ESRF	**Experiment title:** Revealing the coarsening mechanism in phase-separating systems with X-ray photon correlation spectroscopy	**Experiment number**: HS-2032
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Report:

Recently, it was shown that different coarsening mechanisms in phase-separating alloys can be distinguished in an X-ray photon correlation spectroscopy (XPCS) experiment by analysing the fluctuating speckle intensities with the fluctuation analysis (FA) technique [1]. Thereby, so-called fluctuation exponents α that are characteristic for long-term correlations are gained. For long-term correlations one obtains $0.5<\alpha<1.0$. By comparing the dependence of α on the scattering vector with Monte Carlo (MC) simulation results [2] the coarsening mechanism in the two systems Al-6at.%Ag at 140°C and Al-9at.%Zn at 0°C could be verified (coarsening via an evaporation/condensation mechanism – also known as Lifshitz-Slyozov-Wagner (LSW) mechanism – and coagulation of whole precipitates, respectively). Based on this first successful experiment we wanted to determine the coarsening mechanism in another phase-separating system, Al-12at.%Li at 150°C, which is believed to be a classical evaporation/condensation mechanism [2]. Additionally, we tried to follow the coarsening behaviour of Al-9at.%Zn at –15°C in order to find out whether such slow dynamics can be measured.

In both investigated systems precipitates form spheres in a disordered Al-based phase, but in contrast to Al-9at.%Zn precipitates in Al-12at.%Li are $L1_2$ ordered (Al_3Li) instead of purely consisting of the minority system component in fcc order. Because Al and Li have similar sizes the matrix-precipitate interfaces are highly coherent with low elastic energy, as is the case for the Al-9at.%Zn system.

The Al-9at.%Zn sample was homogenised at 400°C for 2 h, then quenched into liquid nitrogen, and then annealed at –15°C for 75 h. The Al-12at.%Li sample was annealed at 250°C for 1 h, then homogenised at 420°C for 6 min, and afterwards annealed at 150°C for 72 h. This treatment should ensure that the samples were in a quasi-equilibrium where only slow coarsening takes place and the mean precipitate size does not change measurably during the measurement.

Time series of speckle patterns were taken with a CCD camera (Princeton Instruments, directly illuminated chip, 1242×1152 pixels, pixel size 22.5×22.5 μm^2, placed ≈ 2.2 m downstream the sample) in small-angle X-ray scattering (SAXS) geometry. The Al-12at.%Li sample was measured in our furnace, the Al-9at.%Zn sample in a SAXS chamber provided by the TROIKA beamline. Both chambers were equipped with capton windows in order to preserve the coherence properties of the beam.

Depending on the refill, measurements were disturbed by periodic beam-position fluctuations with a period of ≈500 s, as was already noticed in a previous experiment (SI-838, see experimental report). Due to the very slow dynamics in case of Al-9at.%Zn at –15°C the measured speckle intensity correlations were dominated by the periodic signal of the beam and cannot be used for evaluation.
In case of Al-12at.%Li at 150°C dynamics was fast enough to slightly dominate the correlation behaviour. But, the very low scattering intensity emerged as an additional problem. In the SAXS maximum, see Fig. 1 for the integrated SAXS curve, at most 2 photons/s were counted per pixel as compared to 5 photons/s in case of the Al-9at.%Zn sample or almost 15 photons/s in case of the previously measured Al-6at.%Ag sample [1]. The main reason for the comparatively low scattering intensity for Al-12at.%Li is that the precipitates consist mainly of Al (Al_3Li), which reduces the contrast to the matrix. The low scattering intensity results in weak long-term correlations visible in the fluctuation functions, shown in Fig. 2. These curves are fitted $\propto t^{\alpha}$, yielding the fluctuation exponents α. The fluctuation exponents α depend on the normalised scattering vector Q/Q_{max}, characteristically for the two different coarsening mechanisms [1,2]. Figure 3 shows the resulting curve from our measurements of Al-12at.%Li, full circles. In order to be able to compare these findings with MC simulations we matched the simulated scattering intensity to the experimental value. Additionally, one photon was added with a probability of 50% to the simulated intensities at each time step, corresponding quantitatively to the measured uncorrelated background. The open circles in Fig. 3 represent the results for coarsening via the LSW mechanism and the triangles for coarsening via coagulation, respectively. Although the agreement is not perfect, one can attribute the measured correlation behaviour to coarsening via the classical evaporation/condensation mechanism, unambiguously.

The present study confirms the feasibility of discriminating between different coarsening mechanisms in phase-separating alloys by comparing XPCS findings with MC simulation results. The necessary scattering intensity must not be lower than in case of our Al-12at.%Li sample. Another point one has to take care of is the beam stability, which varies from refill to refill. Especially, when investigating very slow dynamics like in case of Al-9at.%Zn at –15°C this is a crucial point.

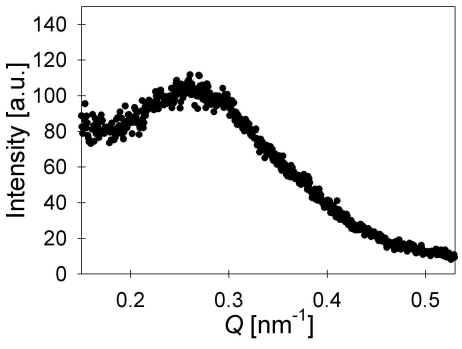

FIG. 1. Integrated SAXS intensity of Al-12at.%Li vs. the scattering vector Q. Maximum intensity at Q_{max} = 0.26 nm^{-1}.

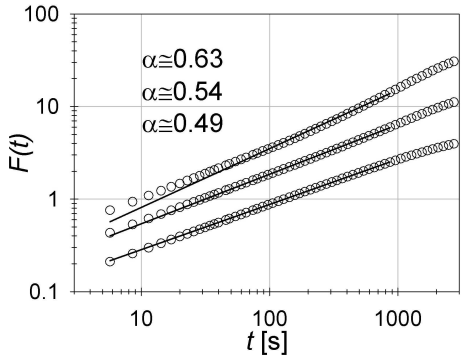

FIG. 2. Typical fluctuation functions $F(t)$ obtained from FA of fluctuating speckle intensities of Al-12at.%Li at 150°C at Q/Q_{max} = 1.0, 1.7, and 2.2, from top to bottom. Straight lines represent fits $\propto t^{\alpha}$.

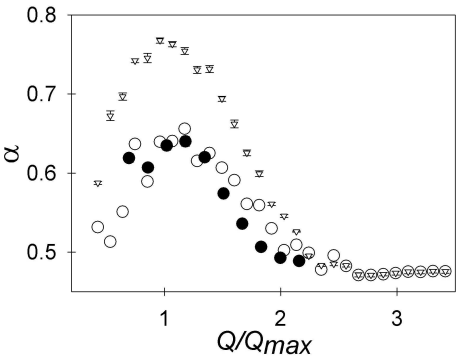

FIG. 3. Fluctuation exponents α as a function of Q/Q_{max}. Al-12at.%Li at 150°C, full circles. MC simulation results for coarsening via …LSW mechanism, open circles, …coagulation mechanism, triangles.

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