Textures and Microstructures, 1989, Vol. 10, pp. 309–323
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Printed in the United Kingdom

Texture Investigations by Neutron Time-of-Flight Diffraction

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(Received March 22, 1988)

For the majority of isotopes the thermal neutron absorption cross section is two or more orders lower than that for X-rays. This makes neutron diffraction well-suited for bulk texture investigations. Some characteristics of neutron diffraction are discussed. The principles of neutron time-of-flight diffraction are described. The pole figure determination by means of TOF technique is considered. The main parameters of the present Dubna texture facility are given. Further developments of the experimental technique are considered. The application of the TOF technique for inverse pole figure measurement is discussed as an approach to direct observation of the texture forming process. The magnetic moments of neutrons can be used to study magnetic textures. Two different techniques are discussed.

KEY WORDS: Neutron diffraction, Time-of-flight method, Pole figures, Inverse pole figures, In-situ measurements, Magnetic scattering.

INTRODUCTION

Modern quantitative texture analysis is based mainly on spectroscopic methods like X-ray or thermal neutron diffraction. Low absorption of neutrons by most of isotopes makes them well-suited for bulk texture studies in large specimen volumes up to 10 cm^3 and more. Texture inhomogeneities are completely averaged in the commonly used techniques. Therefore, neutron diffraction permits to investigate relatively coarse grained aggregates like those often

found in selected metallic materials and in petrofabric problems as well. Furthermore, complete pole figures can be determined without special preparation techniques combining reflection and transmission geometry measurements.

At present, texture investigation facilities at various neutron sources are in operation. Different variants of the angle dispersive neutron diffraction at stationary reactors are widely used, which are similar to the well-known diffraction of monochromatic X-rays. In the previous five years powerful pulsed neutron sources were put into operation (see, e.g. Ananiev *et al.*, 1985 and Leadbetter, 1985). On this base the energy dispersive neutron time-of-flight diffraction is demonstrated by Feldmann (1986) to broaden the spectrum of problems for texture investigations in the direction of low symmetric and multiple phase material.

In the present paper a review is given of the time-of-flight diffraction technique and its application to investigations of various texture problems.

REMARKS ON NEUTRON DIFFRACTION

Some aspects of neutron scattering should be remembered, which may be important in connection with texture investigations. For more detail see the monographs of Bacon (1975) and Windsor (1981). Comparing considerations concerning neutron and X-ray methods were done by Kleinstück *et al.* (1976) and Welch (1986).

Neutron scattering takes place due to the interaction of neutrons with nuclei, but also of neutron spins with the magnetic moments of atomic electron shells in the investigated matter. In usual diffraction experiments the total neutron scattering is measured consisting of elastic and inelastic components. The inelastic part is of less order of magnitude and contributes to the background only. The elastic scattering is described by the differential elastic scattering cross section

$$d\delta/d\Omega = d\delta/d\Omega_{\mathrm{coh}}^{\mathrm{nucl}} + d\delta/d\Omega_{\mathrm{coh}}^{\mathrm{magn}} + d\delta/d\Omega_{\mathrm{inc}} \qquad (1)$$

consisting in general of nuclear and magnetic coherent components as well as an incoherent part. For texture studies information can be obtained from the coherent scattering only.

For the most of isotopes the incoherent scattering is of low intensity. However, for hydrogen it is extremely predominant. Therefore, neutron diffraction measurements including texture studies at materials containing hydrogen like polymers are very complicated. These difficulties can be overcome using deuterated specimens.

Magnetic diffraction takes place only if there is a magnetic ordering of any kind in the studied material. Often, the atomic and magnetic lattices coincide. In this case, there are problems to separate the nuclear from the magnetic component of the Bragg reflection. A non-coincidence of magnetic and conventional lattice texture may lead to inaccuracies in the texture analysis.

Neutron diffraction is determined by the Bragg's law

$$\lambda = 2d_{hkl} \sin \theta \tag{2}$$

There are two possibilities to satisfy this equation for discrete lattice spacings d_{hkl} leading to two different experimental techniques:

- The incident beam is monochromatic. The Bragg angle is varied (conventional angle dispersive method).
- A polychromatic beam consisting of a certain wavelength spectrum is used at constant scattering angle 2θ (time-of-flight (TOF) method).

The following consideration is restricted to the TOF technique.

NEUTRON TIME-OF-FLIGHT DIFFRACTION

The Bragg condition can be satisfied for various lattice spacings d_{hkl} at a fixed scattering angle, if the incident beam consists of polychromatic neutrons. Since there are no wavelength or energy sensitive neutron detectors, the time of flight through a certain distance is measured to determine the energy of a given neutron. Such an experiment can be done if the start time of the considered neutron is known. Therefore, not continuous, but pulsed beams have to be used. In Figure 1 the lay-out of a TOF diffraction experiment is shown. If a neutron with energy E starts from the source at the moment t_0 it is recorded in the detector at time t

$$t - t_0 = T = (L_1 + L_2)(2m/E)^{1/2} \tag{3}$$

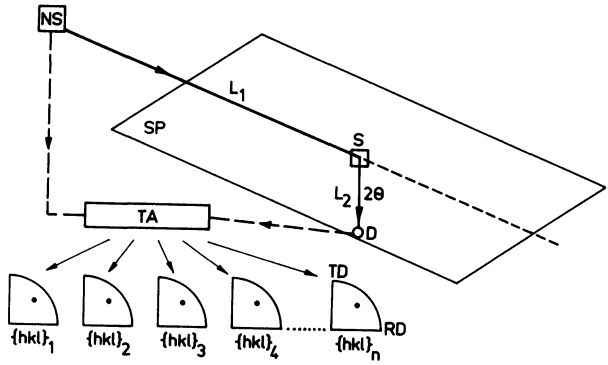

Figure 1 TOF diffraction experiment. TA-multichannel time analyzer, L_1, flight path of the primary beam, L_2, distance from the specimen to the detector. In each pole figure one point at equivalent positions is recorded by one Bragg pattern simultaneously.

The detector signal is stored in a multichannel time analyzer (TA) which is started synchronously with each pulse emission. Thus, the spectrum of recorded counts is built up in dependence on neutron flight time. The energy and the wavelength of a given neutron are correlated by

$$\lambda = 2\pi h/(2mE)^{1/2} \tag{4}$$

where m is the neutron mass and h the Plank's constant. Then the relation between the time of flight, total flight path and wavelength is given by

$$\lambda = T/(b(L_1 + L_2)) \qquad b = 2.528 \times 10^6 \,\text{sec/m}^2 \tag{5}$$

where L_2 is the distance from the specimen to the detector.

RESOLUTION PROBLEMS

Any pulsed neutron source produces pulses of fast neutrons having a finite width. For condensed matter studies thermal or even epithermal neutrons in the energy range from about 10^{-3} eV up to 10 eV are required in general. Therefore, fast neutrons are moderated to a pulse containing a Maxwell-like thermal energy spectrum

as well as remaining high energy neutrons. For commonly used moderators the maximum of the Maxwell-distribution is at 0.15 nm approximately. The time dependent pulse shape is asymmetric now, having a steep raise but a smoother slope. Its width is in the range of 100 μs or more.

The pulse width strongly influences the resolution of the TOF diffraction experiment. Figure 2 illustrates the situation. At the moderator surface x_0 the pulse is composed of all wavelengths. After a short flight path x_1 the pulses for different wave lengths are slightly separated but still overlapped. Arriving at the long distance point x_2 the pulses for the same wavelengths are separated completely with respect to their time of flight.

Therefore, high resolution is obtained by the use of long flight paths. On the other hand, the beam intensity goes down proportional to the square of the flight path. An optimum has to be found. To decrease the loss of intensity neutron guide tubes are widely used. The intensity gain is obtained at the expense of beam divergency using the total reflection of neutrons at metallic mirrors (especially Ni). The critical angle of total reflection is proportional to the neutron wavelength. Therefore, the use of neutron guide tubes may influence the resolution power considerably for long wavelengths.

Considering the vertical const.$-\theta$ line in Figure 3 representing Bragg's law for the TOF experiment, all nonforbidden diffraction peaks are recorded at a fixed Bragg angle simultaneously. The

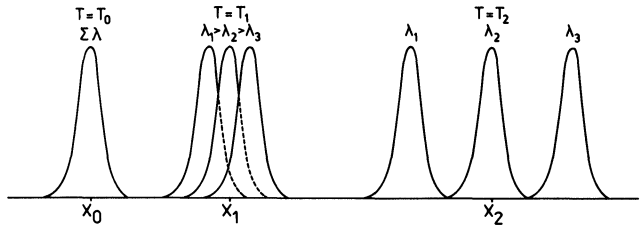

Figure 2 Behaviour of the polycromatic neutron pulse. At time T_0 the pulse leaves the moderator surface. It contains all wavelengths. After a short time T_1 the λ_2-neutrons arrive at the point x_1. The λ_3-neutrons have passed it already, the λ_1 neutrons are slower. Passing a long flight path x_2 (long time T_2) the three different wavelength pulses are separated completely.

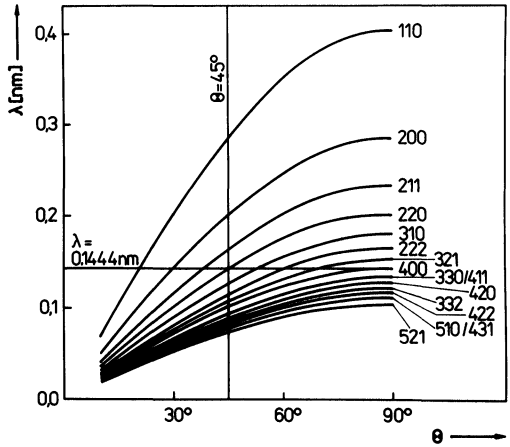

Figure 3 Bragg condition for a bcc-lattice. The horizontal line represents the angle dispersive and the vertical one the TOF diffraction experiment.

low index reflections are favoured for investigation, because the measured intensity is proportional to λ^4 in the TOF method. Moreover, the resolution of peaks improves with increasing wavelength. As an example, Figure 4 shows the TOF diffraction pattern of copper.

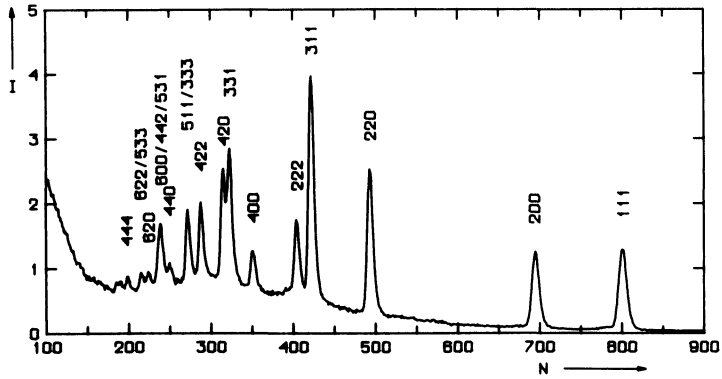

Figure 4 TOF diffraction pattern of copper (intensity versus number of time channel).

NEUTRON TIME-OF-FLIGHT TEXTURE ANALYSIS

In the time-of-flight method like in conventional neutronographic studies, too, complete pole figures can be determined combining transmission and reflection geometry measurements. Eulerian cradles or triple axis goniometers having one vertical and two mutually perpendicular horizontal axes are used for pole figure scanning. The point net may be an equal angle net an equal area net or even any other one.

In the TOF method pole figures from all separable reflections are measured simultaneously by only one scan (see Figure 1). Therefore, the experimental expense is almost independent of the required quantity of pole figures, i.e. the TOF technique becomes more efficient, if the number of necessary pole figures increases. Consequently, the method is especially suitable for preferred orientation studies in samples having low lattice symmetry or in multiple phase materials like those often found in geological specimens (Feldmann *et al.*, 1986).

The representation of reflections as well as background by several points of the spectra increases the statistical reliability of the pole figure data. This advantage can be used to provide very accurate texture investigations.

In TOF diffraction extended area detectors and large cross section neutron beams are available to investigate relatively coarse grained materials.

For the determination of pole density values a computer fit of Bragg reflections has to be done. The information of overlapped peaks may be used. The number of separable pole figures is restricted by the spectrometer resolution, the efficiency of the available fit programme and the lattice symmetry of the investigated sample. To carry out the line profile analysis the diffraction patterns must be normalized with respect to wavelength independent neutron flux. The wavelength distribution of the primary beam is reflected by the spectrum of an incoherent scatterer like vanadium.

The absorption of neutrons by the most of isotopes is lower than that of X-rays by a factor of typically 10^3 to 10^4 (Welch, 1986). Therefore, thick samples up to the order of a centimeter can be measured even in transmission geometry. By neutronographic methods the average volume texture is studied. Surface effects in

general do not influence the result. Consequently, the requirements to specimen preparation are very low.

Nevertheless, pole figure data have to be corrected for beam weakening and effectively irradiated volume at every sample position during the pole figure scan. This procedure may be avoided without loss in accuracy using spherical specimens as described by Tobisch and Bunge (1972). In this method a cube can be considered approximately as a sphere. The absorption coefficient can be calculated correctly, if the sample is bounded by parallel planes like rolled metal sheets. The errors of texture analysis are shown by Bunge (1982) to be significantly less for neutron than for X-ray diffraction.

The absorption coefficient has not to be determined separately in the TOF technique, if the incoherent cross section of the specimen is not too low. Then, the incoherent part of each spectrum or even a range of it can be used for its own normalization. Figure 5 shows the interesting region of the diffraction pattern of a quartzitic rock having high incoherent background. In such cases the influences of weakening and effectively irradiated volume as well as detector and source fluctuations are eliminated also. Multiple diffraction effects, which are increasing with increasing path of neutrons in the sample and the influence of different specimen ranges contributing to the scattering during the pole figure scan remain incorrected because the incoherent scattering does not depend on them. If this proce-

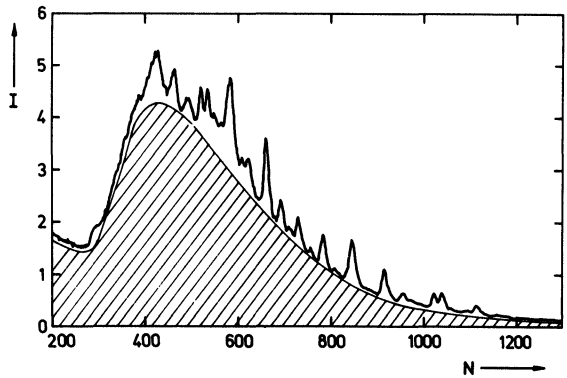

Figure 5 TOF diffraction pattern of a basal granitoid (quartzitic rock). The incoherent background is represented by the shadowed area.

dure is applicable, the only requirement to the sample shape is its thickness.

Comparing with the conventional angle dispersive method, the TOF texture analysis is less convenient concerning the amount of experimental data and its handling. For each sample a set in the order of 10^3 points of the corresponding spectrum has to be stored. Pole density values are determined only via line profile analysis of diffraction patterns. On the other hand, TOF diffraction may be applied to solve a very wide spectrum of preferred orientation problems.

PARAMETERS OF THE DUBNA SPECTROMETER

The present version of the Dubna texture facility NSWR at the IBR-2 pulsed reactor is described in detail by Ananiev *et al.* (1984). The thermal neutron pulse having a slightly wavelength dependent pulse duration of about 300 μs hits the sample at a distance of 32.6 m from the reactor. There the time averaged neutron flux is 3×10^6 n/cm^{2}s. The beam is guided in an evacuated steel tube and collimated to a circular cross section of 50 mm in diameter. The distance from the specimen to the detector is 1.6 m. The horizontal divergency of the primary beam is 0.12°, the minimum divergency of the scattered beam is 0.2°. A resolving power of 1% is achieved for the range of long wavelengths (see Figure 6). A triple axis texture goniometer permits to carry out angle steps down to ±1 degree around each axis. The diameter of the goniometer is 250 mm. The double Bragg angle is variable from 10° to 170°. The exposition time for one sample setting varies from some minutes for

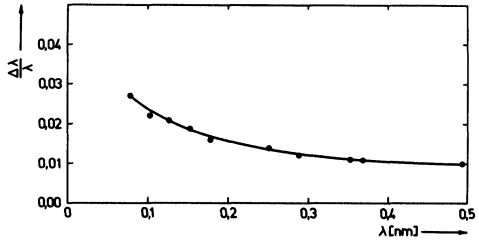

Figure 6 Experimental resolution of the Dubna spectrometer.

metals like iron and copper to about one hour for complex low symmetric metamorphic rocks.

At the IBR-2 reactor a higher resolution texture spectrometer at a 100 m flight path is to go into operation. It is equipped with a neutron mirror guide tube from the source to the specimen. With the beam cross section of $5 \times 17\ cm^2$ it will be especially suitable for preferred orientation studies in metamorphic rocks.

FURTHER EXPERIMENTAL DEVELOPMENTS

Compared with the conventional neutron texture analysis the exposition time in the TOF method is long (see Welch, 1986). To fasten the measurements, similar considerations may be done as in monochromatic beam technique. Using the geometry equivalent to that one at the Risø National Laboratory (Juul Jensen, 1986) the detectors have to be situated on a circle around the primary beam to avoid flight path differences (Figure 7). Therefore, in the TOF method the counters may be placed on any Debye-Scherrer cone, i.e. not necessarily in the plane perpendicular to the incoming beam. The application of discrete counters on the circle segment instead of quasicontinuous position sensitive detectors (PSD) diminishes the amount of experimental data. Points are measured as shown in Figure 7 at equivalent positions in all considered pole figures.

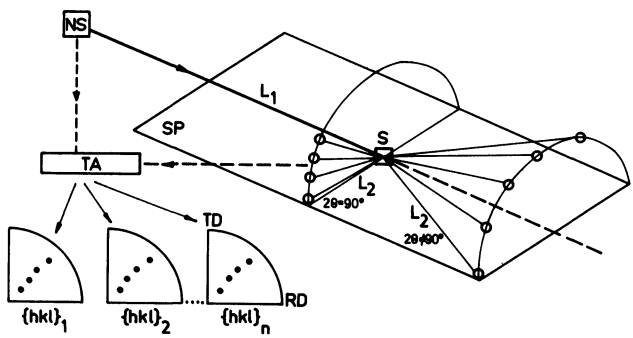

Figure 7 Intensity distribution measurement along a (*hkl*) Debye–Scherrer cone using a multidetector.

Therefore, the detector positions should be chosen with respect to the point net on the pole figure. The pole figure tilt angle depends on the angle between detector and horizontal scattering plane in a nonlinear way. Consequently, the counter areas have to be changed strongly from one to another to ensure equal pole figure windows. Up to now, this geometry for TOF texture facilities is under discussion only.

Another possibility to fasten the measurement is the application of several detectors situated in the scattering plane at different Bragg angles (see Figure 8). The experimental geometry is similar to that one applied by Bunge *et al.* (1986) at the ILL in Grenoble using a PSD which is situated in the scattering plane. In opposite to the PSD at the stationary reactor every counter records the complete diffraction pattern. Therefore, the detector positions may be chosen with respect to the pole figure point net in a way to rotate the specimen only around the vertical goniometer axis and the sample normal for a complete pole figure scan. This geometry avoids the strong beam defocusation at tilting around a horizontal axis. Pole figure points at different tilt angles are corresponding to various Bragg angles, i.e. to other resolution ranges of the spectrum. Therefore, a mutual calibration of them is necessary. At the JINR two detectors are used at present (Drechsler *et al.*, 1987). In the nearest future the measurements will be carried out with six counters simultaneously.

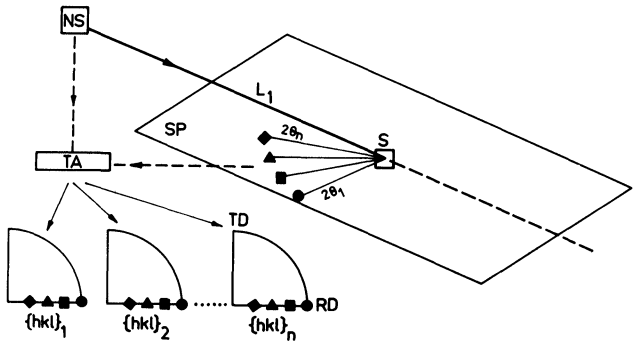

Figure 8 Application of a horizontal multidetector.

INVERSE POLE FIGURE MEASUREMENT

A TOF diffraction pattern consists of all nonforbidden Bragg reflections of the investigated sample measured at constant scattering geometry. This means the information of one TOF spectrum is equivalent to that of the inverse pole figure for the corresponding specimen position. Unfortunately, the available number of separable reflections and their inhomogeneous distribution over the inverse pole figure range complicate sufficiently the procedure of mathematical texture analysis. The majority of experimental points is situated on symmetry lines.

For inverse pole figure construction the TOF diffraction patterns have to be corrected with respect to all wavelength dependent influences, especially the incoming neutron spectrum. This procedure may be somewhat uncertain. Therefore, it seems to be more straightforward to compare the spectra with a measurement at a random specimen.

Nevertheless, the constant geometry makes the TOF diffraction well-suited for the observation of texture component formation in in-situ experiments. Figure 9 shows the behaviour of four low index reflections of copper during the recrystallization process studied by Matz and Feldmann (1982). The exposition time of 30 minutes per spectrum is too long to observe technically relevant processes. Recently Balagurov and Mironova (1986) reported on times of less than 1 minute to observe at the IBR-2 reactor the reaction kinetics of hydration of $Ca_3Al_2O_6$ having very poor scattering parameters. Such exposition times seem to be promising for in-situ texture investigations as well.

RESIDUAL STRESS ANALYSIS

The line profile analysis is applied to determine pole figure values in the TOF technique. The necessary computer program may be used to fit the position, the height and the line width of the studied peak too. Therefore, these methods include the principle possibility to extract information on residual stress anisotropy of the investigated sample besides the texture data from diffraction patterns. Special efforts have to be made to ensure sufficient resolution parameters of the experimental equipment.

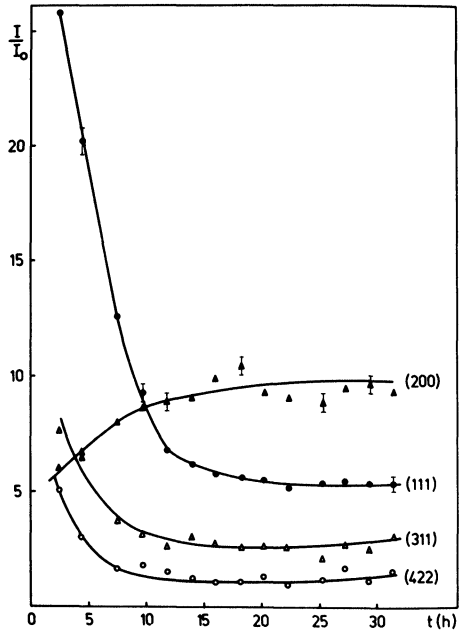

Figure 9 Intensity versus annealing time for four Cu reflections.

MAGNETIC TEXTURE STUDY

Attempts have been made to apply neutron diffraction for investigations of magnetic sublattice anisotropies in magnetically ordered materials (Hennig *et al.* 1983). There are two methods to separate magnetic from nuclear diffraction:

- Contrary to the lattice diffraction the intensity of magnetic reflections decreases with increasing sin θ/λ, i.e. the difference in various order pole figures is expected to represent the pure magnetic texture part. Corresponding careful measurements have been done without satisfactory results. Extinction effects are supposed to be the reason.
- The magnetic diffraction can be suppressed completely, if the specimen is magnetized up to its saturation with magnetization direction parallel to the diffraction vector. The difference in pole

figures measured without and with the field, respectively, represents their magnetic components. Of course, such experiments are not reproducible. Furthermore, a diamagnetic goniometer must be used to avoid strong forces on it in the saturation field.

The TOF method is well-suited to purse both possibilities. In the diffraction pattern the different order reflections are completely separated and not mutually influenced like in the conventional technique. Carrying out measurements in a magnetic field the constant scattering geometry for all pole figures permits to choose an adequate counter position with respect to the dimensions of the magnet.

CONCLUSIONS

Thermal neutron diffraction is shown to be an efficient tool for bulk texture analysis via pole figure determination. The method is applicable for the most of materials with the exception of hydrogen containing substances, e.g. of polymers.

The neutron time-of-flight diffraction permits to study a wide range of texture problems in metallurgy and metal physics as well as in petrofabric analysis. Especially, it is well-suited to investigate low symmetric and multiple phase materials up to rather complex sample compositions. At present, its main drawback are the relatively long exposition times. The application of discrete multidetectors decrease the time for measurements without difficulties in pole figure scanning.

Very short exposition times in TOF method may be promising to observe the kinetics of texture formation caused by external influences immediately.

Magnetic texture studies are confronted by methodical difficulties up to now. Further progress will however be made in the future.

Acknowledgement

The author wishes to thank M. Betzl and A. Mücklich (CINR Rossendorf), J. Tobisch (TU Dresden) and K. Walther (JINR, Dubna) for helpful discussions.

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