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Report:

As could be shown recently, different coarsening mechanisms in phase-separating alloys can be distinguished in an x-ray photon correlation spectroscopy (XPCS) experiment by analysing the fluctuating speckle intensity with the fluctuation analysis (FA) technique, gaining so-called fluctuation exponents – characteristic for long-term correlations – and comparing them with Monte Carlo simulation results [1]. It could be verified that also in bulk coarsening can take place by movement and coalescence of whole precipitates.

Spiridis and Korecki [2] did scanning tunneling microscopy (STM) studies of annealed 10 monolayers (ML) Fe films on Au(001). After annealing at 800 K formation of nanometre sized plate-like precipitates of gold in the Fe layer was observed. At higher temperatures the precipitates migrated and agglomerated, i.e., precipitates coarsened via a coagulation mechanism. Moreover, in surface science, coarsening via coalescence is a well known phenomenon observed at the growth of islands on a surface [3]. STM measurements of 5 ML Au on Fe_3O_4(001) by Spiridis *et al.* [unpublished] showed the formation of Au nanoclusters after annealing at 830 K for one hour. Additionally, a change in the cluster-height distribution and in the cluster shapes (from hexagonal to rectangular shape) were observed after 1 h at 930 K. The question arose how to follow the dynamics of these processes.

Triggered by these observations we performed XPCS measurements in GISAXS geometry of 10 ML Fe on Au(001) and 5 ML Au on Fe_3O_4(001) – both on a MgO(001) substrate – as prepared at RT and at subsequently higher temperatures under UHV conditions. A direct illumination CCD camera (Princeton Instruments, 1242 × 1152 pixels, pixel size 22.5 × 22.5 μm^2) was used as detector. For collecting conventional GISAXS diffraction patterns, see Fig.1, the camera was placed in ≈ 55 cm distance to the sample. For performing XPCS the distance was increased to ≈ 2.3 m in order to resolve the speckle structure. Time series of pictures were taken with exposure times up to 7 seconds. The speckle intensity as a function of time was analysed by FA and detrended FA [4], respectively, giving fluctuation functions $F(t)$. Analysis for a certain value of the momentum transfer $\boldsymbol{Q}$ was done for pixels lying in a 20 × 20 pixels square and then averaged. The resulting curve $\langle F(t) \rangle$ was fitted by a power law $\propto t^{\alpha}$, yielding so-called fluctuation exponents α. Figure 2 shows results for measurements of 10 ML Fe on Au(001) at 723 K after precedent heat treatments up to 873 K, i.e., where Au precipitates should have been present in the iron film. $\alpha > 0.5$

indicates the presence of long-term correlations, similar as were seen for bulk measurements of binary phase-separating alloys undergoing coarsening [1]. Additionally, a certain ***Q*** dependence is recognisable which demonstrates the feasibility of the applied method for investigations of coarsening dynamics on surfaces.

Analysing the XPCS data of the measurements at 5 ML Au on Fe_3O_4 turned out to be much more complicated because dominating periodic correlations with a period of $\approx$ 410 s (derived from the power spectrum of the intensity fluctuations) were observed from RT up to 723 K. This periodic signal can be attributed to variations of the beam position, which was found to be differently stable from refill to refill. At temperatures higher than 723 K the dynamics in the sample dominates the correlation behaviour, as can be seen even when the intensity fluctuations are analysed conventionally by calculating their autocorrelation functions, Fig. 3. This proves the feasibility of applying XPCS to the problem of nanocluster formation on metal oxides. In order to obtain quantitative results it is necessary to have a more stable beam.

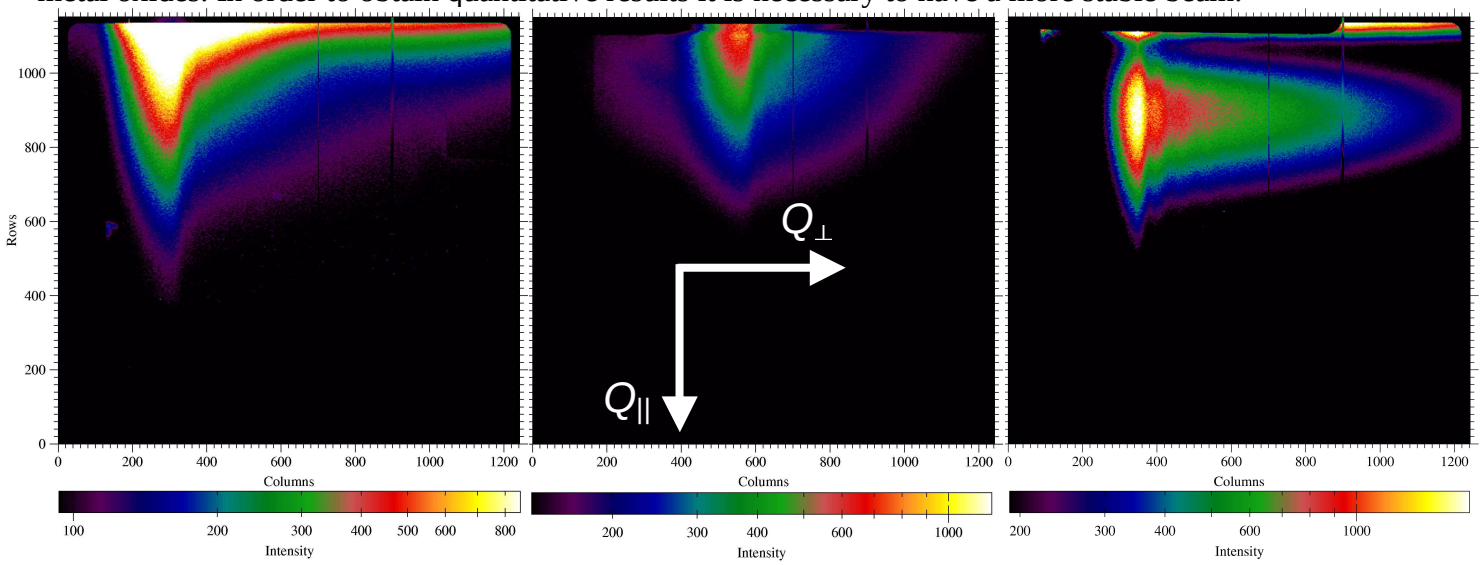

FIG. 1. GISAXS diffraction pattern with logarithmic intensity scale. Momentum transfer in the sample plane, $Q_{||}$, normal to the sample plane, $Q_\perp$. Left: Au precipitates in 10 ML Fe on Au(001) after heat treatment; beam parallel to MgO(001). Grazing incidence angle α_i=0.45°. Middle: Same sample, but beam parallel to MgO(011). α_i=0.45°. Right: 5 ML Au on Fe_3O_4 before heat treatment. α_i=0.3°. Here, the GISAXS signal is caused by lateral correlations in the Fe_3O_4 substrate. The derived correlation length is $\approx$ 19.5 nm.

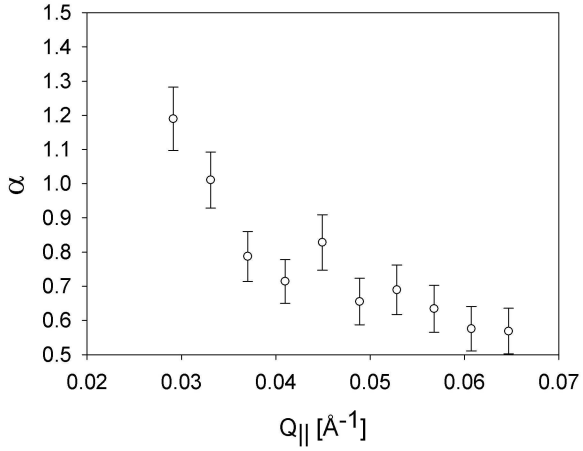

FIG. 2. Fluctuation exponents α for Au precipitates in 10 ML Fe on Au(001) at 723 K. $Q_\perp$=0.059 Å^{-1}.

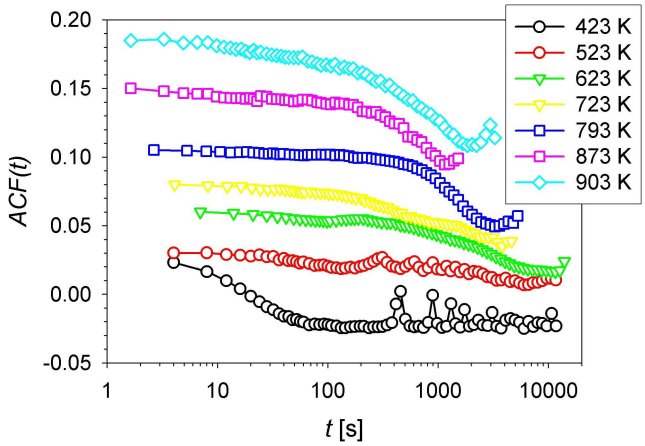

FIG. 3. Autocorrelation functions for different temperatures for 5 ML Au on Fe_3O_4, shifted along the *y* axis for clarity.

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