Studies of atomic diffusion in crystalline alloys by XPCS

M. Stana[*1], **M.Leitner**[1,2] **M.Ross**[1] **and B. Sepiol**[1]

[1]**Universität Wien, Fakultät für Physik, Strudlhofgasse 4, 1090 Wien, Austria,** [2]**TU München, Physik-Department E13, 85747 Garching, Germany**
[*]**markus.stana@univie.ac.at**

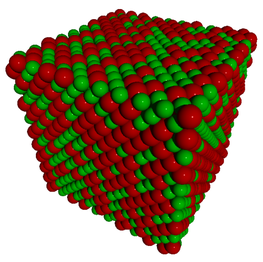
1 Introduction

Measuring diffusion on an atomic level directly in real space is not possible today. Using coherent X-rays, however, we can (ignoring the details of the exact structure factor) use the time fluctuations of the scattering pattern to study dynamics of the material.

With **X**-ray **P**hoton **C**orrelation **S**pectroscopy (XPCS) we collect a series of speckle pattern images representing a detail of reciprocal space at a certain time t and a certain reciprocal space vector ($\vec{q}$). Correlating these images we get correlation times $\tau(\vec{q})$ for different $\vec{q}$. The method was successfully applied on a Cu-Au alloy [LSS$^+$09]. This poster will show its feasibility on $Ni_{97}Pt_3$ solid solution.

2 Theory and experiment schematics

Real Space



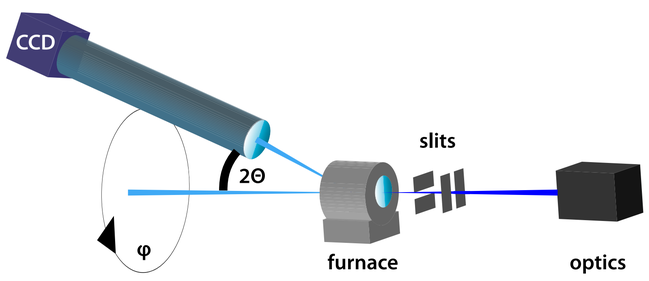
Van Hove Autocorrelation function:

$$G(\Delta\vec{a},\Delta t)=\langle\sigma(\vec{a},t)\sigma(\vec{a}+\Delta\vec{a},t+\Delta t)\rangle$$

with:

$$\sigma(\vec{a},t)=\begin{cases}0 & \text{if } atomtype\ A \text{ at } \vec{a}\\ 1 & \text{if } atomtype\ B \text{ at } \vec{a}\end{cases}$$

Experimental setup:



Fourier Transform

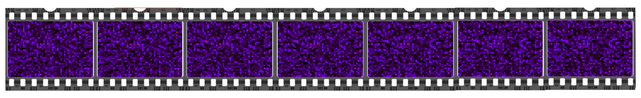
Scattering experiment

Connected by:

$$g^{(1)}(\vec{q},\Delta t)=\mathscr{F}_{\Delta\vec{a}\to\vec{q}}(G(\Delta\vec{a},\Delta t))=e^{-\left(\frac{\Delta t}{\tau(\vec{q})}\right)}$$

with $\tau(\vec{q})$ dependent on $I_{SRO}(\vec{q})$ (short-range order intensity)

A series of images detected at certain $\vec{q}$:



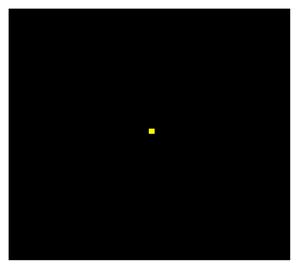
Intensity Autocorrelation function and Amplitude Autocorrelation function linked by **Siegert relation**:

$$g^{(2)}(\vec{q},\Delta t)=1+b(g^{(1)}(\vec{q},\Delta t))^2=1+be^{-2\left(\frac{\Delta t}{\tau(\vec{q})}\right)}$$

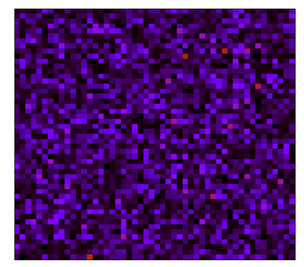
b ..... contrast (due to non-perfect coherence and other effects), Δt ..... time interval (number of frames between successive image recordings)

Reciprocal space

Bragg scattering:



Diffuse scattering:

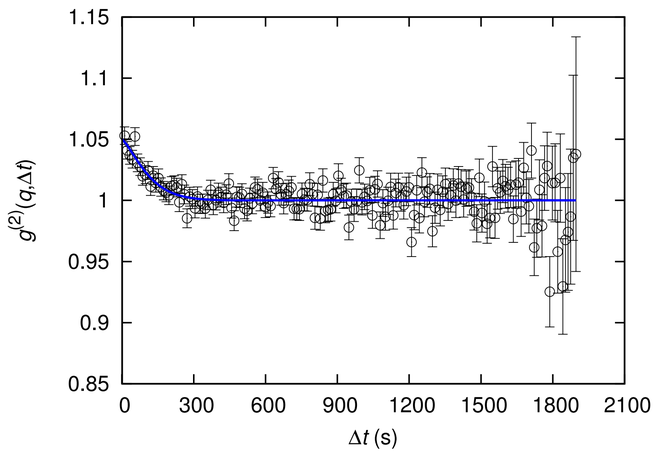


Amplitude Autocorrelation function:

$$g^{(1)}(\vec{q},\Delta t)=\frac{\langle A(\vec{q},t)A(\vec{q},t+\Delta t)^*\rangle}{\langle|A(\vec{q},t)|\rangle^2}$$

Intensity Autocorrelation function:

$$g^{(2)}(\vec{q},\Delta t)=\frac{\langle I(\vec{q},t)I(\vec{q},t+\Delta t)\rangle}{\langle I(\vec{q},t)\rangle^2}$$

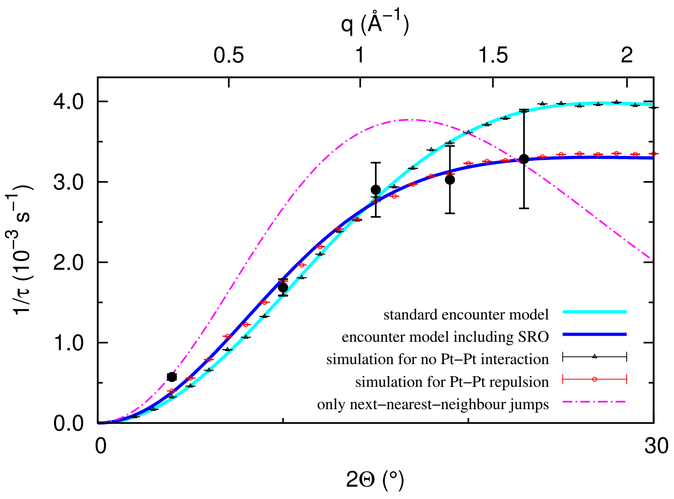


3 Results

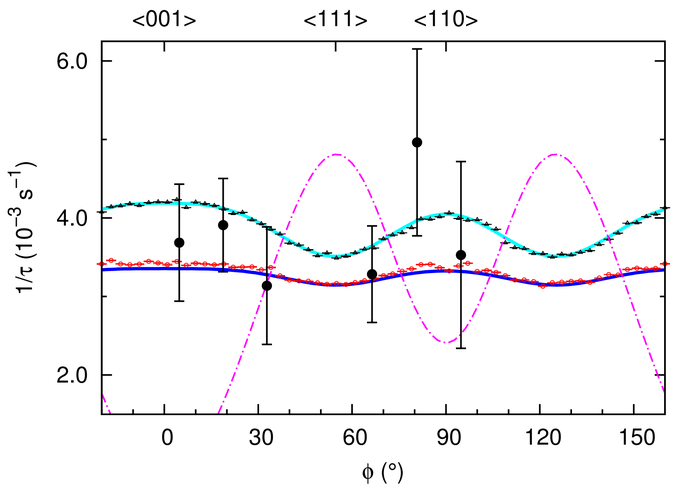
We measured a **$Ni_{97}Pt_3$ single crystal** at beamline ID10A, ESRF, Grenoble. In a solid solution diffusion is supposed to be governed by the encounter model [Wol77].
Two approaches were used for the Metropolis Monte Carlo algorithm for diffusion. One without short-range order (no interaction force between atoms) and one with strong Pt-Pt repulsive force (higher concentration of Pt atoms yields an $L1_2$ configuration [CDK86]).

- average atomic jump freq.: $\nu=1.87(11)\times10^{-3}\ s^{-1}$
- diffusion rate $\tilde{D}=2.29(10)\times10^{-23}\ m^2\ s^{-1}$
- **this means that very low diffusivity could be measured**
- **it is necessary to account for repulsive force between Pt atoms**

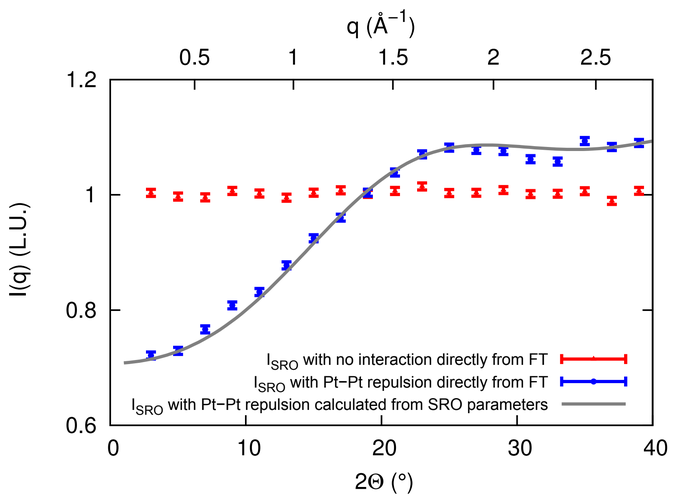
Data measured along scattering angle 2Θ:



and along azimuthal angle (ϕ) compared to theory:



Intensities from Monte Carlo sim. for ϕ = 33.2°:



References

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