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Names and affiliations of applicants (* indicates experimentalists):		
*L.-M. STADLER ¹ , *B. SEPIOL ¹ , *M. SLADECEK ¹ , *M. RENNHOFER ¹ , *M. HARTMANN ^{1,2} , G. VOGL ¹ , G. GRÜBEL ³		
¹ Institute of Materials Physics, University of Vienna, Strudlhofg. 4, A-1090 Wien, Austria		
² Erich Schmid Institute of Materials Science, Austrian Academy of Sciences & Institute of Metal Physics, University of Leoben, Jahnstr. 12, A-8700 Leoben, Austria		
³ European Synchrotron Radiation Facility, F-38043 Grenoble Cedex, France		

Report:

If coherent light is scattered at a disordered sample the diffraction pattern will be highly modulated (speckle pattern). If the sample evolves in time also the speckle pattern will. Analysing the intensity fluctuations of a single speckle provides information about timescales on which the dynamics in the sample takes place. This method using coherent X-rays is called X-ray Photon Correlation Spectroscopy (XPCS).

Single crystals of binary alloys crystallizing in the B2 (CsCl) structure contain antiphase domains [1]. Information about the dynamics of antiphase domains/boundaries can be obtained in an XPCS experiment. This was the result of our first experiment with Co₆₀Ga₄₀ (B2 structure) [2].

In the present experiment we have chosen an Fe₆₅Al₃₅ single crystal (also B2 structure) because information about dynamics of its antiphase domains is available in literature [1]. The (001) superstructure and the (011) fundamental Bragg peaks have been investigated at different temperatures between RT and 1000K with a CCD camera (Princeton Instruments, directly illuminated chip, 1242 × 1152 pixels, 22.5 × 22.5 μm² pixel size). For a typical picture see Fig. 1. Dynamics of antiphase domains should be visible only in the superstructure reflection [3]. It means that we have a possibility to verify whether effects of antiphase domains motion are measured or not. Temperature calibration was made for the surface of the sample measuring the central shift of the Mössbauer line.

Bragg peaks were exposed 1s/frame resulting in ≈1.4s repetition time. The average centre of mass of the intensity in the frames was calculated and used as the centre of circles defining pixels for which intensity fluctuations were analysed. This was done first by calculating the auto-correlation function C(τ) of the time vector of intensities [2]. It was obvious that C(τ) were not simple exponentials. The reason for this behaviour is that the position of the peak varies with time at constant temperature feeding in spurious “correlations”. We suppose that the variation of the peak position can be attributed mainly to small angle grain boundary (GB) movement. The effect of variation of the lattice parameter due to temperature instabilities can be excluded. In our case the temperature was stable to at least ± 2K.

In order to check if we can disentangle the effects of fluctuations resulting from GB and from antiphase boundaries Fluctuation Analysis (FA) [4] and Detrended Fluctuation Analysis (DFA) [5] were applied. Doing FA one calculates for all speckle intensity pairs with the same time-lag *t* a function

$F^2(t) = (Y_{j+t} - Y_j)^2$, where $Y_j = \sum_{k=1}^j \Delta I_k$ with ΔI_k being the fluctuation of the intensity in one pixel at time-step k . For completely random fluctuations $F(t)$ is proportional to t^α with $\alpha=0.5$ (random-walk problem without correlations). If short range correlations with a correlation time τ_c are present, $F(t)$ will be proportional to t^α with $\alpha \approx 1.0$ roughly up to $t=\tau_c$ and for longer times to $\alpha=0.5$ again. If there are non-stationary processes (trends) in the data the situation is more complicated and FA is useless because correlations will be shaded by these trends. Application of DFA n [5] with n being the order of the method is indispensable. DFA1 for example removes piecewisely constant trends in the raw data arising from, e.g., flux fluctuations of the primary beam. DFA2 removes linear trends in the raw data, DFA3 quadratic trends and so on. Fig. 2 clearly shows that spurious trends, that could suggest correlations on the basis of FA or auto-correlation function in the sample at RT, can be removed. Differences between fluctuations measured at elevated temperatures in the fundamental (001) and in the superstructure (001) Bragg directions are shown in Fig. 3 and can be interpreted in the following way: Measurements in the fundamental direction, where no antiphase effects are expected, show short-range correlations which we attribute to small-angle grain boundary motion. Relaxation times for these correlations are slightly below 100s (notice the steeper increase of the $F(t)$ function for shorter times). Opposite to measurements of the fundamental peak, trends, that manifest themselves in crossovers, are much more pronounced for the superstructure (001) peak and cannot be removed even by the DFA3 analysis. Also a definite temperature dependence of the crossover time can be recognized. We attribute the trends in the (001) direction to the antiphase motion in the sample out of the thermal-equilibrium state. Furthermore, short-range correlations caused by small-angle grain boundary motion seem to be shaded by these stronger trends in Fig. 3 (left). More quantitative results are not attainable without extensive Monte Carlo simulations.

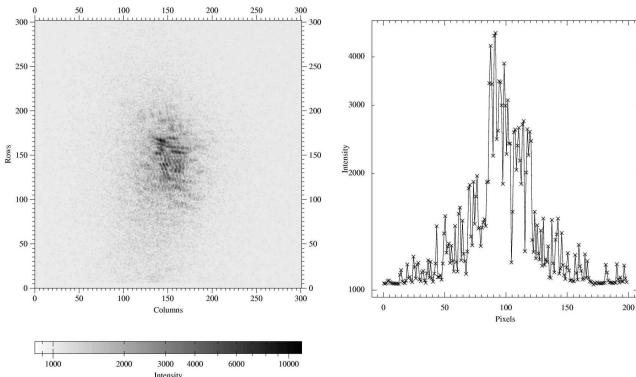


Fig. 1. (left) (001) reflection of $\text{Fe}_{65}\text{Al}_{35}$ single crystal at 500°C. Speckle structure is clearly visible; (right) slice across the Bragg reflection illustrating the effect of coherent illumination - highly structured intensity profile.

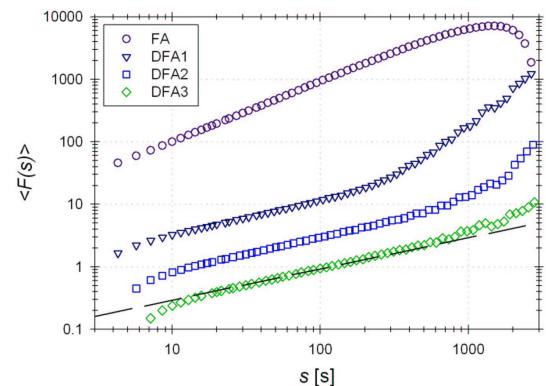


Fig. 2. Application of FA, DFA1, DFA2 and DFA3 analysis to remove trends from intensity fluctuations of the (001) reflection at RT. The slope of the dashed line in this and in the next figure is exactly 0.5.

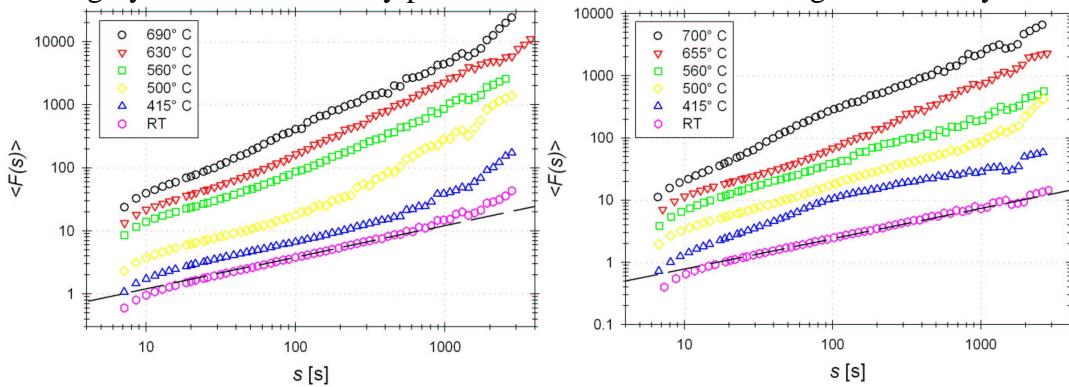


Fig. 3. (001) and (011) reflections (left and right sides respectively) at comparable temperatures analysed with DFA3. For better visibility curves were shifted in y-axes direction.

- [1] S.M. Allen and J.W. Cahn, *Acta Metall.* **27**, 1085 (1979).
- [2] L.-M. Stadler et al., ESRF experimental report HS-1275 (2000); M. Sutton et al., *Nature* **352**, 608 (1991).
- [3] F. Bley, F. Livet, J.C. Leroux and J.P. Simon, *Acta Cryst. A* **51**, 746 (1995).
- [4] L.-M. Stadler et al., ESRF experimental report HS-1578 (2002).
- [5] J.W. Kantelhardt et al., *Physica A* **295**, 441 (2001).