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Report:

Scattering coherent X-rays at a sample displaying disorder leads to a so-called speckle pattern in the scattered intensity, which is nothing but the absolute square of the Fourier transform of the (disordered) electron density in the sample. Recording the temporal fluctuations of the speckle pattern is the principle of X-ray Photon Correlation Spectroscopy (XPCS). This technique therefore measures the lifetime of fluctuations in the sample corresponding to a given scattering vector $\vec{q}$. We have recently demonstrated that it is possible to apply this technique for studying the dynamics at atomic scales: by using a single crystal and varying $\vec{q}$ both in length and orientation relative to the crystal lattice one can elucidate the diffusion mechanism of single atoms [1].

Here we report on our measurements at a single crystal of $Si_{89}Ge_{11}$. Measurements were done with a photon energy of 8 keV, the Si(111)-monochromator gave a temporal coherence of $\Delta E/E = 1.42 \times 10^{-4}$ and the beam-defining slits were set to $10 \times 10\,\mu m^2$. The beam was focussed with the compound refractive lenses-system. The sample was mounted in a custom-built vacuum furnace. The entrance and exit windows of both furnace and flight tube were made from Kapton foil. Frames were taken with ID10A's 1024×1024 CCD camera with $13 \times 13\,\mu m^2$ pixel size, the exposure was 10 s per frame with about one second read-out. The distance sample–detector was 0.5 m. With this set-up, the count rate was about 0.01 photons per pixel and frame, the coherence factor β was about 0.06 at scattering angles $2\theta = 10°$, at higher angles even less.

The results obtained in these measurements were of limited value from the viewpoint of physics, but they provided valuable insights about the method: During the course of the measurements, it was often the case that the two-time auto-correlation functions showed features incompatible with equilibrium dynamics. Fig. 1 as an example shows an apparent slowing-down of dynamics. This was often observed when changing the temperature, but there happened also more complex phenomena. Another point to mention is the form of the decay of the auto-correlation function when selecting a range of frames where the dynamics were apparently stable. As can be seen in Fig. 2, the closest fit is given by a compressed exponential decay with a compressing exponent of about 2.

These findings are incompatible with the knowledge about the system SiGe: There is no long-range order and only very weak short-range order, therefore there is no parameter whose equilibration could lead to the apparent slowing-down. Furthermore, as it is a crystal with two inequivalent sites, the auto-correlation function should be composed of either one or two exponential decays, depending on the mechanism of dif-

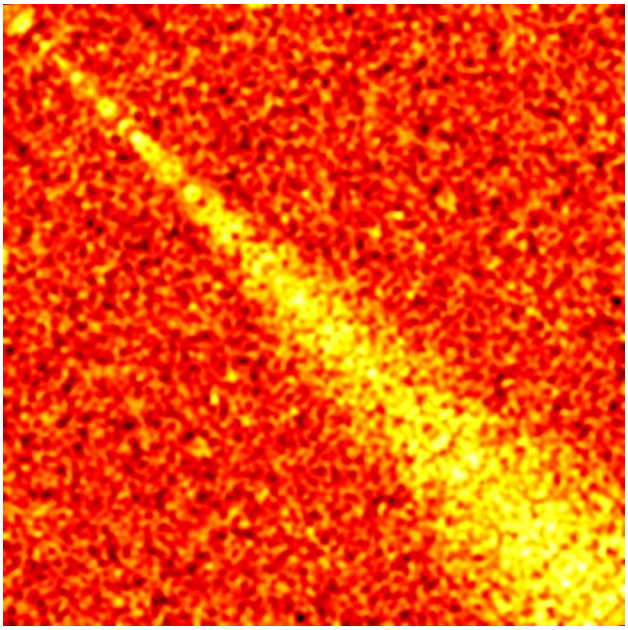

Figure 1: Two-time auto-correlation function showing instability. $(t_1, t_2) = (0, 0)$ is in the left upper corner.

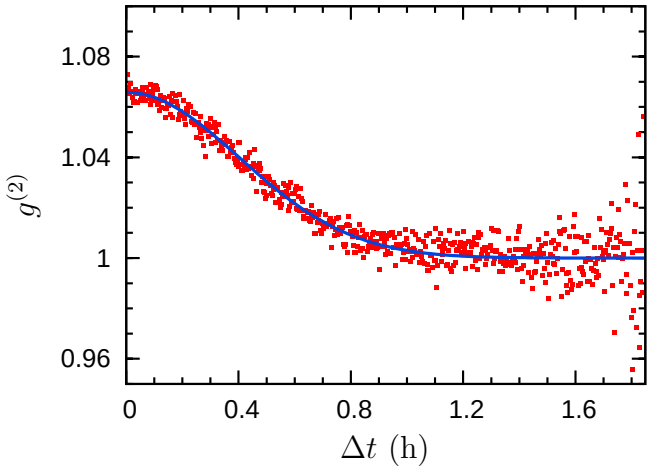

Figure 2: Auto-correlation function computed from stable part, fitted with compressed exponential decay.

fusion. Phenomenologically, this can lead to an apparent stretched decay, but not to a compressed decay.

Our interpretation is that the observed features are not due to dynamics, but to movements of the sample with respect to the beam. Specifically we think that the furnace performs a rotation with the sample. The speckle pattern is essentially just the electron density of the sample in reciprocal space, so if the sample rotates, the speckle pattern rotates in reciprocal space. The same wavevector-transfer therefore probes a succession of points in the speckle pattern, leading to a decay of the temporal auto-correlation function. This also explains the compressed form of the decay: Assuming a speckle form of $(\sin(x)/x)^2$ and taking the finite pixel size into account leads to a function which very much resembles a compressed exponential decay with compressing exponent 2.

Comparing the dimension of the speckles in reciprocal space (about $(10\,\mu\mathrm{m})^{-1}$) with the length of the scattering vector (about $(1\,\text{Å})^{-1}$ at high scattering angles) leads to the requirement of a stability of 0.001° of the orientation of the sample over the duration of the experiment. Obtaining such a stability is a challenging task, especially given the high temperatures (above 800°C) in the present investigations, however, our previous experiments at lower temperatures (about 300°C) have show that it is feasible.

We also used a small part of this beam-time for feasibility studies on the B2-system FeAl, specifically with a single crystal of $Fe_{65}Al_{35}$. As the K-edge of Fe is at 7.11 keV, with a beam energy of 8 keV the amount of fluorescence is orders of magnitude higher than the diffuse elastic scattering. We also tried to measure immediately below the edge, which suppressed the fluorescence to a high degree, but concomitantly also the incident flux went down. This is because of the wavelength dispersion of the Beryllium lenses. An alternative would be to measure at the ID10C-branch, where it is possible to focus 7 keV-radiation at the position of the sample.

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[1] M. Leitner, B. Sepiol, L.-M. Stadler, B. Pfau, and G. Vogl, Nature Mat. **8**, 717 (2009).