Coherent X-ray Studies of Non-Equilibrium Processes

ANDREI FLUERASU,[a,b] MARK SUTTON,[a] ERIC DUFRESNE,[c] AND G.B. STEPHENSON[d]

[a]Department of Physics, McGill University, Montreal, Canada

[b]current affiliation, (ID10A) TROIKA, ESRF, Grenoble

[c]Department of Physics, University of Michigan, Ann Arbor, MI, USA

[d]Material Science Department, Argonne Nat. Laboratories, Argonne, IL, USA

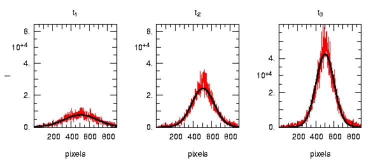
ABSTRACT

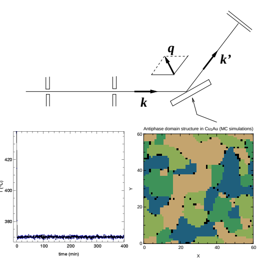
The order-disorder phase transition in Cu_3Au has been studied by x-ray intensity fluctuation spectroscopy (XIFS). The ordering kinetics in Cu_3Au follows a universal behavior as measured by time-resolved incoherent x-ray scattering and predicted by early theories for the coarsening regime in such processes. By using coherent scattering, we measured the fluctuations of the scattered intensity around its average behavior. The covariance of the scattered intensity for a single wavevector, at three different temperatures, was found to be proportional to a scaling function with natural variables $\delta t = |t_1 - t_2|$ and $\bar{t} = \frac{(t_1+t_2)}{2}$ as predicted by theory. Early-time deviations from this scaling form will be discussed.

1 Introduction

1.1 XIFS studies of non-equilibrium systems

→ Coherent X-ray diffraction studies on order-disorder phase transitions in Cu_3Au.



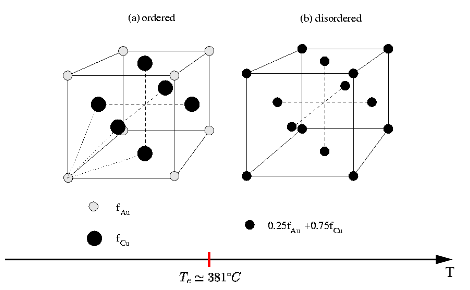


- Random constructive + destructive interference by sample domains results in a "speckle pattern".
- Speckle pattern measure the exact structure factor i.e. exact arrangement of domains.
- XIFS ideal to measure slow dynamics at atomic length scales.

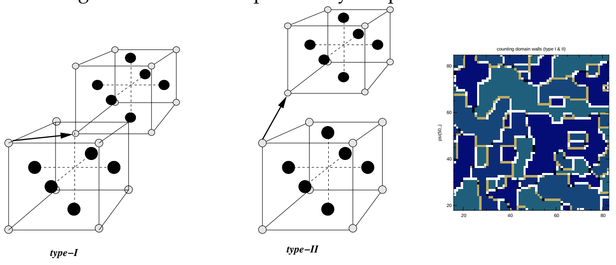
1.2 The order-disorder phase transition in Cu_3Au

Ordered:
→ cubic lattice; 4-atom basis - Au atoms tend to occupy the "corners", and the Cu atoms the face centers.

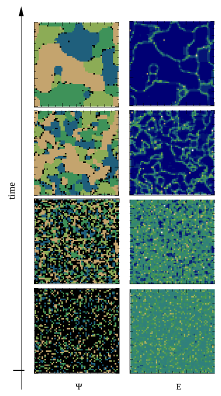
Disordered:
→ fcc lattice, randomly occupied by either a Cu or an Au atom.



The ground state (ordered state) is 4-fold degenerate. Ordered domains in different ground states are separated by antiphase domain walls.



Domain walls:
- type I : formed by "in-plane" translations; low-energy
- type II: formed by "out of plane" translations; high-energy



Phase-ordering following a temp. quench "model A" dynamics [2]:

ψ - phase field (density of ordered material)

$$\frac{\partial\psi(x,t)}{\partial t} = -\Gamma\frac{\delta H}{\delta\psi} + \eta(x,t)$$

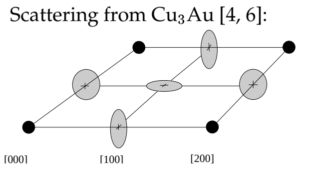
$(\Gamma \to \lambda\nabla^2$ "model B")

$$H(\psi) = \frac{k}{2}|\nabla\psi|^2 - \frac{r}{2}\psi^2 + \frac{s}{4}\psi^4$$

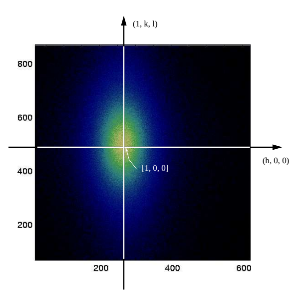
Dimensional analysis:

$$\left[\frac{1}{t}\right] = \left[\frac{1}{L^2}\right] \to L \sim t^{1/2}$$

Scattering from Cu_3Au [4, 6]:

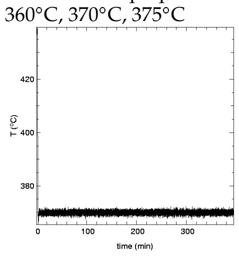


→ superlattice peaks (disks)
→ fundamental peaks (dots)



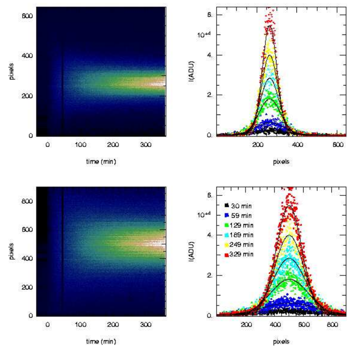
2 Experiments

→ KPPID temp. quenches: 360°C, 370°C, 375°C



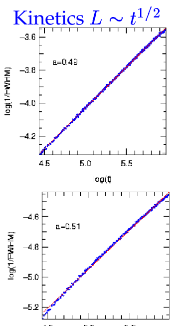
→ Spectrometer APS/IMM-CAT: λ=1.66 Å, $\frac{\Delta\lambda}{\lambda} = 6.2x10^{-5}$ slits: 5μm (H) x 10μm (V)

- X-ray (CXRD) studies of the ordering process

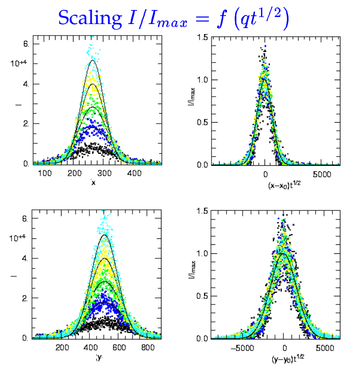


2 One-time analysis (statics)

Kinetics $L \sim t^{1/2}$

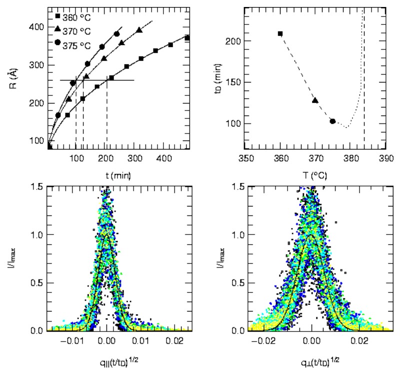


Scaling $I/I_{max} = f\left(qt^{1/2}\right)$



excellent agreement with previous time-resolved XRD studies on the ordering process in Cu_3Au [5].

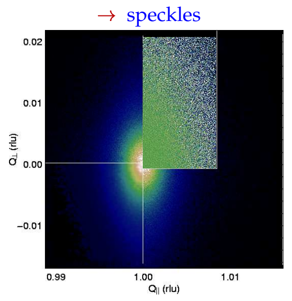
→ Temperature dependence is determined by the competition between and increased thermodynamic force associated with a deeper quench and a reduced atomic mobility



3 Two-time analysis (dynamics)

3.1 Coherent X-rays: speckles

→ speckles

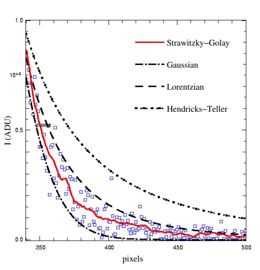


$\langle I(q,t)\rangle$ $\qquad I(\mathbf{q},t)$

- Random constructive + destructive interference by sample domains results in a "speckle pattern".
- Speckle pattern measure the exact structure factor i.e. exact arrangement of domains.
- Intensity fluctuations:

$$D(q,t) = \frac{I(q,t) - \langle I(q,t)\rangle}{\langle I(q,t)\rangle}$$

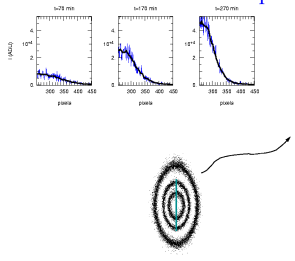
3.2 Evaluating $\langle S(\mathbf{q},t)\rangle$

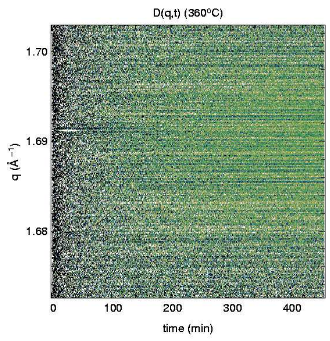


- Equilibrium systems: ✗
→ averaging over time
- Non-equilibrium systems: ✗
→ averaging over rings of constant **q** in isotropic systems
→ fits with various analytical forms: Gaussian, Lorentzian, Hendricks-Teller ✗
- Any systems:
→ Strawitzky-Golay filters ✓

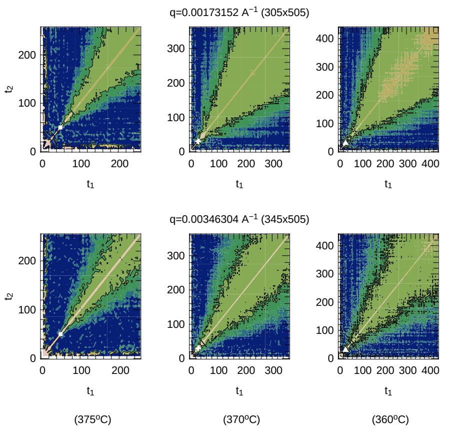
3.2 Results

→ Time-evolution of speckles



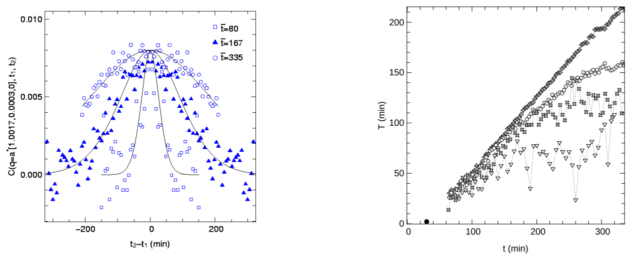


$C(\mathbf{q},t_1,t_2) = \langle D(\mathbf{q},t_1)D(\mathbf{q},t_2)\rangle$

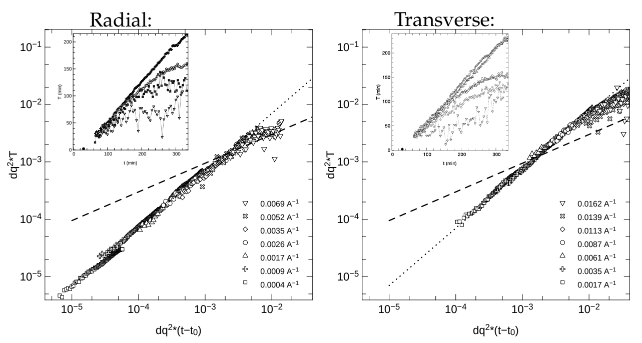


$$C(\mathbf{q},t_1,t_2) = C\left(\mathbf{q}, \bar{t} = \frac{t_1+t_2}{2}, \Delta t = t_2 - t_1\right)$$

$$C_{norm}(z) = \left(\frac{z^2K_2(z)}{2}\right)^2, z = A\frac{\Delta t}{\sqrt{\bar{t}}} = \frac{\Delta t}{T}$$, T - correlation time([3])



Scaling behavior of the two-time correlation functions:



4 Conclusions

- XIFS is ideal way to measure the dynamics of fluctuations in non-equilibrium systems at atomic length scales, if the intensity is sufficient for the characteristic time scales involved.
- First experimental confirmation of the two-time scaling laws in a "model A" system.

References

[1] S. Brauer, G. B. Stephenson, M. Sutton, R. Bruning, E. Dufresne, S. G. J. Mochrie, G. Grubel, J. Als-Nielsen, and D. L. Abernathy, Phys. Rev. Lett. **74**, 2010 (1985); L.B. Lurio, D. Lumma, P. Falus, M.A. Borthwick, S.G.J. Mochrie, J.F. Pelletier, M. Sutton, L. Regan, A. Malik, and G.B. Stephenson, Phys. Rev. Lett. **84** 785 (2000); D. Lumma, Z. Dufresne, T. Nurushev, R. Clarke, and S.B. Dierker, Phys. Rev. E, **65**, 065107 (2002).

[2] For an extensive review, see J.D. Gunton, M. San Miguel and P.S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J.L. Lebowitz (Academic, London,1983), Vol. 8.

[3] G. Brown, P.A. Rickvold, M.Sutton, and M. Grant, Phys. Rev. E **56**, 6601 (1997).

[4] B. Warren, *X-ray Diffraction* (Addison-Wesley, Reading, MA, 1969), Chap. 12, pp. 206-250.

[5] Robert F. Shannon, Jr., Stephen E. Nagler, Curt R. Harkless, and Robert M. Nicklow, Phys. Rev. B **46**, 40 (1992).

[6] K. F. Ludwig, Jr., G. B. Stephenson, J. L. Jordan-Sweet, J. Mainville, Y. S. Yang, and M. Sutton, Phys. Rev. Lett. **61**, 1859 (1988).