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Ordering fluctuation dynamics in AuAgZn$_2$

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The second-order ordering transition of the AuAgZn$_2$ alloy has been studied by coherent x-ray scattering. Within a few degrees above the critical temperature $T_c$, equilibrium critical fluctuations are observed together with some pre-transitional local ordering connected to sample defects. The speckles observed correspond to heterodyne interference between local ordering and fluctuations and show a mixed static and dynamical behaviour in a narrow domain of a few tenths of degree above $T_c$. The dynamical behaviour is shown to correspond to the critical slowing down of the fluctuations in the vicinity of the transition (model “A” of Hohenberg and Halperin$^1$). A rough comparison can be carried out with the classical diffusion models. Some improvements of the method are discussed.

I. INTRODUCTION

When a system is close to the critical point, anomalies occur in its static properties, like the divergence of the fluctuation length $\xi$ or of the susceptibility $\chi$. For systems belonging to the Ising universality class, a large number of experimental studies could be compared with numerical simulations, with the results of series expansion$^2$ and with field theoretical models$^3$. The dynamics of these fluctuating systems where the microscopic elementary process is the atom spin flip exhibits at the transition a “critical slowing down”, corresponding to the divergence of $\xi$, which is much more difficult to experimentally observe. In a simple diffusion model, the characteristic time $\tau$ of the fluctuations should also be divergent at the transition. This model of the dynamics of the transition with “non conserved order parameter”, the “A” model of Hohenberg and Halperin$^1$, see also Ref. 4, introduces a dynamic exponent $z$, which connects $\tau$ to the fluctuations by the following expressions:

$$
\begin{align}
\tau(q, T) & \propto q^z & \text{for } q \ll \xi^{-1} \\
\tau(q, T) & \propto q^{-2} & \text{for } q \gg \xi^{-1}
\end{align}
$$

where $q$ corresponds to the wavevector. Dynamic renormalization group calculations$^5,6,7$ as well as Monte Carlo simulations$^8$ suggest that $z \approx 2$.

The order-disorder transformations taking place in the ternary body centred cubic AuAgZn$_2$ alloy have been extensively studied in the 80’s in order to determine the thermodynamics and the kinetics associated with the stabilized phases. Close to the critical temperature $T_c \approx 330^\circ$C, the AuAgZn$_2$ Heussler alloy exhibits a transition between L$_21$ and B2 structures represented in Fig. 1$^9,10$. The transition corresponds to a change of the ordering state in the Au-Ag sublattice leading to a doubling of the unit cell lattice parameter. Above $T_c$, the two chemical species share a simple cubic lattice. Below $T_c$, Au and Ag atoms are preferentially located on their own face centred cubic lattices. As the development of ordering does not need a long distance transport of atoms, this alloy can be considered as a model for the transition with “non conserved order parameter”, i. e. the “A” model of Hohenberg and Halperin$^1$.

In a previous paper,$^{11}$ the B2→L$_21$ transition was studied with incoherent X-ray scattering measurements close to the $\overline{111}$ Bragg reflection of the B2 structure. The static critical behaviour was investigated with isothermal acquisitions above $T_c$ to probe the thermodynamical equilibrium. The data analysis has shown a well characterized second-order transition with a transition temperature close to $350^\circ$C (varies slightly depending on Zn concentration) and the Ising-like behaviour of the alloy has been confirmed by the values of the critical exponents deduced from the temperature dependence of the intensity profiles. The dynamics of the order-disorder transition was also studied from acquisitions during quenches of the sample. When quench temperatures are well below the critical temperature, the dynamics is mainly associated with the motion of interfaces between ordered domains. The time dependence of the characteristic domains size $L$ was shown to follow the $L(t) \sim t^\frac{1}{3}$ growth law, which applies for non-conserved

FIG. 1. (a) L$_21$ and (b) B2 ordered structures of the AuAgZn$_2$ alloy below and above $T_c$, respectively. The lattice parameter $a$ ($a_0$ in the text) of the B2 phase is close to 3.17 Å at 330$^\circ$C.
dynamics far from $T_c$ (see e.g. Ref.12). From the equation:

$$L^2 = Dt$$  \hspace{1cm} (2)

the diffusion constant $D$ at the critical temperature was estimated to $2.4 \times 10^3 \text{ Å}^2 \text{s}^{-1}$ leading to a microscopic diffusion time of $\tau_0 \approx 40 \mu \text{s}$ between two sites of the Au-Ag simple cubic ($a_0 = 3.2 \text{ Å}$) sublattice (see Fig. 1b). When quenches temperatures are in the vicinity of the critical temperature, the dynamics in a limited time interval is dominated by the time evolution of the critical fluctuations of size $\xi$. From our estimate of $\tau_0$, the fluctuation time $\tau$ should be roughly 1s if $\xi \approx 500 \text{ Å}$ by using a formula similar to Eq.2, with $L$ replaced by $\xi$: $\tau \approx \tau_0 (\xi/a_0)^2 \approx 40.10^{-6} \times (500/3.2)^2$. This is a simple diffusion model (i.e. which assumes $z = 2$). From the asymptotic behaviour $\xi \approx a_0 (T - T_c)/T_c \propto T - T_c$, $\xi$ is larger than 500 Å only for $|T - T_c| < 0.2^\circ\text{C}$ (see Eq. 11 of Ref. 11).

As the elementary process for local ordering is the exchange between neighbouring cubic Au/Ag sites, this system is considered as equivalent to the model “A” of Ref.1.

The experimental observation of the dynamics of the critical fluctuations is a real challenge since heating devices providing high temperature stability, very good accuracy, fast cooling rates as well as small undercooling during quenches are required. A specific water-cooled vacuum chamber with a maximum deviation of 0.01$^\circ\text{C}$ for a fixed temperature between 280$^\circ\text{C}$ and 365$^\circ\text{C}$ and a maximum cooling rate of 2$^\circ\text{C}$s$^{-1}$ was designed for this purpose and used in our previous study to observe the establishment of the critical fluctuations after quenches. Data analysis in Ref 11 has revealed that 50 s were necessary for fluctuations to have a 500 Å correlation length after quench, which was significantly different from the previous estimate of one second and thus not trustworthy. Further measurements in the AuAgZn$_2$ system at the European Synchrotron Radiation Facility (ESRF) and the Diamond Light Source (DLS) showed that only the temperature equilibration at long time scales was observed. The reason was found in the poor thermal contact of the ceramic glue with the heating element under vacuum resulting to inertia effects. Because of too slow quenches, the dynamic critical behaviour of the AuAgZn$_2$ ternary alloy could not be fully characterized in Ref 11 and a reliable comparison with the theoretical predictions is still missing. In the present study, the quality of the thermal contact is improved by inserting a liquid metal drop between the heating element and the samples.

Nowadays, high fluxes of synchrotron sources and high resolutions and counting rates of 2D detectors enable to perform time-resolved diffraction experiments with coherent x-ray beams.\textsuperscript{13} This technique called x-ray photon correlation spectroscopy (XPCS) gives rise to new insights into the structure and its dynamics. In coherent scattering experiments, disorder leads to speckles in the measured scattering intensities\textsuperscript{13}, and the observation of changes in the speckle pattern reflects the microscopic time evolution of the fluctuations.

Pioneering experiments on the dynamics of critical fluctuations have been realized by Brauer et al\textsuperscript{14} in the Fe$_3$Al binary alloy which exhibits a B2-DO$_3$ second-order transition similar to the B2-L2$_1$ transition investigated in this paper.

In this paper, the x-ray photon correlation spectroscopy technique is used to improve the understanding of the dynamics of critical fluctuations and to determine more reliable estimates of their time correlation functions. Difficulties belonging to this approach are also discussed.

The paper is organised as follows. The first part is devoted to the technical description of the experimental setups, to the characterization of the sample probed by the x-ray beam and to the determination of the degree of coherence of the experiment. In the second part, experimental results are analysed in terms of domain structure, critical temperature assessment and correlation time. The comparison with the theoretical predictions is then presented before concluding remarks.

II. EXPERIMENTAL DETAILS

A. XPCS setup

Most of the results presented here were obtained from dynamic experiments carried out at the Advanced Photon Source (APS), Argonne National Laboratory on the 8ID-G beamline. A few preliminary results were obtained at the I16 beamline of Diamond Light Source (DLS). We used a monochromatic beam of wavelength $\lambda = 1.687 \text{ Å}$ (APS) and 1.55 Å (DLS) obtained with Si(111) monochromators. At APS, the flux from the undulator was first low-pass filtered by a small mirror 20 m upstream our setup, and at DLS, mirrors were used for vertical focusing and as low pass filters. The transverse beam partial coherence was achieved by slits opened to $20 \times 20 \mu\text{m}$ upstream the samples. The incident flux at the sample was measured to be $1.8 \times 10^9 \text{ ph/s}$ at the APS and $2.6 \times 10^9 \text{ ph/s}$ at DLS, where pre-focusing optics are used. The scattered intensity was measured with direct illumination CCDs (DI-CCD) from Princeton Instruments. A camera with $1300 \times 1340$ active pixels of 20 $\mu\text{m} \times 20 \mu\text{m}$ size was positioned at 2 meters downstream the sample at APS and a camera with $384 \times 576$ pixels of 22 $\mu\text{m} \times 22 \mu\text{m}$ size was placed at 2.75 meters at DLS. At the selected wavelengths, the efficiency of the detectors is in the 45-55% range. As these DI-CCDs saturate for about 100 x-rays per pixel, experiments were always carried out with multiple frames, with some dead time between frames. In the case of DLS, the dead time was 0.27 s. In the case of the APS measurements, the dead time between two frames was 1.9 s for full frame
FIG. 2. \( \{1 \overline{1} 1\} (Q_0/(2 \times \pi)) \) Bragg peaks observed at room temperature for (a) a polished sample with an ellipsoidal intensity connected to surface mosaicity (elongation in the \( q_y \) direction) and (b) the same sample after disordering and quench showing the isotropic distribution of antiphase boundaries (DLS experiment). \( \vec{q} = |(\vec{Q} - \vec{Q}_0) \) has two components: \( q_x \) (nearly parallel to \( \vec{Q}_0 \)) and \( q_y \) (perpendicular to \( \vec{Q} \)).

(1.7 Mpixels at a 1Mhz frequency). For a 1 s exposition time, the period of \( \approx 2.9 \) s was too slow to observe fluctuation dynamics, and the dynamical measurements were carried out with a 300 x 300 pixels ROI, which limited the dead time to 0.3 s. The “droplet algorithm”\(^{15} \) was systematically used for photon counting. If necessary, the x-ray intensity was damped with filters. All the measurements were carried out on the \( \{1 \overline{1} 1\} (=Q_0/(2\pi)) \) superstructure reflection, which appears owing to the doubling of the unit cell in the ordered phase. In the vicinity of the Bragg position, \( \vec{q} = \vec{Q} - \vec{Q}_0 \) is the relevant scattering vector. At the critical temperature, the Bragg angle is \( \theta_B = 13.32^\circ \) for \( \lambda = 1.687 \text{ Å} \) and the x-ray penetration depth in the sample is about 1 \( \mu \text{m} \). The reflection was measured in nearly symmetrical geometry, except for a surface miscut of 2.3\( ^\circ \) (only for the APS experiment) which was accommodated in the scattering plane. The experimental stability of the setups was confirmed using the static speckle structures from low temperature quenched samples. No significant changes were observed in the structure of the static speckle of the \( \{1 \overline{1} 1\} \) superstructure reflection, which confirmed the good stability in the half hour range at both beamlines.

B. Sample features

Our samples are crystals cut from a single crystal rod 8 mm diameter grown in a vacuum silica ampule, slowly cooled from liquid to 400\( ^\circ \text{C} \) and then cooled down to room temperature. This (relatively) fast cooling (\( \approx 2 - 3^\circ \text{C/s} \)) leads to a pattern of antiphase ordered domains. Samples were then cut with surface normals close to a <111> crystallographic direction, and carefully polished with diamond powder and silica gels.

Experiments carried out at DLS have shown that under vacuum, the surface losses of Zn lead to an inhomogeneous lowering of \( T_c \) in the 1 \( \mu \text{m} \) depth probed by x-rays. Thus, for the measurements at the APS, a 80 Å thick aluminum layer has been deposited by sputtering and the resulting \( \text{Al}_2\text{O}_3 \) layer acts as an efficient chemical barrier.

Fig. 2(a) presents a typical scattering map recorded at room temperature for initial microstructures (as-polished samples). The scattering intensity appears ellipsoidal. If samples are then heated above \( T_c \) and rapidly cooled down, the enlarged Bragg peak has a circular symmetry as shown in Fig. 2b. In both cases, speckles are observed. The anisotropy observed in Fig. 2a is thus connected to mosaicity caused by the sample polishing yielding scattering elongated along the \( q_y \) direction. During this process, a large amount of dislocations are introduced at the sample surface. For temperatures higher than 280\( ^\circ \text{C} \), most of this surface dislocation structure disappears and

FIG. 3. Rocking curves obtained for two neigbouring positions of the sample (20 \( \mu \text{m} \) transverse displacement). Plots from the centers of the peaks show two different values of FWHM: 0.0046\( ^\circ \) and 0.0079\( ^\circ \), and some extra intensity corresponding to defects. The change in the position of the maximum is connected to mosaicity.
a rapid growth of the L2₁ ordered domains is also observed. After some aging, a nearly monodomain sample is obtained, but mosaicity can not be entirely removed. This is illustrated for a sample at 336 °C served. After some aging, a nearly monodomain sample is observed at 336 °C. The study of speckles at large angles has to take account of all lattice defects, as it was observed from dislocations, or even from surface steps. Experience with metals shows that perfect crystal grains rarely exceed a few micrometers except if annealed for hours close to the melting temperature. In this study, the Zn vapor pressure is too high to carry out a high quality restoration. Consequently, our measurements are sensitive to surface dislocations which move and form subgrain boundaries.

C. Speckle contrast

Coherence can be checked from the isotropic scattering of a quenched sample. Fig. 4 shows the scattering close to Q₀ observed at APS after a quench to 75°C. With the hypothesis that no privileged direction in the crystal leads to a systematic anisotropy of the antiphase configuration, the contrast β of our experiment can be estimated from the angular mean square fluctuations of the intensity averaged across rings centered at the Bragg position:

$$\beta(q) = \frac{\langle |I(q)|^2 \rangle_{|q| \in \Delta} - \langle |I(q)| \rangle_{|q| \in \Delta}^2}{\langle |I(q)| \rangle_{|q| \in \Delta}^2}$$

where Δ are domains delimited by circles of increasing radius $q: q - \delta q/2 < |q| < q + \delta q/2$. This equation includes a correction for Poisson counting statistics.

III. RESULTS

A. Domain structure

The average intensity of Fig. 4 is represented in figures 5a and 5b for two q ranges. The Porod’s plot, shown in Fig. 5c, highlights the $q^{-4}$ asymptotic behaviour of the intensity, thus demonstrating the presence of well defined domains, separated by antiphase walls. The speckle contrast calculated with Eq. 3 is represented in Fig. 5(d) for different values of q. In the case of low (i.e. less than one count par pixel) intensity, the Poisson noise is significantly larger than the speckle contrast. A measurable value of β can nevertheless be obtained, owing to the large number of pixels included in the corresponding domains. By averaging the results between the 6 sets of 50 1s frames, we estimate $\beta = 0.075 \pm 0.01$.

A detailed discussion of the speckle contrast is out of the scope of this paper, but this (relatively) low value is explained by the “large” beam size (20 μm) used, by the poor monochromativity of the beam (δλ/λ ≈ 0.00014) with the Si₁₁₁ monochromator, combined with the significant (≈ 1 μm) beam penetration depth. An important contribution to this low contrast is also the asymmetry of the scattering geometry, due to the 2.3° miscut of the surface with respect to Q₀.
The origin of the intensity of Fig. 6, we fitted this asymptotic behaviour. Fig. 7 shows the results of the fit when applied to the scattering of Fig. 6. The fit shows the two regimes of the scattering: for $q > 0.003 \text{ Å}^{-1}$, the fluctuations are dominant, and no speckles are observed. For $q < 0.003 \text{ Å}^{-1}$, the central peak is dominant, it is stable, with visible speckles and its intensity has a $q^{-4}$ behaviour. This means that the pre-transitional peak corresponds to stable limited ordered domains, probably close to the surface.

This “central peak” corresponds to pre-transitional ordering. Moving the sample in the beam modifies its relative weight and shape, but it seems difficult to get rid of it. This behaviour is observed in numerous systems exhibiting second-order transition. It is generally connected to surface defects or surface strain.

### B. Critical temperature

The “double peak” analysis of Eq.4 provides for each temperature an estimate of $S(q=0)$ and of the correlation length $\xi$. These can be used for the determination of the critical temperature from:

$$S(q=0) = A(T - T_c)^{-\gamma} \quad \text{and} \quad \xi = C(T - T_c)^{-\nu}$$

where the standard static critical exponents in the 3d Ising system ($\gamma = 1.241, \nu = 0.631$) are used. Fig. 8 shows the results of the fits. We estimate $T_c \simeq 336.18 \pm 0.01^\circ\text{C}$.
C. Heterodyne observation of dynamic critical fluctuations

The two components of the scattering in Eq. 4, as visible in Fig. 6, have different origins: the small ordered domains are essentially stable, leading to observable speckles and the diffuse scattering exhibits no speckles because of short-time fluctuations. The relative weight and shape of the central peak is dependent on the beam position at the sample, but its intensity is strongly temperature dependent.

For the observation of the dynamics, the measuring time was reduced by choosing a 300×300 pixels ROI. Dynamical results were obtained with 0.2 s (0.5 s total period) and 0.8 s acquisition time (i.e. 1.1 s sampling period).

The standard method is to obtain a doubly-averaged normalized correlation function \( g(q, t) \) from the measured intensity \( I(\vec{q}, t) \):

\[
g(q, t) = \frac{\langle (I(\vec{q}, t + t') \times I(\vec{q}, t)) e^{i\vec{q} \cdot \vec{v}} \rangle_{\vec{q} \in \Delta}}{\langle |I(\vec{q}, t)|^2 \rangle_{\vec{q} \in \Delta}}
\] (6)

The first average, over the time \( t' \) yields a classical time correlation, of poor statistics, and the second average is carried out in the \( q \) circular domains \( \Delta \), assuming isotropic dynamics.

For short time scales, we observe interference between the stable “central peak” and the fluctuating intensity of the diffuse scattering. This interference has already been observed between a polymer sample and a reference\(^2\), and if one assumes identical coherence factor \( \beta \) for the two scatterer, the speckle dynamics can be written:

\[
g(q, t) = 1 + \beta ((1 - x)^2 + x^2 \gamma^2(q, t/\tau) + 2x(1 - x)\gamma(q, t/\tau))
\] (7)

where \( x = \langle (I_c) \rangle / \langle (I) \rangle \) is the mixing, i.e. the part of the intensity connected to the critical fluctuations \( I_c \). \( \gamma(q, t/\tau) \) is the \( q \)-dependent correlation function, which here will be assumed not to differ essentially from an exponential:

\[
\gamma(q, t) = \exp(-t/\tau(q))
\] (8)

In our experiment (see Fig. 6), mixing can be fairly small, and carrying out fits to experimental curves results in \( x \) varying from a few percent in the central part to 0.3. This means that the second order term (in \( x^2 \)) of Eq. 7 is nearly negligible.

Typical results are shown in Fig. 9. These data were obtained at 336.25°C, from 5,000 frames of period 1.1s. Averages were carried out in circles 6 pixels wide, after careful discarding of the mosaic scattering observed in the detector (see Fig. 6). The results of the fits with Eq.7 are shown in the figure. Best estimates of \( \tau \) were obtained from the measurements displayed in Fig. 9, where \( |T - T_c| \approx 0.07°C \). The results for shorter sampling time or for higher temperatures appear somewhat imprecise. In Fig. 10 are given the results of all fits carried out at 336.25°C. Results become unreliable for \( q > q_{\text{max}} \approx 0.002 \ \text{Å}^{-1} \).

In Fig. 10, \( q^2 \times \tau(q) \) is also plotted. In the case \( z = 2 \) is considered valid, this product is constant for \( q \gg \xi^{-1} \). From Fig. 11 of Ref. 11, we estimate \( \xi \approx 1000 \ \text{Å} \) for \( T - T_c \approx 0.07°C \), and though \( q_{\text{max}} \) is not significantly larger than \( \xi^{-1} \), we estimate that this product is close to \( 7 \times 10^{-6} \ \text{sÅ}^{-2} \) for large \( q \) values. This result can be compared to the observed size increase of the sample after quench. In Fig. 11 is plotted the evolution of the characteristic dimension \( L \) of the domains. In this figure, \( L \) is obtained from the inverse of the half width at half maximum (HWHM) of the scattering observed, different form Eq. 2. In the case of the squared Lorentzian approximation (Eq. 4), \( L = L/\sqrt{2 - 1} \). In Fig. 11 a new estimate of the diffusion constant at 334°C is given: \( D = 4.9 \times 10^5 \ \text{Å}^2/\text{s} \). This result is consistent with the previous estimate \( (D \approx 2.4 \times 10^5 \ \text{Å}^2/\text{s}) \) (see Eq. 2 in Ref.
one temperature in the very vicinity of $T_c$, which gives a fluctuation time in the $1-20$ s range. It is derived from the ordering kinetics. As in a "post mortem" study, the diffusion constant in the transition vicinity (at about $612^\circ$C) was estimated to $3.2 \times 10^{-17}$ m$^2$/s. This means an atomic time scale $\tau_0$ of $\approx 2 \times 10^{-3}$ s, 50 times slower than in AuAgZn$_2$. This value is in agreement with Ref. 23 where a different definition of the diffusion constant is used. For large fluctuations, the fluctuation time can be in the $100$ s range. This was studied in Ref. 24 by selecting a volume sample of a few $\mu$m$^3$, and by the observation of the scattering at the superstruture Bragg peak position. The results "at $T_c$" were interpreted as critical fluctuations. The fluctuation time observed "at $T_c$" was found to be $70$ seconds, in agreement with the above estimate.

This is an important remark for future XPCS experiments: when studying large scale dynamics, the microscopic time scale $\tau_0$ which leads the fluctuation dynamics must not be too short, because it is difficult to observe a slowing down of more than $\approx 10^5\tau_0$. For instance, in magnetic or in displacive transitions, if the microscopic time scale (spin flip, atom jump) is in the picosecond-nanosecond range, XPCS will not be observable, except with the new x-ray laser sources.

The Fe$_{0.76}$Al$_{0.24}$ alloy gives larger fluctuation times than AuAgZn$_2$ and this should ease the observation of the dynamics of the speckles. Unfortunately, due to x-ray larger penetration depth (several $\mu$m) and to lower chemical contrast between ordering elements ($\Delta Z = 13$ for Fe-Al vs $\Delta Z = 32$ for Au-Ag), it may be difficult to carry out dynamic speckle experiments with Fe-Al.

For larger $q$ values ($q > 10^{-3}$ Å$^{-1}$) and shorter fluctuation times ($\tau < 2s$), improved measurements are now attainable:

1. intensities of the order of $10^{10}$ coherent photons (for instance ID10 at ESRF, P10 at PETRA III.) are now obtained with improved sources and optics.
2. beam focusing provides beams a few microns wide, opening the opportunity of selecting a smaller sample region (about $10$ $\mu$m$^2$ area vs $2000$ $\mu$m$^2$ area in this paper) with a smaller number of defects.
3. the new pixel detectors provide a close to unity efficiency and negligible dead time. Their poorer resolution ($\approx 55\mu$m) is well adapted to smaller beams at the sample in a coherent scattering experiment.
4. in the coming years, many synchrotron facilities will upgrade their storage ring to an achromatic lattice. This should result in an increase a factor $\approx 30$ of the source brilliance.

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