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Report:

Our recent X-ray Photon Correlation Spectroscopy (XPCS) experiment was motivated by Monte Carlo investigations of precipitate coarsening at late stages of phase separation in binary alloys [1, 2]. These studies yielded 2 possible mechanisms: The so-called Lifshitz-Slyozov-Wagner (LSW) mechanism and the coagulation mechanism. In case of the LSW mechanism coarsening proceeds via preferential diffusion of individual atoms from smaller precipitates to larger ones, whereas in the other case whole precipitates move and coalesce. The simulations showed that the coagulation mechanism can indeed play an important role in solids. Roughly, the question which mechanism plays a dominant role in a system can be answered as follows: If vacancies are preferably found in the matrix, they foster the diffusion of single atoms through the matrix. Thus, the LSW mechanism will dominate. If vacancies are localised preferably in the precipitates or on their surfaces, the mobility of whole precipitates is enhanced. This increases the importance of the coagulation mechanism dramatically – it can dominate. To get a first, rough estimation of where vacancies are found one can consider the melting temperature of each alloy's constituent [2]. If the melting temperature of the precipitating constituent is lower than that of the matrix, vacancies will rather be found in precipitates than in the matrix and vice versa.

Additionally Weinkamer [1] simulated time dependent speckle intensities for a XPCS experiment [3] in SAXS geometry for the two cases described above. By analysing speckles fluctuating intensity with Fluctuation Analysis (FA) he obtained different, characteristic results for the two coarsening mechanisms, shown in figure 1.

FA works in the following way: Starting point is a vector of N intensities I_k – each representing the intensity of a certain speckle at time step k . Subtracting the intensity average gives a new vector with the speckle's intensity fluctuations ΔI_k . In analogy to the mean-square displacement in classical diffusion theory, one calculates $F^2(t) = (Y_{j+t} - Y_j)^2$ with $Y_j = \sum_{k=1}^j \Delta I_k$ for all pairs with the same t . Doing this for all t between 1 and N a function $F^2(t)$ is obtained, which can be well fitted by $F^2(t) \propto t^\alpha$. If the intensity fluctuations I_k are

completely random, the exponent α will be exactly equal to 1. But if there are long range correlations in the data, then α will lie between 1 and 2.

In our experiment we took time series of pictures of speckle patterns with a CCD camera (Princeton Instruments, directly illuminated chip, 1242×1152 pixels, pixel size $22.5 \times 22.5 \mu\text{m}^2$) in SAXS-geometry [3]. When measuring near the SAXS fringe the exposure time was typically 1 second. To save readout time and disk space we chose broad rectangles with small heights as regions of interest. Samples were Al-6at.%Ag between room temperature and 190°C and Al-9at.%Zn at ice water's temperature. Both are systems where precipitate growth is well known [4], [5]. In both cases precipitates form spheres, coherent with the matrix. The obtained SAXS rings confirm this, see figure 2. Samples were homogenised for several hours at high temperature ($\approx 400^\circ\text{C}$), quenched into (ice) water and then annealed for a certain time at the measurement temperature before the measurement. This should ensure that measurements were done in quasi equilibrium with constant precipitates volume fraction and only very slow coarsening – in contrast to measurements on AlLi [6]. To check this, 2θ -scans were performed before and after each measurement. We saw that the shift of the SAXS peak towards smaller values of the scattering vector k due to coarsening was indeed negligible, which confirms our assumption of quasi equilibrium.

The analysis of the accumulated data was done in the following way: For each pixel lying in a 20 pixel wide ring, which corresponds to a certain k , $F^2(t)$ was calculated as described above and fitted with the function ax^α . Results for Al-6at.%Ag at 140°C and for Al-9at.%Zn at 0°C are shown in figure 3 (compare with figure 1, bottom). One clearly sees that α is varying in k much slower for Al-6at.%Ag than for Al-9at.%Zn. Also the curvatures are remarkably different. Comparison with the simulation brings us to the conclusion that the coarsening mechanism in Al-6at.%Ag is the LSW mechanism, presumably. In case of Al-9at.%Zn results indicate coarsening via the coagulation mechanism. Considering the melting temperature of each alloy's constituent – see above – one finds that this fits well together [2].

For the first time it was possible to determine the coarsening mechanism in binary alloys experimentally. This result is extremely promising for future investigations of coarsening systems.

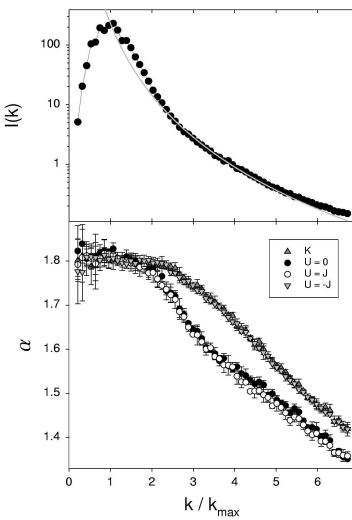


Fig. 1: Top: Simulated SAXS-profile. Bottom: calculated α vs. k/k_{max} . Triangles for LSW, circles for coagulation mechanism.

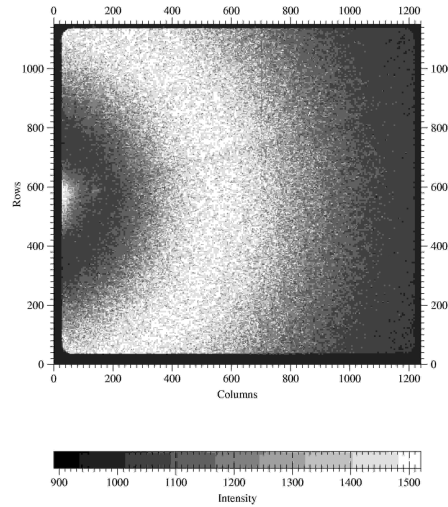


Fig. 2: SAXS spectrum of Al-6at.%Ag. The perfect ring structure indicates spherical precipitates; speckle structure is clearly visible.

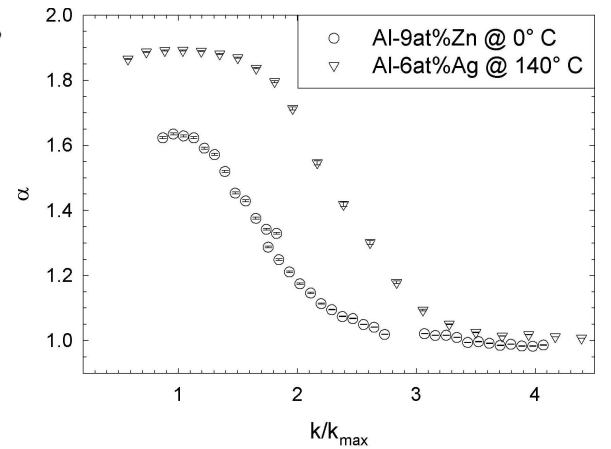


Fig. 3: Results from our measurements. Error bars (smaller than symbols) indicate the fit error. α varies much less with k in case of Al-6at.%Ag than in case of Al-9at.%Zn.

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