



Experiment title: Study of the Diffusion Jump by Nuclear Forward Scattering of Synchrotron Radiation

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HS-117

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6

Local contact(s):

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Report:

In this project nuclear forward scattering of synchrotron radiation was used in order to study the diffusional dynamics of atomic motion in solids. As was shown in first measurements on a Fe_3Si single crystalline foil [1] and on the molecular glass former dibutylphthalate [2], diffusive motion of ^{57}Fe atoms gives rise to a loss of coherence of the forward scattered field resulting in an accelerated decay in delayed intensity. Here we report on measurements on FeSi as a function of temperature **and** crystal orientation. Mounting the sample environment on a goniometer head allowed for a precise alignment of the [111] and [113] crystal direction parallel to the synchrotron beam. Data were collected at various temperatures between 827K and 967K.

The nuclear forward scattered intensity depends not only on temperature but also on crystal orientation (Fig. 1). Besides the determination of the ^{57}Fe diffusivity the microscopic diffusion mechanism may be derived. Evaluating the orientational dependence of the forward scattered signal we are now able to answer the question which has been asked in this experiment:

There is complete parallelity between diffusional line broadening in Mössbauer spectroscopy and accelerated decay in nuclear forward scattering of SR.

A correct fit of spectra measured at the same temperature but along different crystal directions requires the knowledge of a proper diffusion model as an input in the fitting procedure. Using "classical" quasielastic Mossbauer spectroscopy (QMS) on the same system we established a simple model with nearest neighbour jumps of iron atoms between α and r-iron sublattices [3] The iron atom jumps rapidly over its own sublattices without disturbing the degree of order.

A superficial consideration of the time-dependent spectra would raise a false impression that information which can be obtained from fits would be very limited. The results of the fitting procedure showed actually that nuclear forward scattering is extremely sensitive to details of the diffusion model. Consistent fits of all spectra were impossible using the simple model obtained with QMS.

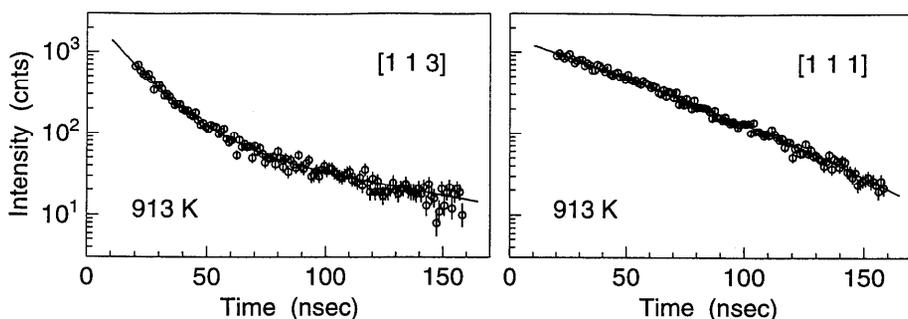


Fig. 1. Nuclear forward scattering of SR measured along the [113] and [111] directions at 913 K.

A satisfactory fit (Fig. 1) was obtained after a small admixture of iron atom jumps via antistructure sites to the microscopic diffusion model. We explain this difference to QMS with the properties provided by synchrotron radiation:

(a) nuclear forward scattering offers an ideal resolution function which depends only on the life time of the excited state. There is no additional line broadening and change in spectral shape of the observed resonance level characteristic for a strong radioactive Mößbauer source.

(b) an excellent divergence of the synchrotron beam (ID 18: $\sim \mu\text{rad}$) allows for a precise determination of the diffusional line broadening resulting in a considerably reduced smearing of effects depending on crystal orientation as compared to QMS.

A disadvantage of nuclear forward scattering seems to be a limitation towards fast diffusion. The enhanced acceleration of the forward scattered intensity requires measurements at short times. However, the detector overload due to the intense prompt pulse limits the accessible time window to some 20 nsec. Further improvement of the avalanche photo diode detector should overcome this problem.

Apart from measurements of full spectra, we also developed a new measuring mode - collecting simply all delayed counts versus crystal orientation. A scan of the orientation dependent integral nuclear forward scattered intensity (ODIN) enables a very fast determination of geometrical relations connected to the investigated structure and its orientation, e.g. to find crystal directions where the projection of the jump vector on the incident wave vector results in a maximal and minimal line broadening. Afterwards, to investigate the microscopic diffusion mechanism in detail one can measure a full time dependent spectrum in this selected directions.

First results were already presented at the DIMAT conference [4]. We are currently preparing a manuscript to report this experiment in detail.

References

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