Coherent X-ray Study of Fluctuations During Domain Coarsening [1]

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In recent years, increasing theoretical attention [2] has been focused on the physics of fluctuations during domain coarsening. It has been predicted that such fluctuations differ from classical fluctuations, in that they are (almost) "frozen in" to each instance of a coarsening system, leading to domain arrangements at later times which are strongly correlated with those at earlier times. To test these ideas we have used X-ray intensity fluctuation spectroscopy (XIFS) to observe the dynamics of the exact structure factor (speckle pattern) during phase separation in a sodium borosilicate glass. In the past few years XIFS has been demonstrated in studies of equilibrium dynamics such as fluctuations near a critical point and diffusion in colloids and micellar systems [3]. In this study we apply the technique to a non-equilibrium process, domain coarsening during phase separation.

The XIFS experiments were performed in a small-angle scattering geometry at the IMMCAT beamline 8-ID-A. In order to obtain a sufficiently intense coherent X-ray beam, the full 2.5% energy bandwidth of the undulator first harmonic was used [4]. A detailed description of the production and characterization of X-ray speckle using this beamline was reported earlier [5]. In a typical experiment, the sample was heated to $T = 1033$ K to equilibrate it in the single-phase state, and then rapidly quenched to and held at a temperature below $T_c = 1026$ K so that phase separation occurred isothermally. During this process, scattering patterns were recorded using a CCD camera at a rate of one frame per 1.24 s.

To analyze the fluctuations in the exact structure factor we use a two-time correlation function, defined by

$$C(q,t_1,t_2) = \frac{\langle I(t_1)\,I(t_2)\rangle - \langle I(t_1)\rangle\langle I(t_2)\rangle}{(\langle I^2(t_1)\rangle - \langle I(t_1)\rangle^2)^{1/2}\,(\langle I^2(t_2)\rangle - \langle I(t_2)\rangle^2)^{1/2}}. \quad (1)$$

Here $I(t)$ is the intensity in a pixel at time t normalized to the total intensity. Since $\langle I\rangle$ is isotropic, ensemble averages were taken over pixels having the same q. A typical two-time correlation function for a specific wavenumber is shown in Fig. 1. The coordinate along the direction of the central ridge is the average time after the quench $\bar{t} \equiv (t_1 + t_2)/2$. If one makes a cut across this ridge along the time-difference coordinate $\Delta t \equiv t_1 - t_2$, the width of the peak gives the correlation time at that $\bar{t}$. For an equilibrium system, this width would be constant as a function of $\bar{t}$. From Fig. 1, it is clear that the width increases with $\bar{t}$. The correlation time τ was evaluated for each $\bar{t}$ by determining the value of $|\Delta t|$ which gives a correlation value $C = 0.5$. The resulting $\tau(\bar{t})$ at each q are shown in the inset in Fig. 2. The correlation times
increase with time at each wavenumber, and decrease with wavenumber. In addition, τ is always a large fraction of $\bar{t}$ indicating that the speckles are persistent.

Simulations of domain coarsening using a Langevin model [2] have found that τ follows a scaling law, which we write as

$$\tau(q,t) = (t_{max}(q) - t_0)W_2(x), \quad (2)$$

where t_{max}, t_o, and $x \equiv (t - t_0)/(t_{max} - t_0)$ are determined by the scaling behavior of the average structure factor $\langle I\rangle$. The scaling function $W_2(x)$ is found to have a power-law form $W_2 = ax^p$ with the exponent $p = 1 - n$ [2]. Figure 2 shows this function W_2 obtained by normalizing the experimental correlation times by $t_{max} - t_0$. The scaled correlation times at both temperatures fall along the same curve, indicating that proposed scaling works for this system. To test the prediction for W_2, the experimental data were fit to the power law form, giving $a = 0.72 \pm 0.02$ and $p = 0.65 \pm 0.04$. The value of p agrees with the prediction of $1 - n = 2/3$. The near-unity value of a indicates that the speckles are quite persistent, and agrees within a factor of two with simulation results.

The observed scaling of the correlation time at both temperatures provides the first experimental confirmation of the scaling law (2) for τ. These results demonstrate the persistence of speckles during domain coarsening, behavior which differs qualitatively from that previously observed with XIFS in equilibrium systems. We believe that the two-time correlation function measured by XIFS can be a useful quantitative probe for understanding the dynamics of non-equilibrium systems.

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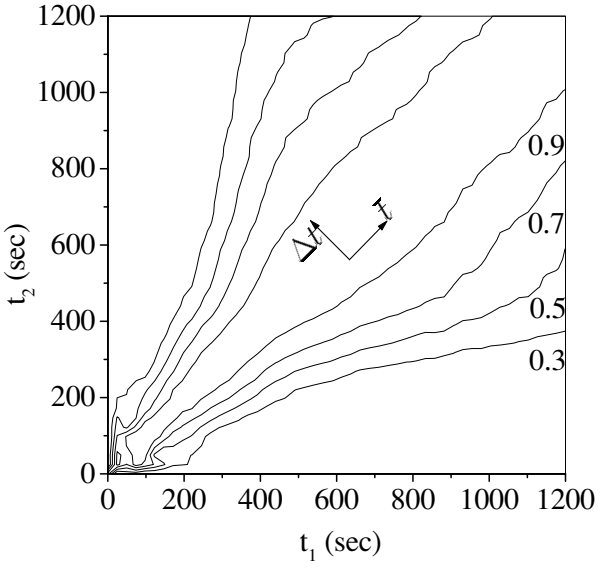

Figure 1: Contour plot of two-time correlation function at q=0.01 Å for the 963 K quench. Values of C for each contour are shown. Directions of the alternative coordinates $\bar{t}$ and Δt are indicated.

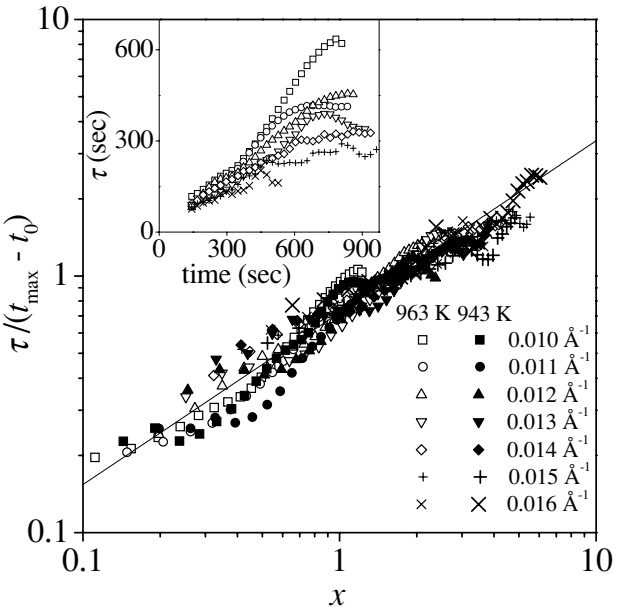

Figure 2: Scaled correlation times for quenches to 963 K and 943 K, plotted versus $x \equiv (t - t_0)/(t_{max} - t_0)$. The line is a power-law fit to the data. The unscaled correlation times for the 963 K quench are shown in the inset.

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