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

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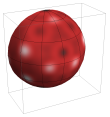
1 Theoretical background

Solving the *rate equation* for discrete atomic jumps on a Bravais lattice yields a correlation time $\tau(\vec{q})$:

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

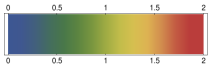
n … number of neighboring shell
p_n … jump probability to certain position in nth shell
$\Delta\vec{r}_{nj}$ … relative jump vector to jth position in nth shell
$\vec{q}$ … scattering vector
I_{SRO} … short-range order intensity in Laue units



For a coherent method like XPCS short-range order plays a role, hence the I_{SRO} in Eq. (1).

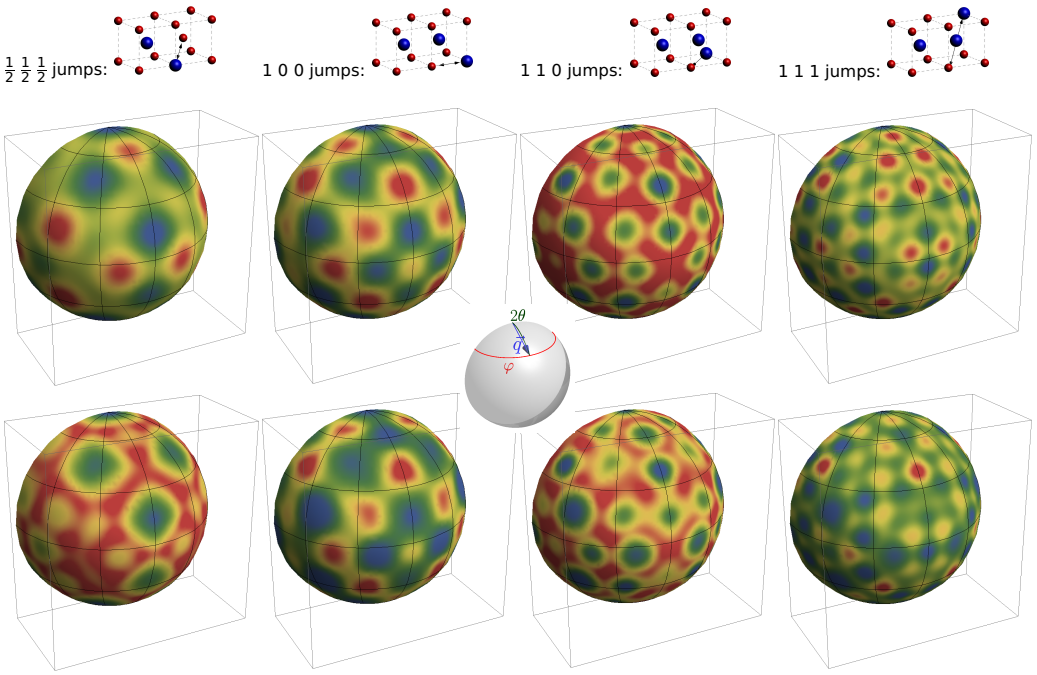
$\Delta\vec{r}_{nj}$ depends on lattice constant d and the symmetry of lattice. $\vec{q}$ depends on the parameters described below. For a certain choice of such parameters an inverse correlation time $1/\tau(\vec{q})$ can be calculated (here bcc).

$1/\tau(\vec{q})$ (∝ Hz)



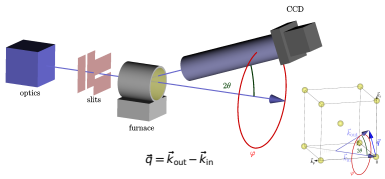
Images on the right show different jump mechanisms without I_{SRO}-correction ($1/\tau_{\mathrm{inc}}$) (*top*) and corrected with I_{SRO} shown above (*bottom*).

$\frac{1}{2}\frac{1}{2}\frac{1}{2}$ jumps: 1 0 0 jumps: 1 1 0 jumps: 1 1 1 jumps:



2 Experiment

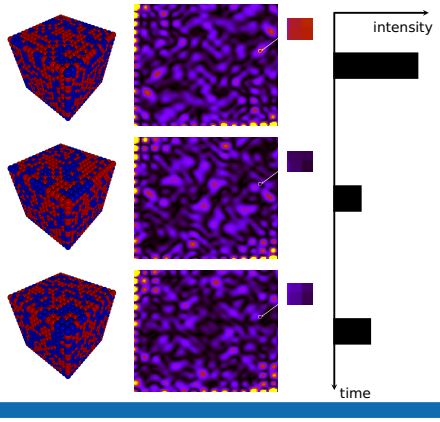
A CCD camera is used to collect a series of images from a detail of reciprocal space corresponding to a certain $\vec{q}$ and for a certain T:

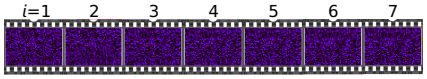


Variable parameters in the experiment:

- scattering vector $\vec{q}$ { scattering angle 2Θ; azimuthal angle φ; $|\vec{k}_{\mathrm{in}}| = |\vec{k}_{\mathrm{out}}| \propto E_{\mathrm{photon}}$
- temperature T
- sample orientation relative to $\hat{k}_{\mathrm{in}}$
- exposure time → frame rate

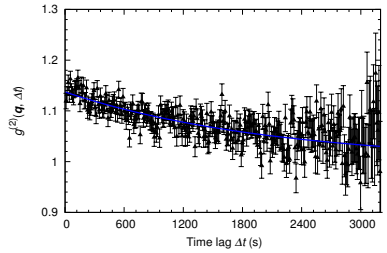
Different atomic configurations in real space yield different intensities in the diffuse regime (here in (100) plane):





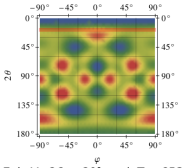
CCD images for different t at particular $\vec{q}$ are used to calculate the Intensity Autocorrelation function:

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

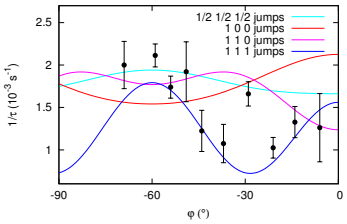


3 Results for $Fe_{55}Al_{45}$

Projection of model for $\frac{1}{2}\frac{1}{2}\frac{1}{2}$ - jumps. Models from section 1 ($I_{\mathrm{SRO}}(\vec{q}) = 1$) compared with φ-scan at 2θ=20° for $Fe_{55}Al_{45}$ binary intermetallic alloy[1]:

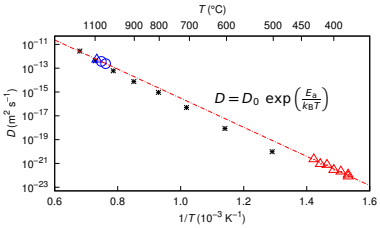


[1] Data measured at PETRA III at $E = 7$ keV, $2\Theta = 20°$ and $T = 653$ K (preliminary results).



Without I_{SRO} - correction 111 jumps appear to be the dominating process.

Arrhenius plot of aXPCS data in comparison to literature values acquired by different techniques:



✳ $Fe_{52}Al_{48}$ measured with tracer diffusion technique (^{59}Fe) [2]
◯ $Fe_{50.5}Al_{49.5}$ measured with QMS [3]
△ $Fe_{55}Al_{45}$ measured with QMS [4]
△ $Fe_{55}Al_{45}$ measured with aXPCS [5]

[2] M. Eggersmann and H. Mehrer, Phil. Mag. A 80:5, 1219 (2000)
[3] G. Vogl and B. Sepiol, Acta metall. mater. 42, 3175 (1994)
[4] R. Feldwisch, B. Sepiol and G. Vogl, Acta metall. mater. 43, 2033 (1995)
[5] preliminary data

As atomic-scale X-ray Photon Correlation Spectroscopy (aXPCS) is a coherent method, it requires information about short-range order in the system. Reliable I_{SRO} measurements are therefore essential for data evaluation.
aXPCS is a valuable tool to determine atomic diffusion mechanisms. It is applicable over a wide range of temperatures being only limited by CCD-readout times and intensity towards fast processes and by setup and system stability towards slow processes. This allows for investigations at relatively low temperatures that were unaccessible to any other atomistic method so far. Furthermore there is no limitation to special isotopes.

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