NEUTRON LARMOR DIFFRACTION
FOR THE DETERMINATION OF ABSOLUTE LATTICE SPACING

J. Repper¹, T. Keller², M. Hofmann¹, C. Krempaszky³, W. Petry¹, E. Werner⁴

¹Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), TU München, 85747 Garching, Germany
²Max-Planck-Institut für Festkörperphysik, 70569 Stuttgart, Germany
³Christian-Doppler-Labor für Werkstoffmechanik von Hochleistungslegierungen, TU München, 85747 Garching, Germany
⁴Lehrstuhl für Werkstoffkunde und Werkstoffmechanik, TU München, 85747 Garching, Germany

ABSTRACT
The Neutron Larmor diffraction method is applied for the first time to determine absolute lattice spacing of a Silicon powder and polycrystalline IN718 samples. The absolute value of lattice spacing can be measured to an accuracy of $7 \cdot 10^{-5}$ Å. However, absolute determination of the lattice parameter is limited by the quality of the reference specimen used. Absolute lattice spacing determined by Larmor diffraction measurements of IN718 samples are shown and compared to the results obtained via conventional neutron diffraction.

INTRODUCTION
Residual stress analysis using neutron diffraction relies on the comparison of the lattice spacing $d$ of a stressed component with the lattice spacing $d_0$ of a macroscopically stress free reference sample. In neutron Larmor diffraction the lattice spacing $d$ is encoded in the spin precession of a polarized monochromatic neutron beam scattered at the sample. In contrast to conventional neutron diffraction methods, Larmor diffraction is independent of the divergence of the incident beam, the spread of the wavelength and the scattering angle $2\theta$ [1]. Based on this, the Larmor technique is insensitive to alignment errors of the sample, which may cause large shifts in measured $2\theta$ values in conventional diffractometer techniques [2]. Hence, the method enables the measurement of relative lattice spacing changes with a resolution of $\Delta d/d \sim 10^{-6}$ hitherto not possible using classical diffractometers [3].

In this paper we present for the first time results of the determination of absolute values for lattice spacing using neutron Larmor diffraction. The accuracy of the method is demonstrated for an NBS-Silicon powder sample with a well known lattice constant and is then applied to determine absolute lattice spacing of the industrially used Ni-base superalloy Inconel 718. The latter one is compared to results from conventional diffraction experiments.
This document was presented at the Denver X-ray Conference (DXC) on Applications of X-ray Analysis.

Sponsored by the International Centre for Diffraction Data (ICDD).

This document is provided by ICDD in cooperation with the authors and presenters of the DXC for the express purpose of educating the scientific community.

All copyrights for the document are retained by ICDD.

Usage is restricted for the purposes of education and scientific research.

DXC Website — www.dxcicdd.com

ICDD Website - www.icdd.com
METHOD
High resolution Larmor diffraction (LD) first proposed by Rekveldt [1, 4] is based on the neutron spin echo technique (NSE) with inclined magnetic field boundaries. In this technique the phase of a neutron spin (precession) in perfectly constant and homogeneous magnetic fields located before and after the sample is analysed. An comprehensive introduction to this method can be found in [5]. Due to technical reasons the static magnetic field $B$ is replaced by two pairs of rf spin flip coils in the so called neutron resonance spin echo (NRSE) method (figure 1). Whereas, usual NRSE applications work with the phase difference of the spin precession before and after scattering, LD sums the phases of precessions in both spectrometer arms. It allows the measurement of the lattice spacing with a typical resolution of $\Delta d/d \sim 10^{-6}$ for single crystal and powder samples [3]. This is at least one order of magnitude better than the resolution achieved by the best conventional neutron or X-ray diffractometers. The resolution is independent of the beam divergence and wavelength spread. Therefore, LD can be used with open collimation to ensure a high intensity beam. The basic principle of the LD technique is to mark each single neutron by a Larmor precession phase such that the phase only depends on the lattice spacing $d$ and is independent of the Bragg angle or the velocity of the single neutron. An initially polarized neutron beam runs through a homogeneous magnetic field $B$ with the field boundaries oriented parallel to the lattice planes of the diffracting crystal (figure 1a). The initial polarization is perpendicular to $B$. The total Larmor phase $\phi_{\text{tot}}$ after passing $B$ twice (incident and scattered beam) is given by the time $T$ the neutron spends in the field. The time $T$ only depends on the components $k_{1,2\perp}$ of the wave vector $k$ which are perpendicular to the field boundaries:

$$k_{1\perp} + k_{2\perp} = \text{const.}$$ [1].

Figure 1: a) Basic principle of Larmor diffraction b) Larmor diffraction setup at TRISP, FRM II. The magnetic field $B$ is replaced by two pairs of rf spin flip coils with a distance $L$. The field boundaries are defined by the precise windings of these coils. Tilting the lattice planes by a small angle $\alpha$ leaves the precession phase constant in a first order approximation ($k_{1\perp} + k_{2\perp} = \text{const.}$) [1].
The variable $\omega_L$ is the Larmor frequency. The parameter $L$ is the length of the magnetic field perpendicular to the lattice planes (compare figure 1), $m$ is the mass of the neutron and $\hbar$ is Planck’s constant. The required diffraction condition is given by the Bragg equation:

$$k \sin \theta_B = \frac{G}{2}$$

where $G/2=2\pi/d$ is the modulus of a reciprocal lattice vector. For magnetic field boundaries which are parallel to the diffracting planes, every neutron satisfying the diffraction condition has the same perpendicular component of wave vector $k_\perp = k_{2\perp} = G/2$ and, therefore, the same Larmor precession resulting from the magnetic fields located in the neutron path before and after the sample. Tilting the lattice planes by a small angle $\alpha$ (figure 1b) leaves the precession phase constant to the first order ($k_\perp + k_{2\perp} = \text{const.}$) [1]. Thus, the method also works for polycrystalline samples using beam divergences in the order of one degree and a relatively high intensity can be achieved.

The total phase of the neutron is given by

$$\Phi_{tot} = \frac{2m\omega_L L}{\pi \cdot \hbar} d.$$  \hspace{1cm} (3)

This phase is independent of the neutron wavelength and of the angle of incidence. For the determination of lattice spacing the total phases of two samples obtained after the same field length $L_{\text{ref}}$ will be compared. For an absolute value of the spacing the lattice spacing of the reference sample $d_{\text{ref}}$ must be known precisely. The unknown lattice spacing $d_{\text{sample}}$ of the second sample can then be calculated using equation (3) by a comparison of the total measured phases for the reference ($\Phi_{\text{tot,reference}}$) and the second sample ($\Phi_{\text{tot,sample}}$):

$$d_{\text{sample}} = \frac{\Phi_{\text{tot,sample}}}{\Phi_{\text{tot,reference}}} d_{\text{reference}}.$$ \hspace{1cm} (4)

For practical purposes the determination of total phases is difficult. The phase of the spin precession, however, is proportional to the number of spin precessions within the magnetic field length $L_{\text{ref}}$. Using the number of spin precessions of the sample ($N_{\text{sample}}$) and of the reference ($N_{\text{reference}}$) in equation (4) this gives

$$d_{\text{sample}} = \frac{N_{\text{sample}}}{N_{\text{reference}}} d_{\text{reference}}.$$ \hspace{1cm} (5)

The number $N_{\text{sample}}$ within $L_{\text{ref}}$ can be written as:

$$N_{\text{sample}} = N_{\text{int,sample}} + \Delta N_{\text{sample}},$$ \hspace{1cm} (6)
with \( N_{\text{ref,sample}} \) an integer and \( \Delta N_{\text{sample}} \) a neutron spin precession typically smaller 1. The number of spin precessions for the reference sample has to be an integer \((N_{\text{reference}} = N_{\text{int,reference}})\) due to physical boundary conditions. Hence, the integer numbers \( N_{\text{int,reference}} \) and \( N_{\text{int,sample}} \) can be determined free from errors. This, in addition with the large number of spin precessions (of about 450) within the field length \( L_{\text{ref}} \) allows the determination of absolute values of lattice spacing with a high accuracy. The Gaussian error propagation of the errors of \( t J_n.N_{\text{sample}} \) (typically \( \sim 10^{-3} \)) and \( d_{\text{reference}} \) results in errors in the order of \( 7 \times 10^{-5} \) Å for \( d_{\text{sample}} \). A detailed description goes beyond the scope of this report and will be published elsewhere.

**EXPERIMENTAL**

A powder sample of SRM640c silicon was used as specimen to verify the results of Larmor diffraction [9]. The lattice parameter \( a_{\text{silicon}} \) of this powder and therefore the lattice spacing \( d \) for lattice planes \( \{hkl\} \) were exactly determined in former experiments. A comparison of \( d_{\{hkl\},\text{theo}} \) calculated with \( a_{\text{silicon}} \) and \( d_{\{hkl\},\text{Larmor}} \) obtained by Larmor diffraction allows the verification of the results of a Larmor diffraction experiment. For a comparison of the results received by Larmor diffraction with results obtained in a conventional neutron diffractometer two sample sets of cylindrical Inconel 718 samples were measured. Each set consisted of three samples with different radii (1 mm, 5 mm and 10 mm). The samples were cut from two differently treated pancakes (diameter = 240 mm, thickness = 20 mm) at the radial position of \( r = 80 \) mm. Both pancakes were forged at a temperature of 990°C and cooled down to room temperature in different ways: The first disk was water quenched (WQ), whereas the second disk was cooled down in air (AC). The LD measurements were performed at the thermal neutron triple axis spectrometer TRISP [6] at FRM II (see figure 2), which incorporates the NRSE technique. A perfect Germanium single crystal (fcc) with a lattice parameter of \( a = 5.65726(7) \) Å [7] served as reference specimen. With this lattice parameter it is possible to calculate the lattice spacing for each \( \{hkl\} \) of the reference sample with the same error bars. An incident wave vector \( k_i \) of 3.53 Å\(^{-1} \) (\( \lambda = 1.78 \) Å) was used for the Germanium and the IN718 samples. For the measurements on silicon the incident wave vector was changed to \( k_i \) of 3.45 Å\(^{-1} \) (\( \lambda = 1.82 \) Å) to keep the scattering angle constant at \( 2\theta = 110^\circ \). Due to their similar lattice spacing, the (333) Bragg reflection of Germanium \( d_{\text{Ge(333)},\text{theo}} = 1.088740(70) \) Å and the (311) Bragg reflection of the IN718 Ni-matrix were chosen for the measurements, while for the Silicon powder the (422) reflection was used.

To gain a high neutron intensity on the sample position no collimation and an averaged beam size of approx. 40 x 40 mm\(^2\) were used. The count times needed for good counting statistics varied between 30 min and 4 hours for the IN718 samples due to different sample volumes. The count time for the Silicon powder was of about 11 hours. Due to the air conditioning in the experimental hall of FRM II the temperature fluctuations during the measurement of one sample
were smaller than 0.08K for all IN718 samples and about 0.16K for the Silicon powder, owing to the long measurement time. The thermal expansion coefficients are $\alpha_{\text{Silicon}} = 2 \times 10^{-6} \cdot \text{K}^{-1}$ for the Silicon powder and $\alpha_{\text{IN718}} = 11 \times 10^{-6} \cdot \text{K}^{-1}$ for the IN718 samples. Therefore, the changes in lattice spacing due to thermal expansion during the measurements are less than $10^{-6}$ for all measured samples and were neglected in the following discussion.

The conventional diffraction experiments were carried out at the neutron materials science diffractometer STRESS-SPEC at FRM II [8]. For a determination of absolute lattice spacing with conventional neutron diffraction it is necessary to know the exact wavelength, to determine the scattering angle $2\theta$ accurately and to align the sample with high precession. A wavelength of $\lambda = 1.5527(2) \, \text{Å}$ with an averaged beam size of $7 \times 20 \times 15 \, \text{mm}^3$ was selected. Because of the high absorption cross section of IN718, an absorption correction depending on the radius of each sample had to be undertaken.

RESULTS AND DISCUSSION

The theoretical $d$ spacing of the Si{422} Bragg reflection can be calculated from the lattice constant $a = 5.4311946(92) \, \text{Å}$ as $d_{\text{Si,th}} = 1.108638(9) \, \text{Å}$ [9]. The determination of the $d$-spacing from Larmor diffraction data is $d_{\text{Si,Larmor}} = 1.108620(71) \, \text{Å}$. This gives a difference of $\Delta d = 1.8 \times 10^{-5} \, \text{Å}$ (see table 1) which is within error bars. The relatively large error of $d_{\text{Si,Larmor}}$ ($7.1 \times 10^{-5} \, \text{Å}$) results from the large uncertainty in the lattice constant of the Germanium reference sample ($7 \times 10^{-5} \, \text{Å}$). Therefore, the precision of the determination of absolute lattice spacing is limited by the quality of the reference. The reduction of the error in the lattice spacing of the

![Figure 2: The thermal neutron triple axis spectrometer TRISP in the experimental hall of FRM II.](image)
reference specimen of one order of magnitude to $7 \cdot 10^{-6}$ Å would result in errors for d-spacing determined by Larmor diffraction of about $1 \cdot 10^{-5}$ Å.

Table 2 shows the $d$-values for the Ni{311} lattice planes of the IN718 samples as determined by Larmor diffraction and conventional neutron diffraction experiments. A good agreement of the two methods can be found for the samples with radii of 5 and 10 mm (differences in the order of $10^{-5} - 10^{-6}$ Å). In case of the 1 mm samples a larger difference in the values of the lattice spacing (in the order of $8 \cdot 10^{-4}$ Å) can be observed. This can be explained by the difficulties to align the sample in the center of gravity of the conventional diffractometer. Small shifts in the sample position can cause large shifts in the $2\theta$ values and as a consequence also in the lattice spacing $d$. For the larger samples small misalignment effects are negligible compared to the sample size. As discussed for the Si-values, also the errors in the absolute lattice spacing of the IN718 samples are relatively high using the Larmor diffraction method. However, they are much lower than the errors for lattice spacing determined by conventional neutron diffraction. In table 2 all lattice spacings are determined with data measured at TRISP compared with values determined with STRESS-SPEC data.
spacing show the same error of $7 \cdot 10^{-5} \text{ Å}$. This shows the dominate effect of the error in the lattice spacing of the reference sample (also $7 \cdot 10^{-5} \text{ Å}$). For a smaller error in the reference lattice spacing of about one order of magnitude, the errors would change from sample to sample, due to different counting statistics. A comparison of the lattice spacing of the different samples of the same sample state shows a relative resolution between $10^{-5} - 10^{-6} \text{ Å}$, as expected.

CONCLUSION
The possibility to determine absolute lattice spacing using neutron Larmor diffraction was shown. The potentially high accuracy (in the order of $7 \cdot 10^{-5} \text{ Å}$) of the method was demonstrated on a SRM640c Silicon powder with a well known lattice parameter. However, it was shown that the precision is mainly limited by the accuracy of the lattice spacing of the reference sample. Therefore the use of another reference sample as Germanium (e.g. Si) promises an even better accuracy. As our measurements on IN718 show, the absolute lattice spacing can be determined at least one order of magnitude more accurate using Larmor diffraction than by conventional neutron diffraction experiments. Furthermore, the insensitivity of the Larmor diffraction method to alignment errors was demonstrated. This offers the possibility to measure reference values $d_0$ used for the analysis of residual stresses by conventional diffraction methods with a high accuracy. The usable large beam cross section of $40 \times 40 \text{ mm}^2$ provides the opportunity to measure accurate stress free reference values also at strained samples for which it can be difficult to cut a reference sample. This method also takes the mean of all inhomogeneities, e.g. in chemical composition, affecting the scattering angle $2\theta$ and therefore the reference value in a conventional diffraction experiment. The use of smaller gauge volumes is possible, but increases the measurement time. In addition another Larmor diffraction instrument set up allows the determination of fluctuations around mean value of absolute lattice spacing. The combination of this set up with the measurement method presented here, has the potential to deliver information about crystallographic phases not resolvable with conventional diffractometers. Results will be published elsewhere.

ACKNOWLEDGMENTS
The authors acknowledge the FRM II for providing beam time and the DFG for funding this research within projects WE 2351/11-1 and PE 580/7-1.

REFERENCES