Studies of atomic scale diffusion by x-ray photon correlation spectroscopy

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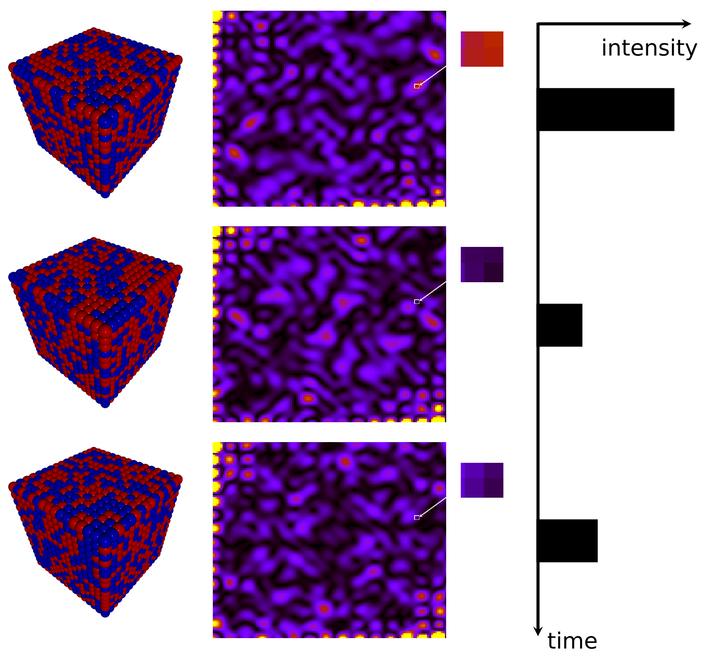
1 Introduction

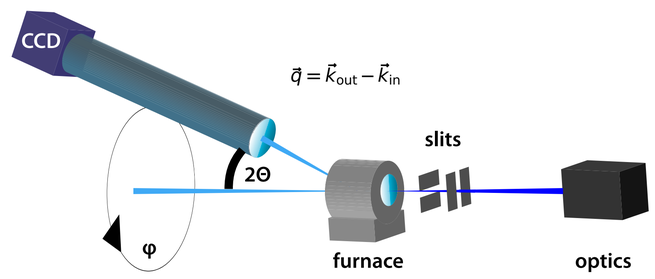
This poster gives an overview of the relatively new technique of **a**tomic scale **X**-ray **P**hoton **C**orrelation **S**pectroscopy (aXPCS), which uses the time evolution of speckle patterns in the diffuse intensity regime to reveal diffusion mechanisms in real space. This is done via the so called **intensity autocorrelation function**.
The advantages of this new technique are its **sensitivity to jump mechanisms**, its applicability at very low temperatures, i.e. for **very low diffusion rates** and for a broad variety of systems (**not element specific**).

2 Experiment and theoretical background

Different real space configurations yield different intensities in the diffuse regime (here in (100) plane):

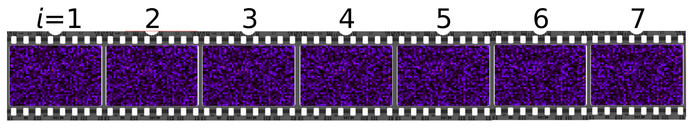


In the experiment a CCD camera collects a series of images from a detail of reciprocal space corresponding to a certain $\vec{q}$ and for a certain T:



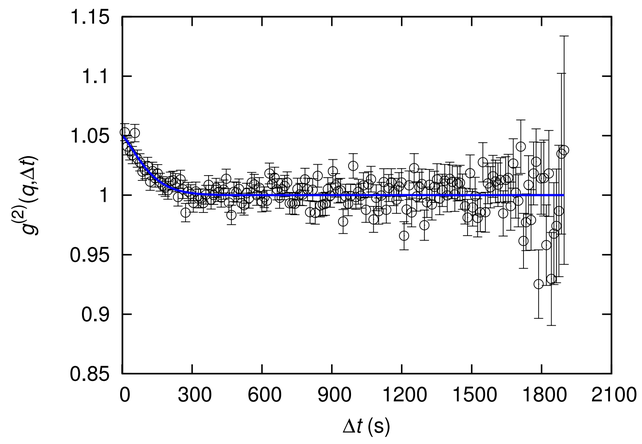
Variable parameters in the experiment:

- scattering vector $\vec{q}$ $\begin{cases} \text{scattering angle } 2\Theta \\ \text{azimuthal angle } \varphi \\ |\vec{k}_{in}| = |\vec{k}_{out}| \propto E_{beam} \end{cases}$
- temperature T
- sample orientation relative to $\hat{k}_{in}$
- exposure time $\rightarrow$ frame rate



CCD images for different t at particular $\vec{q}$ are used to calculate the Intensity Autocorrelation function:

$$g^{(2)}(\vec{q},\Delta t) = \frac{\langle I(\vec{q},t) I(\vec{q},t+\Delta t)\rangle}{\langle I(\vec{q},t)\rangle^2} = 1 + \beta \exp\left(-2\frac{\Delta t}{\tau(\vec{q})}\right) \quad (1)$$



3 Finding a diffusion model

General form of correlation time $\tau(\vec{q})$:

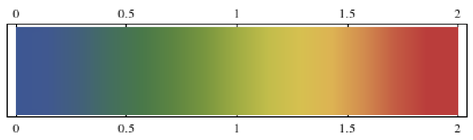
$$\tau(\vec{q}) = \tau_0 \frac{I_{SRO}(\vec{q})}{\sum_n P_n \sum_{\Delta\vec{a}_{nj}} \left(1 - \exp\left(i\vec{q}\cdot\Delta\vec{a}_{nj}\right)\right)} \quad (2)$$

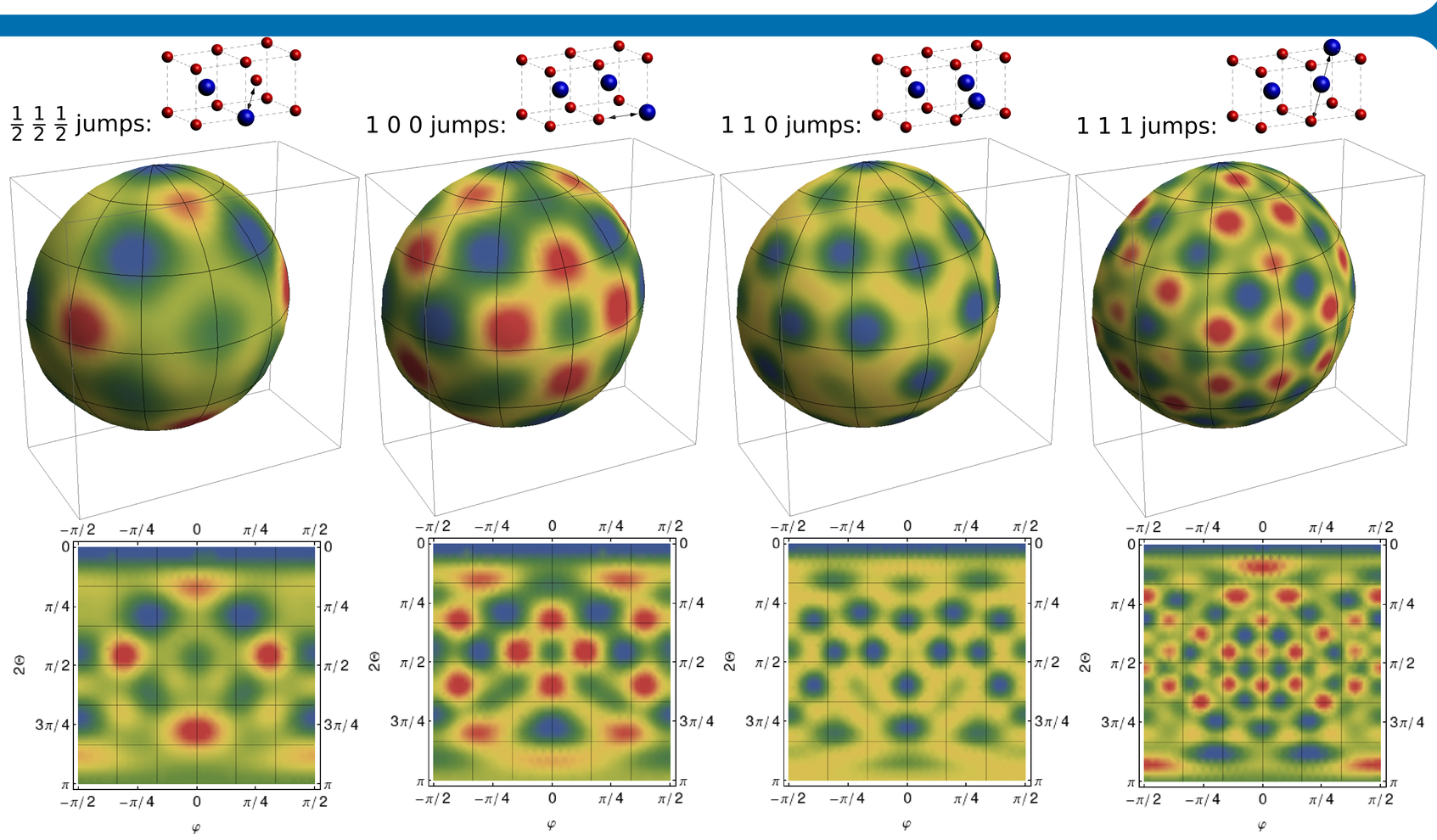
This can be calculated for a particular system and certain parameters:

- short-range order intensity I_{SRO}
- neighbors for certain lattice type $\Delta\vec{a}_n$
- lattice constant d
- crystal orientation

Notice: the sample to detector distance is only a scaling factor.

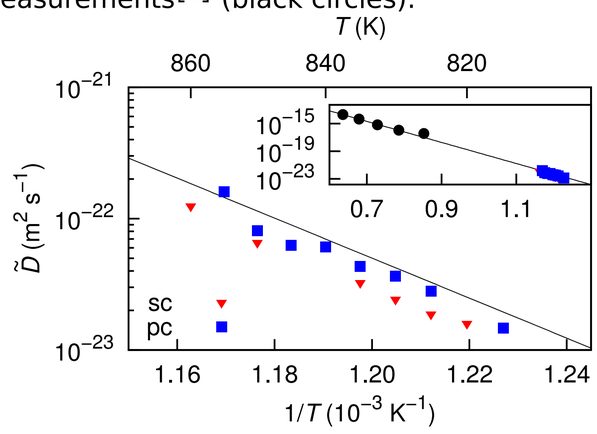
An example of inverse correlation times $1/\tau(\vec{q})$ in a bcc system oriented with $\hat{k}_{in} \parallel \langle 110\rangle$ and with $I_{SRO} = 1$ for different jump vectors is given to the right:



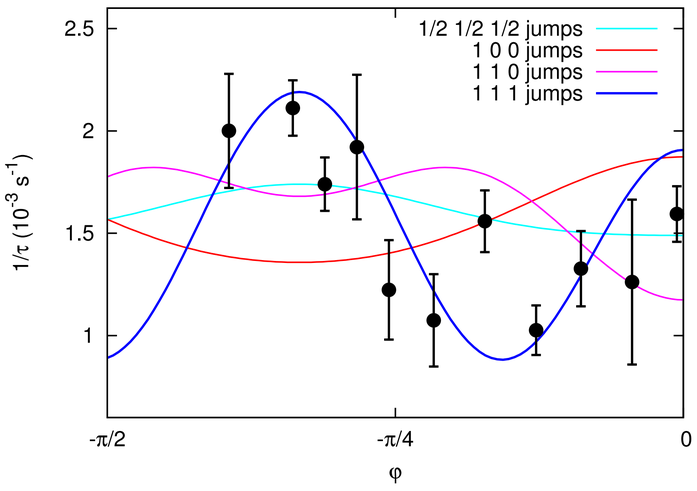


4 Some results

$Ni_{97}Pt_3$ solid solution[1] single crystalline (sc) and polycrystalline (pc) sample measured at different temperatures compared with tracer measurements[2] (black circles):



$Fe_{55}Al_{45}$ binary intermetallic alloy[3] compared with diffusion models with $I_{SRO}(\vec{q}) = 1$:



In the first example of $Ni_{97}Pt_3$ one can see that aXPCS measurements agree well with tracer measurements. The advantage of aXPCS measurements here is the possibility to measure at very low diffusion rates corresponding to very low temperatures. The other big advantage, as can be seen in the example of $Fe_{55}Al_{45}$, is the possibility to identify diffusion mechanisms. In this case, even tough the inclusion of $I_{SRO}(\vec{q}) \neq 1$ could still change this picture, the 111 jump mechanism seems dominant.

[1] M. Stana, M. Leitner, M. Ross, and B. Sepiol, J. Phys.: Condens. Matter 25 (2013) 065401.

[2] M. S. A. Karunaratne and R. C. Reed. Acta mater., 51(10) (2003) 2905-2919.

[3] Data measured at PETRA III at $E = 7$ keV, $2\Theta = 20°$ and $T = 653$ K (preliminary results).

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