmetries leaving the free-energy invariant, in addition to the usual acoustic phonons, new hydrodynamic modes are defined, the so-called phason modes, associated with degrees of freedom along $E_{\perp}$. In QCs, long-wavelength phason fluctuations are diffusive modes and correspond to correlated atomic rearrangements where atoms jump between sites with similar local environments [1]. Within the elasticity theory of i-QC phases, phason fluctuations give rise to an anisotropic diffuse scattering intensity around Bragg peaks in the reciprocal space in addition to the thermal diffuse scattering originating from phonons. In the i-AlPdMn QC phase, such diffuse scattering due to phasons was shown to dominate the overall diffuse scattering intensity [2] with a temperature evolution suggesting the pre-transitional nature of those fluctuations [3].

Phason modes are diffusive. A given set of phason fluctuations with a wavelength λ should thus exponentially decay in time with a characteristic time decay τ_0 proportional to λ^2. A same time dependence is expected for the associated instantaneous diffuse intensity if measured at a distance $q=2\,\pi/\lambda$ far from a Bragg peak. This dynamical process was recently evidenced in the i-AlPdMn QC using coherent x-ray scattering on ID20 [4-5]. In agreement with frozen-in phason fluctuations at room temperature, the intensity correlation function $F_{cor}(q,t)$ remained constant up to 500°C. At 650°C, an exponential time decay was observed as a signature of the temperature-activated phason fluctuations. The time τ_0 of the order of a few seconds could be extracted as well as the diffusion coefficient D(T=650°C) derived from the obtained 'dispersion' relation: $(1/\tau_0) = D.q^2$. Those measurements were performed for a $(\tau,-1,0)$ direction of the wavevector q as imposed by the experimental set-up and by the orientation of the sample.

For a different direction of q, for a given T, a different value of D is expected since phason modes with a different polarisation are selected.

We used the same experimental set-up as in ref [3] and [6]. The sample was placed in an evacuated furnace under secondary vaccuum. A set of 60x60 µm² secondary slits located at 3.6 m before the sample is the effective source. A 10 µm pinhole achieves the degree of coherence β of the incoming beam (E=8 keV). A directly illuminated Princeton 2D CCD located at 1.8 m after the sample is used to record the diffuse scattering patterns. The sample was oriented with a 2-fold axis perpendicular to the surface allowing to investigate phason fluctuations propagating along a (1,0,0) direction.
The coherence of the beam β was estimated to 26% for θ=0. Due to short longitudinal coherence, β decreases to 10 ± 1 % and 5 ± 1% for the low- and high-angle 2-fold (2-f) 8/12 and 20/32 Bragg peaks, respectively.

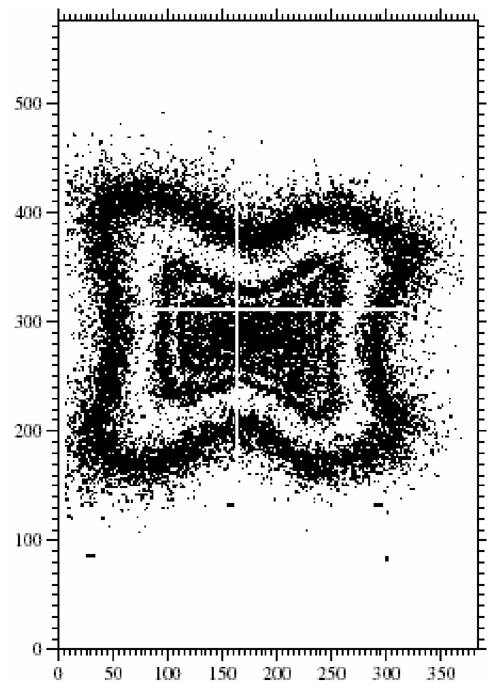

fig. 1 Isocontours of the diffuse intensity at 600°C for q=0.005 Å^{-1} (δθ=0.15°) away from the 8/12 Bragg peak. The white intersecting lines indicate the position of the wavevector q lying along a 2-fold axis almost orthogonal to the plane of the CCD.

Whatever the q position far from the selected Bragg peak, the diffuse intensity was found strongly anisotropic as clearly visible in fig.1 and as expected in this direction for a diffuse scattering due to phason fluctuations. Within our data analysis, in the calculation of the $F_{cor}(q,t)$ correlation function, after a mean over the time t of the intensity-intensity correlation function I(**q**,t)(I(**q**,t+t'), an average is performed on successive circular shells centred around a (100) axis originating from the 2-f reflection of interest. Such strong anisotropy actually limitates the validity of our data treatment to the single central shell.

From room temperature up to 400°C, $F_{cor}(q,t)$ was found constant over a time of measurement of 1000s, in agreement with previous measurements and with frozen-in phason fluctuations.

For higher temperatures (T>500°C), several experimental problems occured making the analysis still complex to carry out :
- deficiencies in the thermal contact between the sample and the furnace were observed and the sample holder had to be changed.
- the total number of scattered photons on each frame was found to display strong variations in the time with as main consequences a drastic decrease of the coherence. Those variations in the scattered intensity were found directly linked with fluctuations in the number of incident x-rays indicating unstabilities of the optics.

Data analysis is in progress in order to determine if the observed time evolutions of the coherence for temperatures above 500°C can be related or not with dynamical effects due to phason fluctuations.

References :
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