Atomic-scale diffusion in ordered intermetallics and glasses studied by X-ray photon correlation spectroscopy

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INTRODUCTION

Macroscopic tracer methods achieve extremely high sensitivity and are widely applicable in diffusion studies. Sometimes, however, if we want to find out the diffusion mechanism it is an advantage to use methods measuring atomic movements *directly* in space and time. Only few scattering methods are sensitive enough to resolve jump vectors on the atomic scale but they are limited to a restricted number of suitable isotopes. Moreover, they are always restricted in the time/energy resolution and thus limited to very fast diffusion processes.

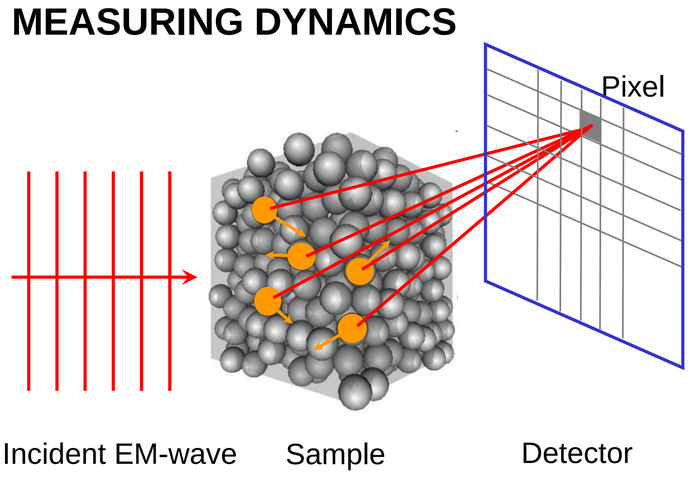
Atomic-scale X-ray photon correlation spectroscopy (aXPCS) seems to be very promising in the study of dynamics with atomic resolution.

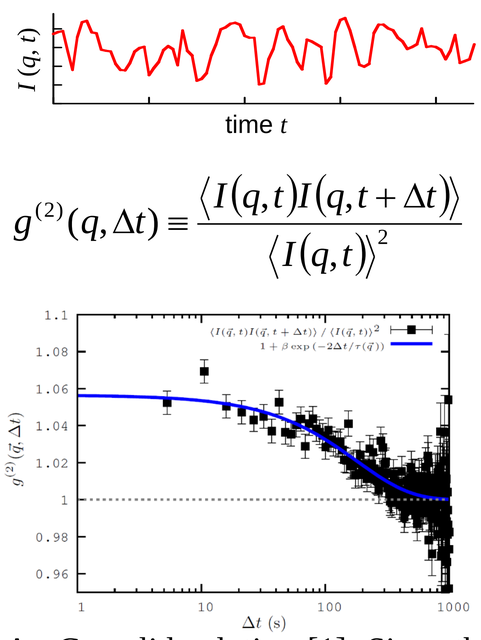
METHOD

The aXPCS method relies on the fact that a particular arrangement of atoms in a sample produces a characteristic *speckle* pattern when it scatters from a coherent beam of X-rays. A change in the arrangements of atoms leads to a modification in the the speckle pattern. Studying these fluctuations as a function of time, one can obtain information on the atomic dynamics at various wave-vector transfers (i.e. at different length scales).

This technique operates in the time regime and measures fluctuations in atomic positions. The time resolution towards faster dynamics is limited only by the readout time of the detector and the intensity of the X-ray beam. In the direction of slower dynamics it is restricted by the finite measurement time.

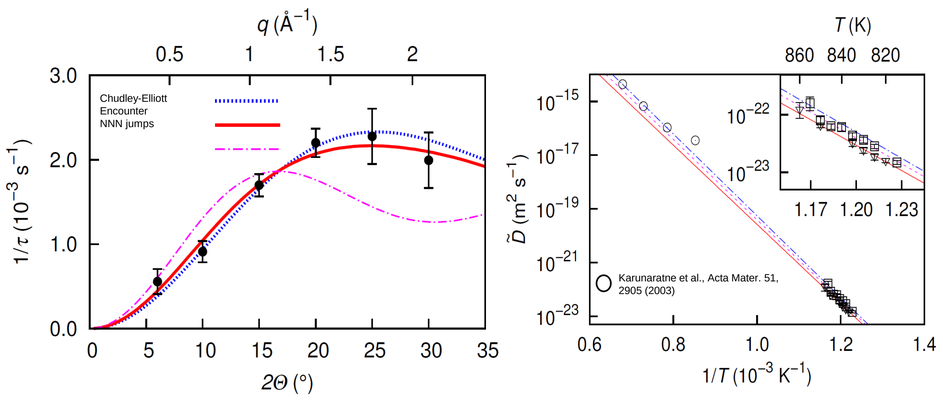
MEASURING DYNAMICS





We carried out our first successful aXPCS experiment on Au-Cu solid solution [1]. Since then we measured solid solutions like Ni-Pt [2], well-ordered B2 intermetallics like Fe-Al [3], Ag-Mg or Ni-Al, metallic glasses as well as we directly observed single atomic motion in lead silicates [4] and in fast ionic conductors alkali borate glasses.

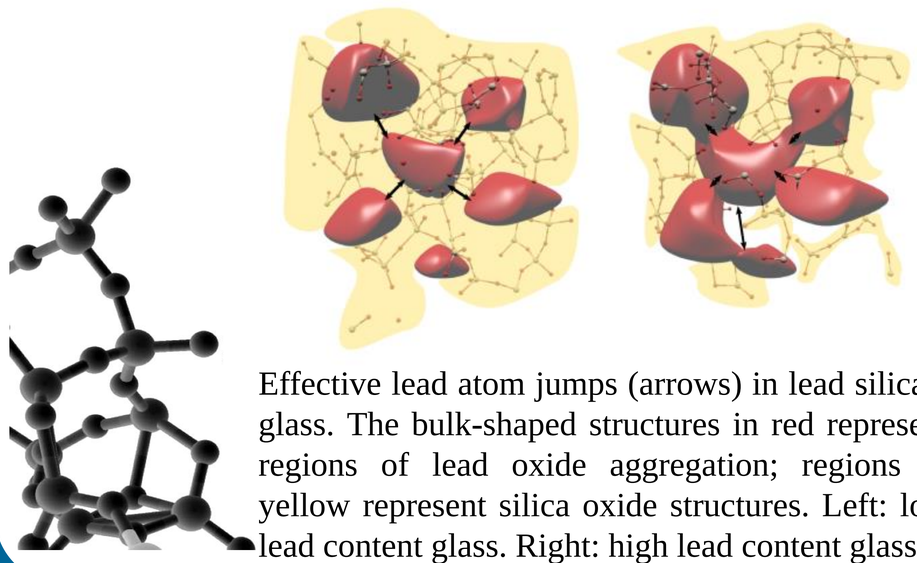
POLYCRYSTALLINE Ni-3at.%Pt sample



Ni-Pt is a model binary system. We proved that *polycrystalline* samples can be also used to study atomic-scale diffusion, although single crystalline samples like Au-Cu [1] yield more information about the microscopic dynamics. Experimental data are well described by the encounter model and an Arrhenius plot of diffusion coefficients yields satisfactory results comparable to the well-established tracer technique outcomes.

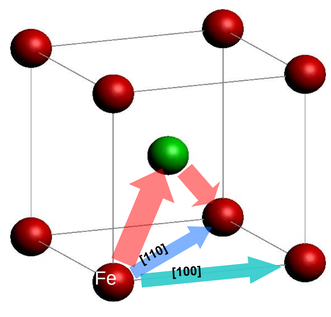
OXIDE GLASSES

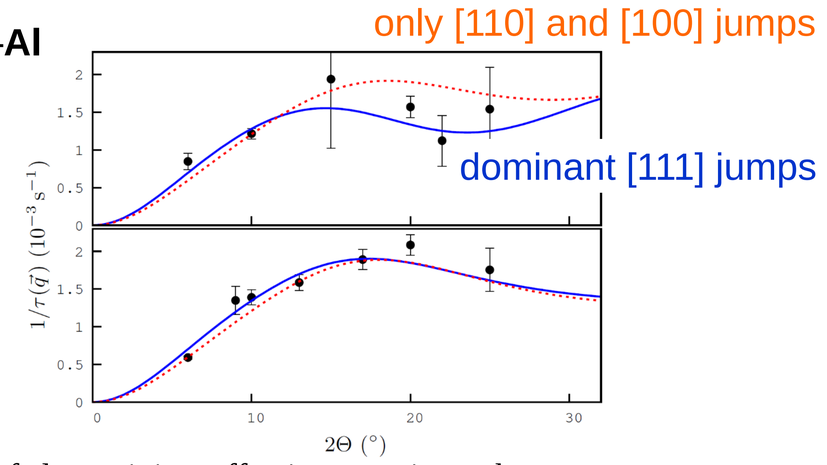
We showed that the dynamics on the atomic level in lead silicate glass changes significantly below and above the hypothetical transition from network-forming silica oxide to the network-forming lead oxide. For low lead oxide concentration only diffusion via long-range jumps was observed, while for high lead oxide concentration the percolation of the network facilitated a large amount of additional short-range jumps. This shows that aXPCS is a viable tool to study diffusion in glasses below the glass transition.



Effective lead atom jumps (arrows) in lead silicate glass. The bulk-shaped structures in red represent regions of lead oxide aggregation; regions in yellow represent silica oxide structures. Left: low lead content glass. Right: high lead content glass.

EFFECTIVE IRON JUMPS in Fe–Al





We could show that aXPCS is capable of determining effective atomic exchanges at temperatures unaccessible to other methods of diffusion studies with atomistic resolution. In the non-stoichiometric long-range ordered $Fe_{54}Al_{46}$ system at 0.42 T_m, the dominating diffusion mechanism leads to effective ⟨111⟩-exchanges of Fe and Al atoms.

SUMMARY

- aXPCS is a versatile tool for studying dynamics of single atoms in solids
- Characteristic jump distances and residence times are accessible in crystalline solids and in glasses
- Very slow diffusion at low temperatures is measurable
- New insights into collective dynamics in glasses are possible

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