

Atomic Diffusion in B2-NiAl

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X-ray photon correlation spectroscopy (XPCS) was in recent years established as a new, time-resolved scattering method for investigating diffusion on the atomic scale. Temperature-driven movement of individual atoms is the fundamental dynamic process in solids. It is, however, very difficult to follow this motion with common methods. The most general approach to this problem is an application of the tracer technique. However, this method does not preserve information about effective atomic jump directions. The present XPCS technique allows a completely different approach in the investigation of dynamics in solids, using highly brilliant synchrotron sources to measure time-resolved scattering patterns. Thereby XPCS enables studies of very low diffusivities. It is neither limited by the structure nor by the elemental constitution and therefore makes a variety of systems accessible for investigation.

After the successful application of the technique in a Cu-Au single crystal [1], this project addresses the long-standing problem of the atomistic diffusion mechanism in the cubic B2-ordered Ni-rich Ni-Al phase. Contrary to self-diffusion in pure elements, where the mechanism is practically always a vacancy-mediated nearest-neighbor jump, in intermetallic compounds more complex mechanisms have to be at work. In the B2-ordered structure all nearest-neighbor lattice sites of one species (e.g. atoms of type A) are exclusively surrounded by the atoms of the opposite element (B-atoms). The crucial factor for determining the hopping mechanism in B2 alloys is finding a defect structure model. In the Ni-Al phase a defect structure model first proposed by Bradley and Taylor [2] can be applied: on the B-rich side of the phase diagram only an A-vacancy is formed (anti-structure majority atoms on the minority sublattice) while on the A-rich side an A anti-structure defect (ASD) exists (vacancies on the majority sublattice). This type of defects are called *structural* or *constitutional*, i.e. those defects which survive at zero temperature in order to maintain the deviation from stoichiometry. In the Ni₅₈Al₄₂ alloy investigated the dominating ASD is thus a majority (Ni) atom on the Al sublattice.

A Ni₅₈Al₄₂ single crystal, oriented with the $\langle 110 \rangle$ -axis normal to the sample surface was mounted in a specially designed vacuum furnace and measured in transmission geometry. Images were taken with a Pixis-XB CCD camera with 1340×1300 , $20 \times 20 \mu\text{m}^2$ pixel size. The exposure time was 3 s per frame with 2.4 second read-out time. The distance sample-detector was 0.83 m. With this set-up, the count rate was about 0.006-0.008 photons per pixel and frame and the coherence factor about 5% at the scattering angle $2\vartheta = 20^\circ$. Positions of photons in the frame were processed via droplet algorithm. To ensure that the sample is in thermal equilibrium, the same detector positions were repeatedly measured two or three times and finally averaged. It should be noticed, that in a previously investigated Al-rich Ni-Al phase the antisite vacancy concentration was much higher, thus the scattering intensity was just high enough to be measured at ESRF. In the present Ni-rich phase the antisite vacancy concentration and thus the scattered intensity is much lower due to dominating Ni-ASDs and a resulting lower electronic density contrast. At the PETRA III synchrotron, however, high enough scattering intensities were obtained.

The resulting correlation function was fitted with a simple exponential decay. Reciprocal decay times for a fixed detector position, i.e., a given scattering vector \vec{q} are shown in Fig 1(a) for different temperatures. The fitted value for the activation enthalpy is about 2.1 eV. This agrees very well with values obtained by tracer measurements in this system [3], where the highest concentration in Ni-Al was 56.6 at.% Ni. The decreasing trend in activation enthalpy as a function of growing

Ni concentration shows a clear trend towards $Q = 2.1$ eV for 58 at.% Ni. This result confirms the accuracy of the XPCS measurement.

Of even greater interest is the variation of the correlation time depending on the orientation of \vec{q} with respect to the crystalline lattice at a fixed scattering angle $2\vartheta = 20^\circ$ (i.e. with constant length of $|\vec{q}|$) and at constant temperature, as shown in Fig 1(b). The broken line represents a preliminary fit indicating an effective movement of Ni atoms mainly along the edges of the cubic B2 lattice, i.e. in the $\langle 100 \rangle$ direction with only small admixture of body-diagonal $\langle 111 \rangle$ jumps. Nearest-neighbor jumps can thus be excluded, as well as cube face-diagonal $\langle 110 \rangle$ jumps. One should notice that using the XPCS technique we basically follow changes in the positions of Ni-ASDs which produce an electronic density contrast resulting in a speckle pattern. To find the correct atomistic picture, however, additional work is needed. Especially evaluation of the short-range order (SRO) is a challenging task. Knowledge of the SRO is necessary to correctly calculate the \vec{q} -dependence of correlation times, cp. theory from [1]. SRO of Ni-Al was measured in dedicated diffuse scattering experiments [4], unfortunately with insufficient quality.

In conclusion, we have successfully measured the diffusive dynamics in Ni-rich Ni-Al. This is, to our knowledge, the second measurement ever performed in a B2-ordered compound where the motion of majority atoms could be followed directly (the first being the investigation of iron motion in Fe-rich Fe-Al [5]). The atomistic picture of diffusion is different from that of Fe-Al and comprises second-nearest-neighbor jumps of nickel with small admixture of further jumps.

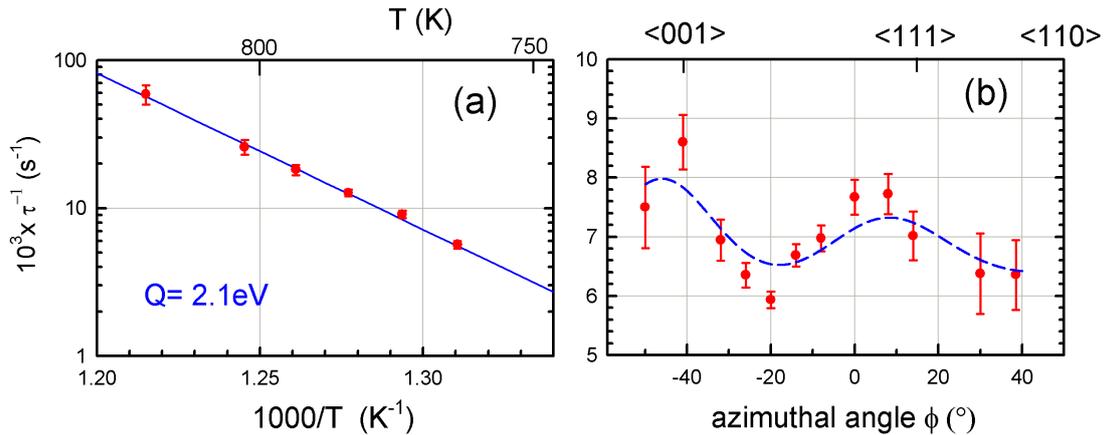


Figure 1: (a) Arrhenius plot of reciprocal correlation times. (b) The same values in $(1\bar{1}0)$ - plane measured at a constant temperature of 783K as a function of azimuthal angle.

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