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Report:

Using the coherent part of the x-ray beam of third-generation synchrotrons the nanometre-scale dynamics of thin samples can be followed in the time domain using the technique of x-ray photon correlation spectroscopy (XPCS). Due to the coherence of the incident x-rays the scattered radiation shows modulation ("speckles"), which is given by the squared modulus of the Fourier transform of the electron density in the sample. Determining the lifetime of the speckles at a given scattering vector $\boldsymbol{q}$ allows to draw conclusions on the dynamics in the sample.

Here we successfully demonstrate the feasibility of following the diffusion of single atoms by XPCS. As the evolution of disorder on atomic scales manifests in the scattering at q-values on the order of Å^{-1}, the scattered intensity is spread over a large solid angle. This entails very low scattering intensities per detector pixel. As a consequence one preferably wants to measure systems that show much atomic-scale disorder, with metallic glasses as the most promising candidates. Here we report on preliminary results obtained from samples of the metallic glass $Zr_{65}Ni_{10}Cu_{17.5}Al_{7.5}$ [1] and of a short-range ordered single crystal $Cu_{90}Au_{10}$ [2].

The samples were mounted in a custom-built vacuum furnace with direct connection to the flight tube in order to reduce parasitic scattering at air and kapton windows. 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 $7 \times 7\,\mu\text{m}^2$. In order to increase the scattered intensity, the beam was focussed with beryllium refractive lenses. Frames were taken with HASYLAB's 1340×1300 CCD camera with $20 \times 20\,\mu\text{m}^2$ pixel size. With the detector at a distance of 1.25 m from the sample, a contrast of $\approx 3\%$ was obtained at a scattering angle of $2\theta = 37°$. The exposure time per frame was 5 s with additional 2.3 s for reading out the whole chip. Aforementioned $2\theta = 37°$ means $q = 2.6\,\text{Å}^{-1}$, which is the position of the first diffraction maximum of this glass, corresponding to characteristic atomic distances of 2.4 Å. With this geometry, count rates were typically about 6 photons per pixel and hour. Varying the temperature between 260°C and 370°C, the instantaneous dynamics were followed. Data were evaluated by intensity auto-correlation functions. In accordance with Fig. 7 in Faupel et al. [3], a strong influce of annealing was foung, see Fig. 1 here. Further evaluation will show whether our data can be described with Faupel et al.'s Eq. (3.3), where the diffusivity is given by a stationary contribution due to thermal fluctuations and a

transient contribution due to structural relaxation, and whether the stationary contribution is really just a function of temperature, independent of sample history.

In order to obtain information about the diffusion mechanism we attempted to record the correlation times as a function of q, the modulus of the scattering vector. To this end measurements were performed with a fixed temperature of 360°C, but with 6 different scattering angles between 18° and 42°. Unfortunately, apart from the measurement at 37°, none of them shows any correlations, see Fig. 2. The reason for that can either be that the counted photons did not come from the sample but were parasitic scattering for all measurements except the one at 37°, or that the dynamics are at least a factor of ten faster outside the peak than in the peak. Both possibilities seem unlikely to us, and we plan to redo these measurements with smaller steps in 2θ in order to see whether the contrast goes down or indeed the correlations become faster.

We also measured the dynamics in the solid solution $Cu_{90}Au_{10}$. Fig. 3 shows the dynamics in the vicinity of principal crystallographic directions. This information can be used to fit a diffusion model to the data.

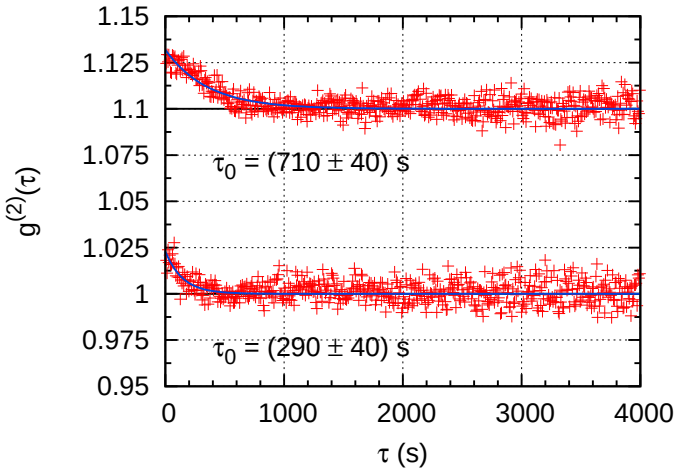

Figure 1: Influence of structure relaxation in metallic glass: sample heated for first time to 350°C and measured (lower data set), same sample again at 350°C after 2 h at 370°C (shifted up by 0.1).

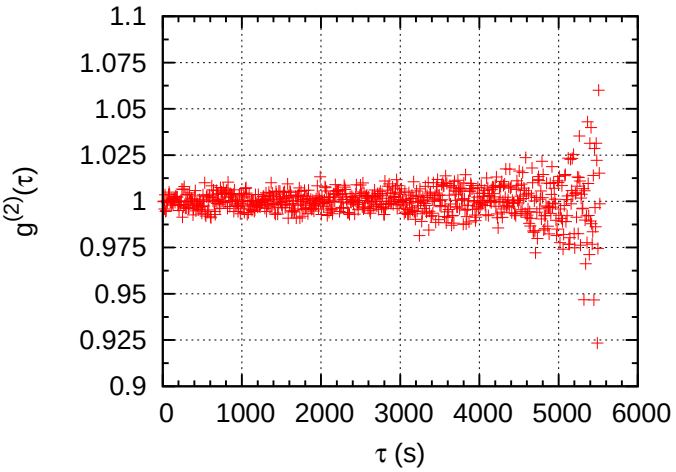

Figure 2: Auto-correlation function at $2\theta = 32°$ showing no contrast.

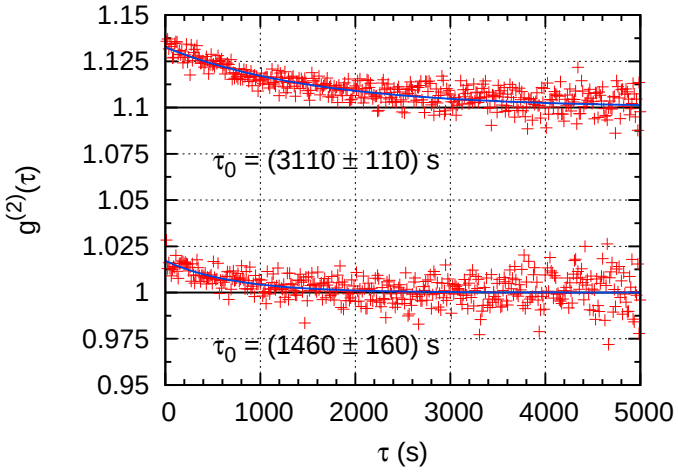

Figure 3: $\boldsymbol{q}$-dependence of relaxation time in $Cu_{90}Au_{10}$ at 270°C, $2\theta = 25°$: auto-correlation function with fit for $\boldsymbol{q}$ close to $\langle 111 \rangle$ (lower data set), auto-correlation function with fit for $\boldsymbol{q}$ close to $\langle 001 \rangle$ (shifted up by 0.1).

[1] Sample provided by A. Meyer, Physik Department E13, TU München.

[2] Sample provided by B. Schönfeld, Institut für Metallforschung, ETH Zürich.

[3] F. Faupel et al., Rev. Mod. Phys. **75**, 237-280 (2003).