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ANALYSIS OF TEXTURE DISTRIBUTION IN NdFeB HARD MAGNETS BY MEANS OF X-RAY DIFFRACTION IN BRAGG-BRENTANO GEOMETRY

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Three different methods are used for the quantitative local texture analysis of fibre textures. Two methods are based upon the evaluation of BRAGG-BRENTANO diffraction diagrams. In the first method relative intensity parameters are calculated, whereas in the second Rietveld preferred orientation factors are determined. The third method uses measured pole figures and the series expansion (Bunge).

The method is applied to hot backward extruded NdFeB ring magnets. A local texture determination dependent on a sample surface distance is possible. The results are compared with magnetic measurements.

KEY WORDS: Local texture analysis, texture inhomogenities, Rietveld, NdFeB, backward extrusion, ring magnets

INTRODUCTION

Magnetic properties of polycrystalline ferromagnetic materials are influenced by preferred crystallographic orientation (texture) due to direction dependence of the magnetic crystal anisotropy. By means of formation of a certain texture application properties can be improved.

The texture is completely described by the three-dimensional orientation distribution function (ODF), which can be determined from pole figure measurements using X-ray diffraction. The characterization of texture inhomogeneities by means of pole figure measurements is time consuming. For simple distributions of crystalline orientations as fibre texture it is not necessary to measure complete pole figures. The diffracted intensities in BRAGG-BRENTANO geometry of a given lattice plane (hkl) are proportional to the volume fraction of crystallites with orientation (hkl) perpendicular to the sample surface. The measurement of only one BRAGG-BRENTANO diagram enables a reduction of measuring time.

The aim of this paper was the quantitative analysis of texture distribution by means of BRAGG-BRENTANO measurements. In the first part a quantitative comparison of the texture parameter of a sintered $Fe_{14}Nd_2B$ hard magnet obtained from pole figure measurement is given with those from BRAGG-BRENTANO measurement. The ODF shows the existence of a fibre texture in this material with similar texture parameters as estimated from the BRAGG-BRENTANO measurements. In a second part the texture distribution is backward hot-pressed $Fe_{14}Nd_2B$ magnets was investigated by means of this method. The correlation to the magnetic properties is discussed in part 3.

EXPERIMENTAL PROCEDURE

Two kinds of NdFeB hard magnets were investigated:

First, $Nd_{14.9}Fe_{77.7}Al_{1.5}B_{5.9}$ magnets were prepared in the usually powder metallurgical way by crushing and milling the cast material. To obtain an alignment of powder particles an applied field $H = 2.0T$ was used during the compaction process. Subsequently, the green compacts were sintered at 1065°C for 60 min and then annealed at 620° for 30 min to obtain a good coercivity. The achieved dimensions of the samples are 5 * 5 * 5 mm.

Secondly, NdFeB hard magnets were prepared by hot backward extrusion. Whereas hot deformation by the die up-setting process results in an alignment parallel to the pressing direction, the backward extrusion (Figure 1) can be used to produce radially oriented ring magnets. This is much advantageous because the magnetic properties of these rings surpass those of sintered ring magnets. Commercial melt quenching powder (MQP) was used as starting material. It was compacted in vacuum at 725°C to a dense preform. The backward extrusion was performed in an inert gas at 800°C. The ring magnets have an outer diameter of 30 mm, an inner diameter of 24 mm, and a height of about 30 mm (Grünberger, 1995).

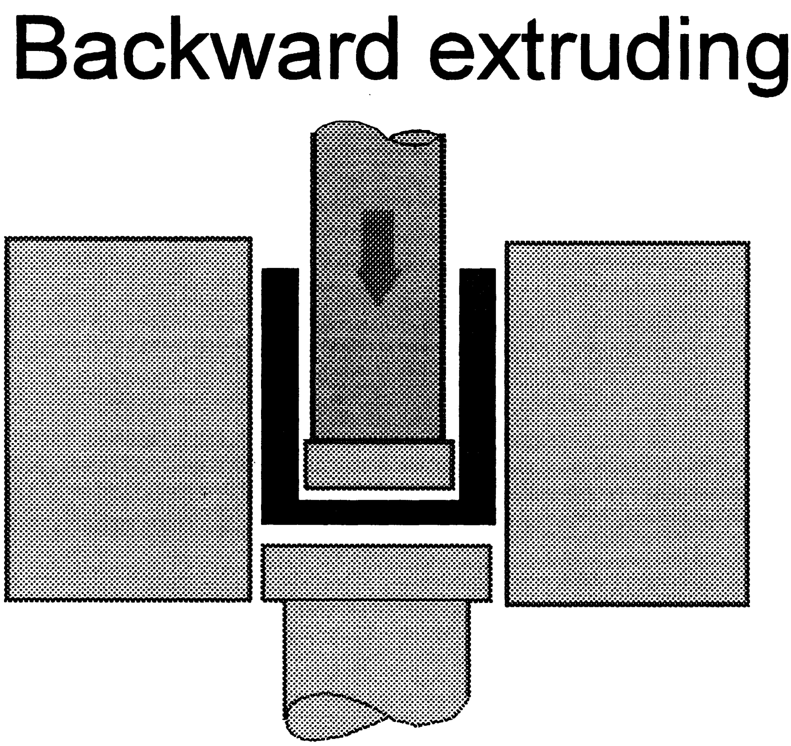

Figure 1 The backward extrusion process.

The magnetic properties of the samples $\leq 3 * 3 * 3$ mm^3 were measured with a vibrating sample magnetometer (VSM) after external magnetization in a pulsed field of $\mu_0 H = 7$ T. In order to investigate the positional dependence in the cross-section, the sample planes lying perpendicular to the radial direction were successively ground. The material was removed from the outer surface of the sample.

The pole figure measurements were carried out with a Seifert texture goniometer in reflection geometry using Co $K\alpha$ radiation. The range for the tilt angle ψ was chosen between 0° and 70° and for the rotation angle φ between 0° and 360° in 5° intervals.

The BRAGG-BRENTANO diagrams were measured with a Philips PW3020 diffractometer. For the determination of the texture distribution in radial direction of the backward extruded hard magnets, the inner surface of a ring segment was ground planar in several steps to the outer surface.

THEORETICAL BACKGROUND

Orientation Distribution Function (ODF)

From the measurement of several pole figures with independent **hkl** the three-dimensional orientation distribution function (ODF) was calculated using the series expansion method (Bunge, 1982; Dahms 1987). The ODF $f(g)$ describes the volume fraction dV of crystallites with a specific orientation g with respect to a sample reference system.

$$\frac{dV}{V} = f(g) * dg \tag{1}$$

V is the volume of the whole sample.

The orientation distribution function (ODF) is expressed in Euler angles $(\varphi_1, \Phi, \varphi_2)$. The ODF is described by symmetric generalized spherical harmonic terms $\dot{\dot{T}}_l^{\mu\nu}$

$$f(\varphi_1, \Phi, \varphi_2) = \sum_{l=0}^{\infty} \sum_{\mu=1}^{M(l)} \sum_{\nu=1}^{N(l)} C_l^{\mu\nu} \dot{\dot{T}}_l^{\mu\nu} (\varphi_1, \Phi, \varphi_2) \tag{2}$$

with the coefficient $C_l^{\mu\nu}$.

The complete ODF allows the calculation of unmeasurable pole figures. In our case the basal pole figure (*001*) is used, which is preferredly situated parallel to the sample surface.

A cut through the calculated (001)-pole figure leads to the profile of the preferred orientation distribution.

Relative Intensity Parameters

A requirement for this method is a fibre textured sample symmetry. The method is based on the comparison of a BRAGG-BRENTANO diagram of a textured sample to a random one (Lima, 1994).

For a texture analysis the intensities of distinguish peaks of the textured and the random samples are determined. The relative intensities are calculated according to:

$$I_{rel,tex} = \frac{I_{tex}}{\Sigma I_{tex}} \tag{3}$$

$$I_{rel,random} = \frac{I_{random}}{\Sigma I_{random}} \tag{4}$$

$$I_{rel} = \frac{I_{rel,tex}}{I_{rel,random}} \tag{5}$$

The relative intensities of reflections can be presented as a function of their angle to the fibre-axis. Then, the diagram corresponds to a pole figure cut through a rotational-symmetrically fibre texture. The distribution of the relative intensities versus the angle to the (001)-plane is described by a distribution function. The shape and the FWHM (Full Width Half Maximum) of the distribution function characterize the anisotropy of the sample.

Rietveld Analysis

The Rietveld method is a least-squares refinement obtained between observed powder diffraction pattern and the calculated pattern based on the simultaneously refined models for the crystal structure and other specimen characteristics (e.g. lattice parameters, preferred orientation, instrumental factors) (Young, 1993).

The calculated counts $\mathbf{y}_c$ are determined by summing up the distributions from neighbouring Bragg reflections **K**, plus the background $\mathbf{y}_b$:

$$y_c = sS_rA\,\Sigma\,[|F_k|^2\Phi(2\theta - 2\theta_k)L_kP_k] + y_b \tag{6}$$

where

s is a scale factor.
S_r is a function to model the effects of surface roughness.
A is an absorption factor.
F_k is the structure factor.
Φ is a reflection profile function.
L_k contains the Lorentz, polarization and multiplicity factors.
P_k is a preferred orientation function.

Diffracted intensities from axially symmetric specimens can be corrected for preferred orientation with a single pole-density function P_k. A convenient procedure is to approximate the profile with a function, the variable parameters of which fitted during structure refinement.

The March-Dollase function (Dollase, 1986) as a preferred orientation model has in comparison to other functions a number of advantages for samples with a strong single-pole orientation:

1. it has a valid theoretical basis related to the major mechanism (grain rotation) that produces preferred orientation
2. it is a true probability distribution with unit integral
3. the same expression applies to disc and rod grain shapes
4. it has a single variable parameter

The March-Dollase function is defined as

$$P_K = (G^2 cos^2 \alpha + (1/G) sin^2 \alpha)^{-3/2} \tag{7}$$

with G as the preferred orientation parameter and α as the tilt angle to the preferredly oriented plane (in our case 001).

The function describes the pole-density distribution produced by rigid-body rotation of inequant crystallites (i.e. crystallites with unequal sides) upon axially symmetric volume. The value $G = 1$ for an isotropic sample and $G = 0$ for an ideally oriented one.

RESULTS AND DISCUSSION

Texture Parameters of Sintered NdFeB Magnets

Figure 2 shows the obtained diffraction diagrams of a random preform sample and a textured one. The reflections indicate the tetragonal $Nd_2Fe_{14}B$ phase (space group: P42/mnm). The intensities of the preform sample correspond to those of a standard powder diagram with random orientation distribution. The textured sample shows preferably the (001)-reflections. Therefore, the c-axis is aligned perpendicular to the surface of the sample. The Rietveld evaluation of the textured sample is presented in the third curve.

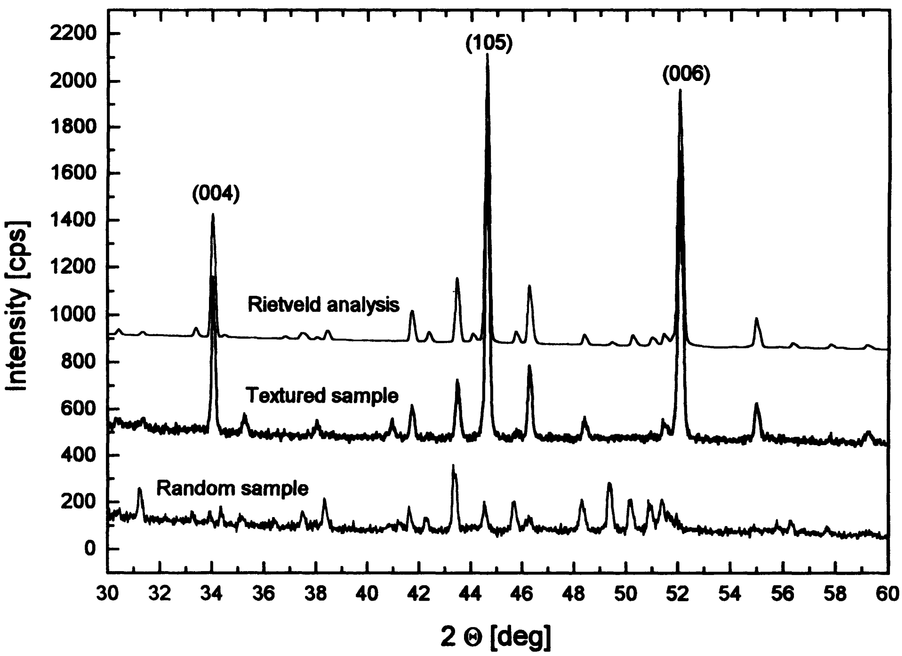

Figure 2 The Θ-2Θ-diagrams for a random, a textured NdFeB sample, and the Rietveld analysis for the textured one. The curves are shifted vertically with an offset.

Figure 3 shows the ODF of a sintered NdFeB magnet. The maxima are situated on a line with $\phi = 0°$ and $\varphi_2 = 0°$ in the Euler space. The position of the line is nearly independent on the angle φ_1. The location corresponds to an orientation of the (001) basal plane perpendicular to the applied magnetic field $\vec{H}$. The result confirms, that the sample has a fibre texture in the c-axis direction. Therefore, the application of the Rietveld analysis and the Relative Intensity Parameter method are possible.

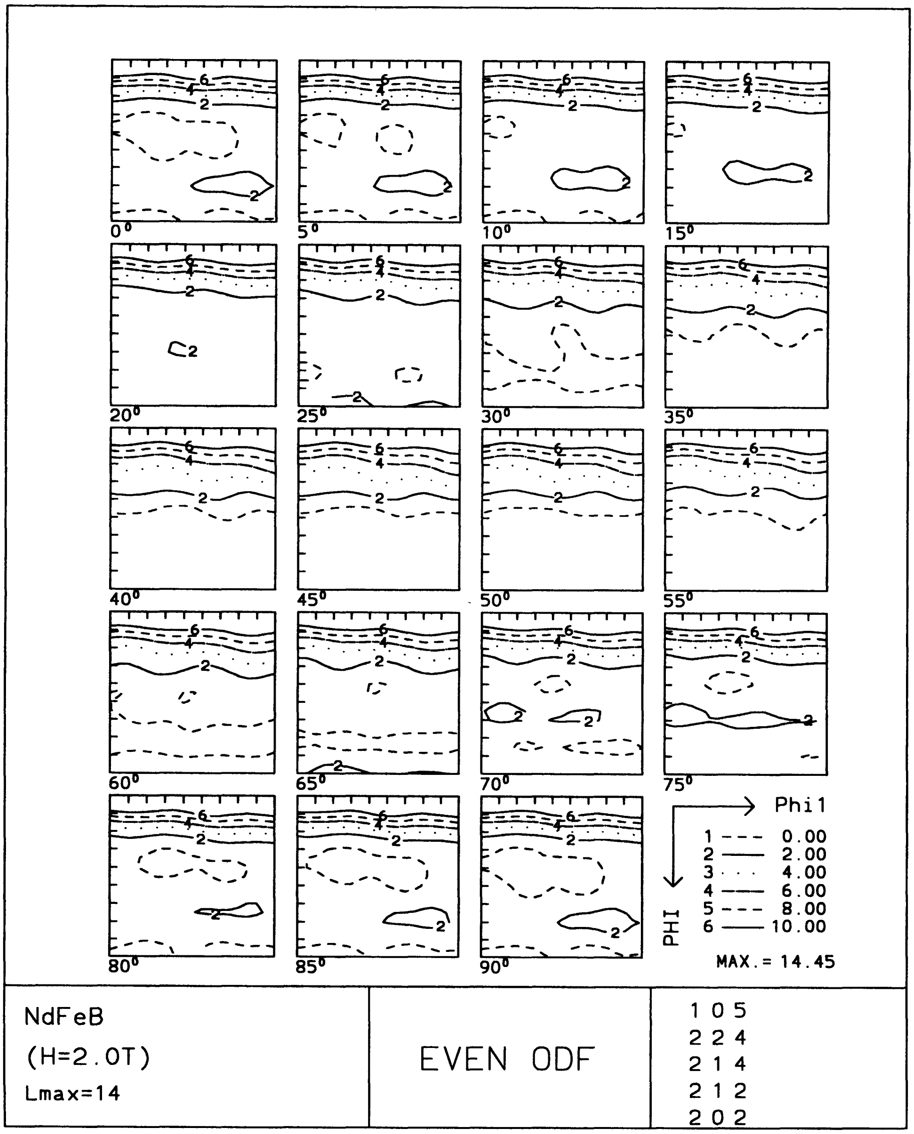

Figure 3 The orientation distribution function (ODF) of a NdFeB magnet pressed in the magnetic field.

Figure 4 shows a comparison of the different methods at the same NdFeB-sample. The cut of the recalculated (001)-pole figure (curve 1) has the largest FWHM (Full Width Half Maximum). The reason can probably be found in the low series expansion degree. For a strongly preferred orientation in the tetragonal lattice a lot of pole figures are necessary, but the superimposed peaks prevent a sufficient number of measurements for a high series expansion degree. Therefore, the calculated textures with classical texture programs are too weak.

The other curves represent an adaptation of BRAGG-BRENTANO diagrams. The calculated Relative Intensity Parameters versus the tilt angle are fitted with a Lorentz function (curve 2). The difference between the second and third curve is, that the relative intensity parameters reflect only a determined number of partly superposed peak intensities whereas the MARCH-DOLLASE function (RIETVELD-analysis) (curve 3) is related to the whole $\Theta - 2\Theta$-diagram.

The diagram shows, that the texture results between the different methods are comparable. In the case of one-axial fibre textures (Figure 4) the RIETVELD-analysis is the most suitable for an approximate texture calculation, because the other methods need a lot of more measurement and calculation time.

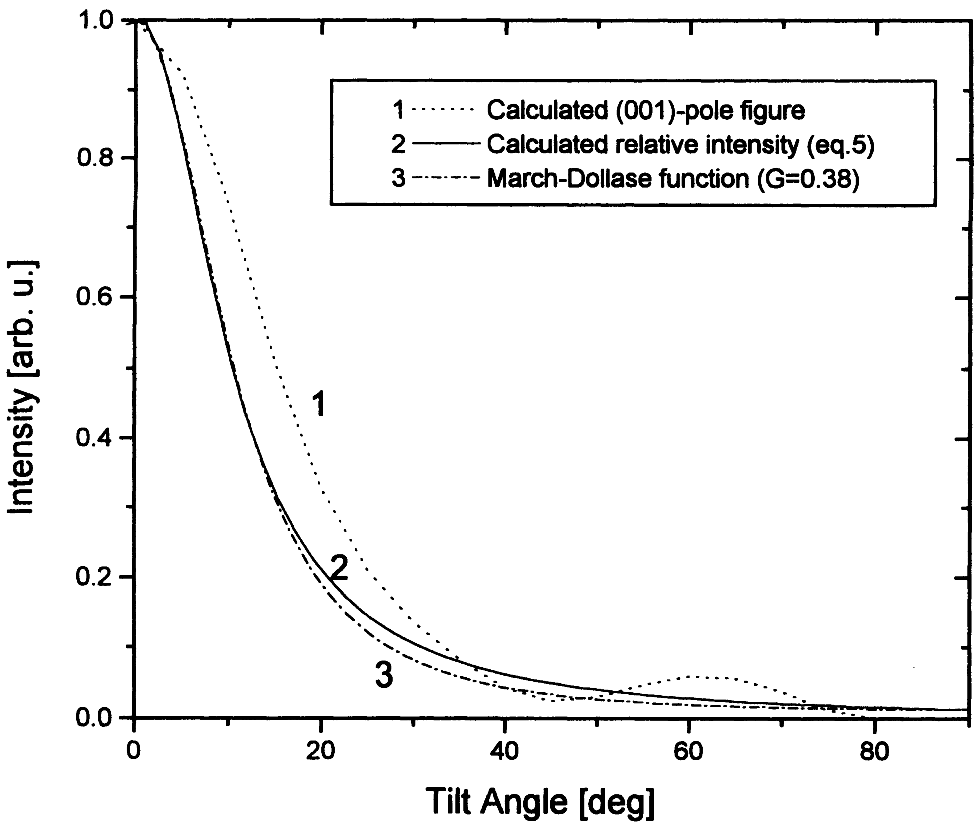

Figure 4 Comparison between three methods for the quantitative texture analysis.

Texture and Distribution in Backward Hot-Pressed NdFeB

The investigation of the texture inhomogeneities in thickness direction of NdFeB ring magnets compared with magnetic results is an application of the described method.

Figure 5 shows the calculated relative intensities of reflections of two different distances (0.41 mm, 2.64 mm) from the inner surface of a backward extruded NdFeB ring magnet. The relative intensities are plotted as a function of their angle to the (001)-plane (i.e. c-axis). The (001)-direction shows a preferred orientation. It can be noticed, that the relative intensities of the (001)-planes near to the inner surface (0.41 mm) are distinctly higher than in the outer region (2.64 mm). Therefore, the anisotropy of the inner surface is more aligned. The FWHM-values are calculated from these curves.

Figure 6 demonstrates the inhomogeneity of the texture versus the distance from the inner surface of the ring. The results show a good agreement with the above presented statements. Regions with a high degree of alignment have a small preferred orientation parameter G according to the March-Dollase definition.

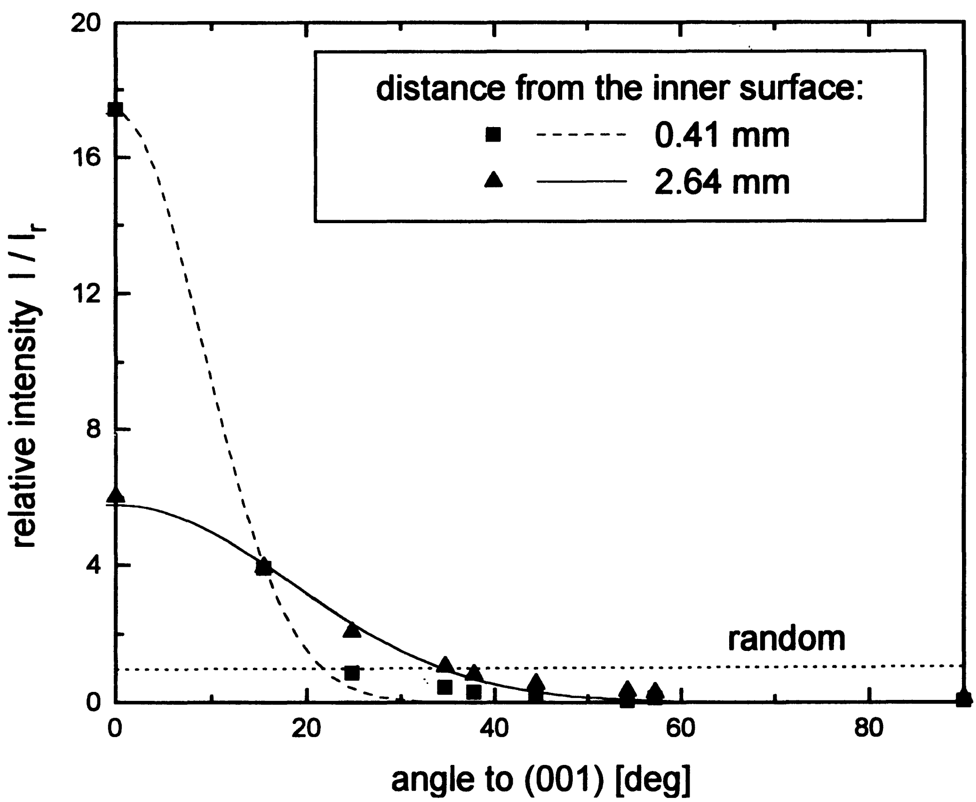

Figure 5 Calculated relative intensities (eq. 5) of all reflections vs their angle to the (001) plane at distances from the inner surface of 0.41 mm and 2.64 mm.

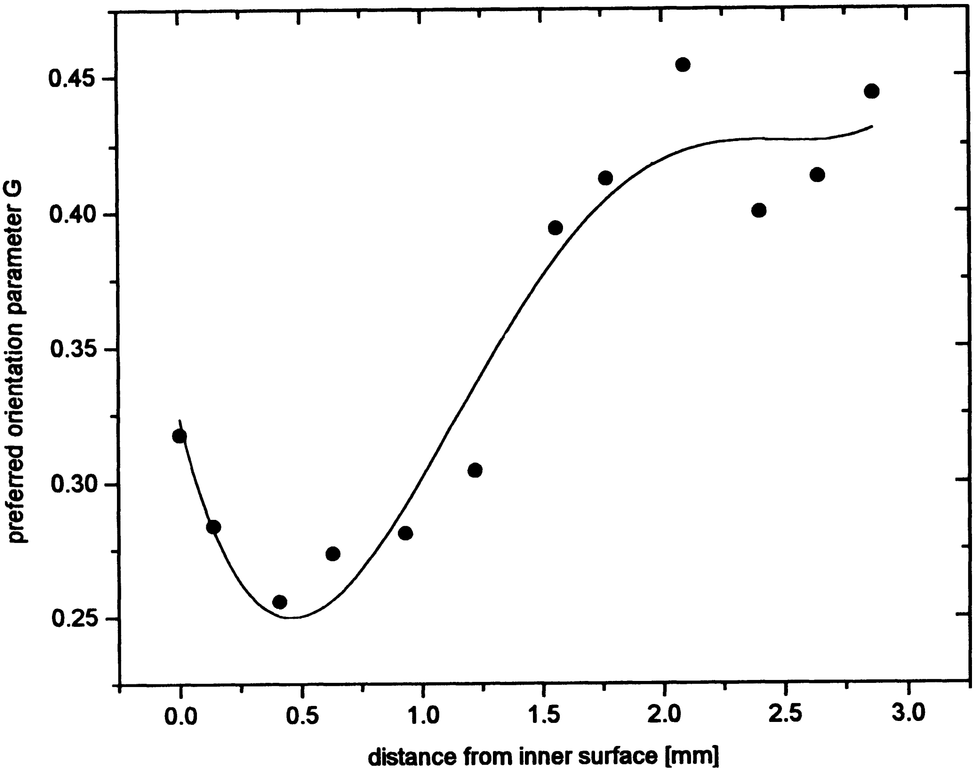

Figure 6 The preferred orientation parameter G vs distance from the inner surface.

Correlation of Texture Parameter with Magnetic Properties

After backward extruding and subsequent heat treatment the coercivity of the sample is about $\mu_0 H_{cj} = 1.1$ T. After the deformation the magnetic properties have the expected alignment of the c-axis in radial direction. Figure 7 shows the demagnetization curves of a ring magnet after annealing. In the radial direction the remanence was above 1.2 T, whereas in the axial and tangential directions the remanences were lower than 0.5 T. The agreement of the remanences in axial and tangential direction and the difference to the radial direction confirms the supposed fibre texture.

Magnetic measurements on the NdFeB sample ground by ten steps from 3.0 to 0.5 mm, beginning from the outer surface, demonstrated an enhancement of remanence from 1.24 T to 1.3 T (Figure 8). The remanence is thereby always related to the remaining sample volume.

Due to the high anisotropy of the single grains, high remanence values mean a strong grain alignment. The curve confirms the X-ray texture results. A remanence maximum near the inner surface of the ring magnet is missing, because a minimum of 0.5 mm sample thickness is necessary for a magnetic measurement. Within these regions only the quantitative local texture analysis leads to results.

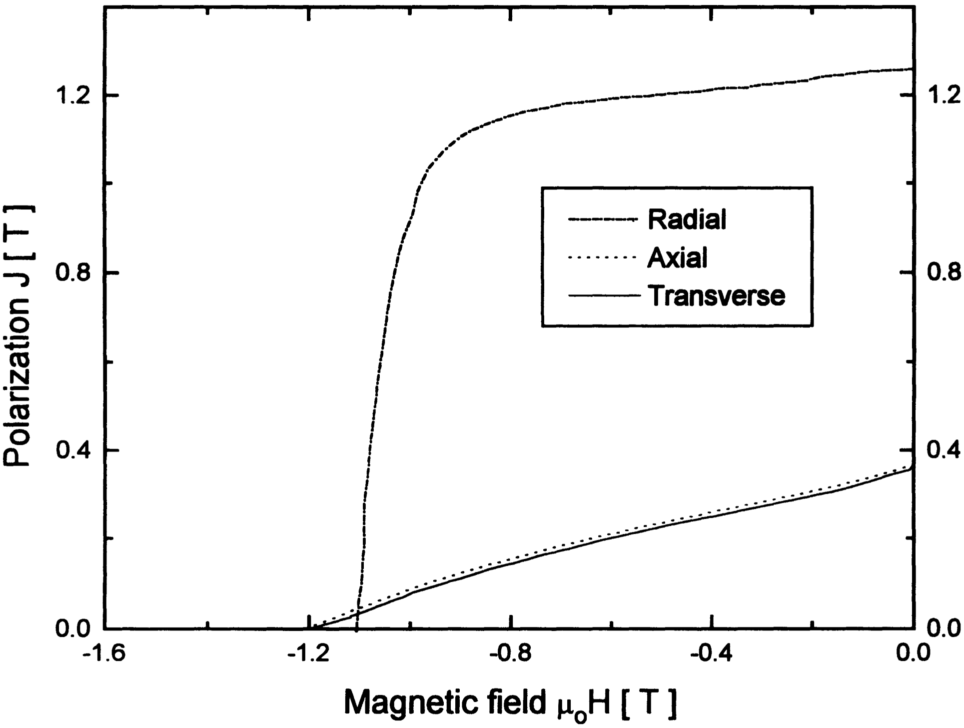

Figure 7 Demagnetization curves of a backward extruded and heat treated NdFeB magnet.

The reason for the inhomogeneity of the NdFeB ring magnet alignment is probably the inhomogeneous stress state during the deformation process due to the special tool design of the backward extrusion. These conditions result in the different material flow in the inner and outer regions of the cross-section and the different alignment of flakes and orientation of the grains within the flakes.

SUMMARY

The following statements can be made:

- Quantitative local texture analysis is possible with BRAGG-BRENTANO diffraction methods for the description of local fibre textures.
- The March-Dollase function of Rietveld analysis of high fibre-textured materials leads to meaningful results for the preferred orientation.
- Especially, for materials with strong superposed peaks the RIETVELD-analysis has an advantage to the other methods.
- The texture analysis by means of relative intensity parameters, Rietveld method, and pole figure measurements are comparable.
- The local texture methods confirm the magnetically determined inhomogeneities.

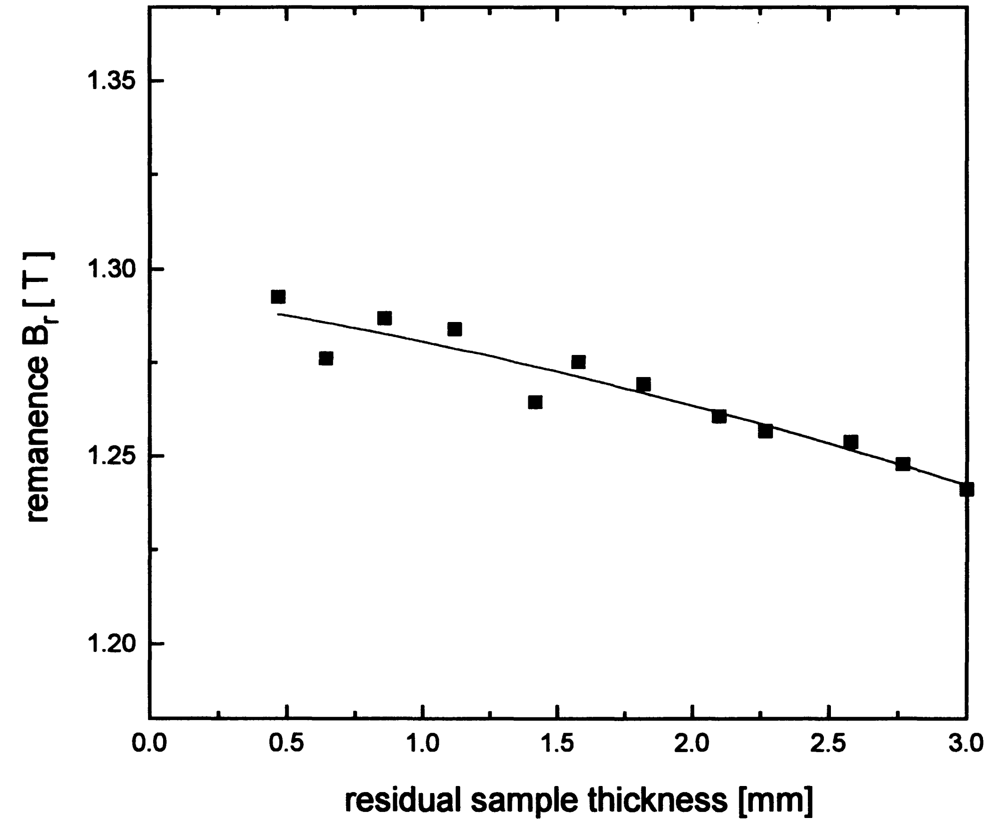

Figure 8 Remanence vs residual thickness of a ground sample.

Acknowledgements

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