



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

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Introduction

Understanding diffusive dynamics at the atomic level is of fundamental importance for comprehending material properties in solid state physics. Especially for high performance materials like iron-aluminum alloys this is true.

In our group **Q**uasielastic Mößbauer **S**pectroscopy (QMS) was used for many years to investigate such processes. More recently we have been using the relatively new technique of **a**tomic scale **X**-ray **P**hoton **C**orrelation **S**pectroscopy (aXPCS) to do this. This new technique operates in the time rather than in the energy regime. This poster gives an overview on both approaches, selected results are presented and the advantages and drawbacks of both methods are discussed.

Quasielastic Mößbauer spectroscopy

Different diffusion mechanisms lead to different broadenings in the Mößbauer spectrum called "quasielastic diffusional broadening". These broadenings are strongly angle-dependent. The experimental setup is shown below:



Figures from R. Röhlsberger, Nuclear Condensed Matter Physics with Synchrotron Radiation, Springer 2004.

In a B2 ordered binary intermetallic alloy the most probable jump mechanisms are:



Atomic scale x-ray photon correlation spectroscopy

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



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} $\begin{cases} \text{scattering angle } 2\Theta \\ \text{azimuthal angle } \varphi \\ |\vec{k}_{\text{in}}| = |\vec{k}_{\text{out}}| \propto E_{\text{beam}} \end{cases}$

• temperature T • sample orientation relative to \hat{k}_{in} • exposure time \rightarrow frame rate





In the case where atoms of type A only reside very shortly on the sublattice B (on antistructure sites), the interplay of the different diffusion mechanisms leads to a total broadening of the Mößbauer line $\Gamma(\vec{q})$. Therefore this broadening is significant for the jump mechanisms taking place. CCD images for different t at particular \vec{q} are used to calculate the Intensity Autocorrelation function:

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

 $\tau(\vec{q})$... angle-dependent correlation time



Fe₅₅**Al**₄₅ **sample with QMS**

The sample was measured at T = 1363 K, which corresponds to 0.88 $T_{\rm C}$. The spectrum was taken in the (110) plane with 14.4 keV Mößbauer γ radiation.





* Fe₅₂Al₄₈ measured with tracer diffusion technique (⁵⁹Fe) [²] • Fe_{50.5}Al_{49.5} measured with QMS [³] • Fe₅₅Al₄₅ measured with QMS [⁴]

Fe₅₅**Al**₄₅ sample with aXPCS

The sample was oriented with the (110) direction parallel to the incoming beam. An x-ray energy of 7 keV was chosen to stay below the iron fluorescence edge. The temperature in the furnace was T = 653 K which corresponds to 0.42 $T_{\rm C}$. Measurements for different φ were taken at 2 $\Theta = 20^{\circ}[5]$:



Published in R. Feldwisch, B. Sepiol and G. Vogl, Acta metall. mater 43, 2033 (1995).

The fitted model includes 110 jumps and 100 - jumps with a ratio of $\frac{W_{110}}{W_{100}} = 1.7.$ \sim Fe₅₅Al₄₅ 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

Conclusion: even though the activation energy (E_a) is the same, the jump mechanisms change with temperature.

φ

At this temperature the 111 jump mechanism seems to be the dominant one (only 111- jumps).

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Advantages of each method

aXPCS works in the time regime and its temporal resolution is only dependent on the stability of the experimental setup and the time spent. It can therefore measure very low diffusivities corresponding to low temperatures. In comparison, Mößbauer spectroscopy only works at elevated temperatures close

to the melting point, because within the lifetime of a Mößbauer nuclear level at least one atomic jump is necessary to produce detectable energy broadening. Another advantage of aXPCS is that it is theoretically not restricted to certain elements. With today's synchrotron sources, however, some limitations to this still apply (high scattering contrast in elements is required). On the other hand Mößbauer spectroscopy is a non-coherent method, which does not require correction for coherent phenomena like short-range order intensity and therefore allows a more straightforward interpretation of experimental results.