Magnetic texture determination using nonpolarized neutron diffraction

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Based on neutron-diffraction data and a maximum entropy analysis, a method to determine the orientation distribution of magnetic moments in polycrystalline materials is proposed. This distribution, termed magnetic texture, was investigated in a nonoriented silicon steel sample. To obtain the magnetic contribution to the total scattering, (110) pole figures were measured in a demagnetized sample and also in a sample magnetized at various values of an external magnetic field. The proposed interpretation of experimental data allows us to obtain the resulting magnetic texture of a specimen.

INTRODUCTION

Neutrons are scattered by interaction with nuclei and with atomic magnetic moments. Due to the latter interaction, neutron diffraction has become the preferred tool for analyzing magnetic structures. The possibility of applying neutron diffraction for measurement of magnetic texture (the preferred orientations of magnetic domains) has attracted researchers already for some time. The first attempt to measure magnetic texture by Szpunar *et al.*[1] and also other research work which followed[2–4] were, however, not conclusive. The present experiment was undertaken to address this problem. A method is proposed to study the statistical distribution of the magnetic moments from the magnetic part of neutron-diffraction data. This distribution will be denoted herein as the magnetic texture.

Silicon iron is an alloy used for magnetic cores in electrical motors and generators. In this material, the magnetocrystalline energy keeps the atomic magnetic moments aligned along the easy magnetization directions. Several situations are readily considered:[4]

(a) In the demagnetized state, the magnetic moments are distributed with equal probability along the six $\langle 100\rangle$ directions. With this assumption, the magnetic texture is identical with the (100) crystallographic texture.

(b) In weak magnetic fields, the spontaneous magnetization is still aligned along the easy magnetization directions. Thus, the magnetic texture is still related to the crystallographic texture, but the symmetry of the orientation distribution of magnetic moments is no longer cubic.

(c) In high magnetic fields, the spontaneous magnetization need no longer be parallel to $\langle 100\rangle$ crystallographic directions. In this case, the magnetic texture will be related only to the sample coordinate system.

When the neutron beam is unpolarized, the diffracted intensity from the lattice plane $\{hkl\}$ is proportional to the differential scattering cross section of an atom which contains a nuclear and a magnetic part:[7]

$$d\sigma = b^2 + q^2 p^2. \tag{1}$$

In this formula, b is the coherent scattering amplitude of the atomic nuclei and p is the magnetic scattering amplitude of atoms. The factor q is the magnitude of the magnetic interaction vector, which is equal to $\sin\Psi$, where Ψ is the angle between the magnetic moment and the direction of the diffraction vector. The diffraction vector bisects the incident and diffracted neutron beams.

There are two major problems to be solved before any information about the magnetic texture can be obtained. The first problem is to separate the magnetic and nuclear scattering contributions to the scattered intensity measured as a function of specimen orientation. Such orientational scans generate pole figures, which are stereographic maps of the density of crystallographic $\langle hkl\rangle$ plane normals in the reference frame of the specimen. A pole figure is measured with a special texture goniometer.[5] The pole figure is normalized, such that

$$\int_0^{2\pi}\int_0^{\pi/2} P_{(hkl)}(\chi,\eta)\sin\chi\, d\chi\, d\eta = 2\pi, \tag{2}$$

where $P_{(hkl)}(\chi,\eta)$ is the density of the crystal plane normals (poles) versus coordinate angles (χ,η) in the reference frame of the specimen. Following normalization, the pole densities are expressed in multiples of the density that would be obtained from a random distribution of crystallite orientations.

In a diffraction experiment, neutron intensity is measured as a function of scattering angle, 2θ. The neutron wavelength is denoted as λ. The magnetic scattering amplitude p decreases rapidly with increasing values of $(\sin\theta)/\lambda$. A previous attempt was made to exploit this dependency to analyze the magnetic contribution to the total scattering.[1,2] We have also attempted to separate the magnetic contribution to the total diffracted intensity by measuring and analyzing the (100) and (220) pole figures. This method of separation however has not produced reliable results because various geometrical corrections have to be introduced and the interpretation of the data is questionable.

In order to separate the nuclear and magnetic contributions to the pole figure, we first measured the texture of the demagnetized specimen. In this state, the specimen does not display remnant magnetization in any direction. The distribution of magnetic moments however may not necessarily be random, but it is expected to follow the distribution of easy magnetic directions, $P_{(100)}(\chi,\eta)$. Thus, for the demagnetized specimen, the magnetic contribution can be estimated by calculating the average value of the orientation distribution of magnetic moments using the equation

$$D(\chi,\eta)=\frac{1}{2\pi}\int_0^{\pi/2}\int_0^{2\pi}\sin^2\Psi P_{(100)}(\chi',\eta')\sin\chi' d\chi' d\eta' \simeq \frac{2}{3}. \tag{3}$$

$D(\chi,\eta)$ is approximately constant for our sample which has a weak texture, i.e., a nearly uniform $P_{(100)}(\chi,\eta)$. In the above equation, the magnitude of q^2 is

$$\sin^2\Psi=1-[\sin\chi\ \cos(\eta-\eta')\sin\chi'+\cos\chi\ \cos\chi']^2. \tag{4}$$

However, our assumption on the magnetic texture of the demagnetized specimen is not a limitation factor for the present analysis. The magnetic texture of the demagnetized state can be always measured, even for hard magnetic materials (by demagnetizing them above the Curie temperature), and then used as a reference in the analysis of magnetic texture below the Curie temperature.

In an external magnetic field, the magnitude of the interaction vector also depends on the orientation of the magnetization with respect to the diffraction vector. As the applied field is increased, the magnetic domains are preferentially aligned and, in effect, $P_{(100)}(\chi,\eta)$ is skewed. Thus, to separate the magnetic part from the total scattering, we will use results obtained for different magnetic-field strengths.

The second problem to be solved is to relate the experimental magnetic pole figure to the underlying distribution of magnetic moment orientations. Unlike the diffraction from crystal planes, scattering from a magnetic domain yields intensity over all specimen orientations, weighted by q^2. Thus, the measured magnetic pole figure is always a broad distribution from which the underlying magnetic texture must be calculated. We propose for the restoration of the underlying magnetic texture the maximum entropy method which yields the underlying distribution of magnetic moment orientations with the least possible structure, consistent with the measured magnetic signal. This method is well described in the context of image reconstruction,[6–8] NMR spectral analysis,[9,10] and in more general works.[11,12]

A plausible magnetic texture is obtained. The described experiment demonstrates the feasibility of magnetic texture studies by neutron diffraction. In principle, the applicability of this technique is not limited to soft magnetic materials. However, there is a need for optimization of experiments depending on what materials and fields are to be used.

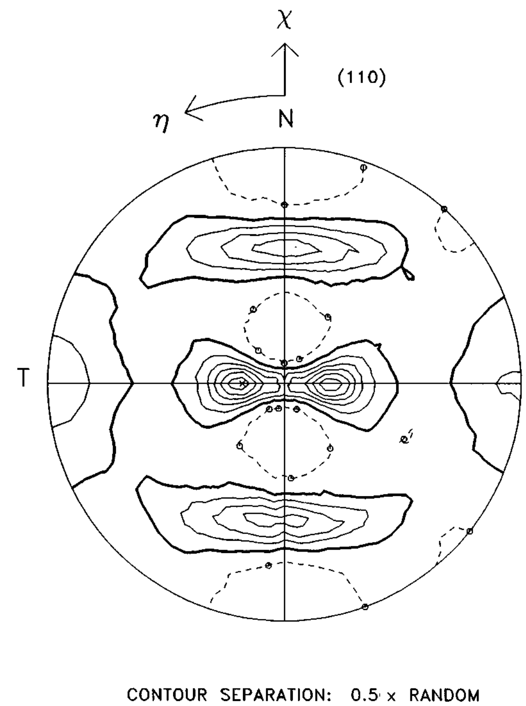

FIG. 1. Normalized (110) pole figure of the demagnetized sample.

GENERATION OF THE MAGNETIC SIGNAL

The experiments on magnetic texture using nonpolarized neutron diffraction were performed at Chalk River Nuclear Laboratory on nonoriented silicon steel. The average grain size of the material was 30 μm, so the directional dependence of neutron diffracted intensity was statistically smooth.

Strips having the dimensions of $10\times50\times0.4$ mm^3 were cut from the original rolled sheet and clamped together to form a parallelepiped with cross section area of 10 mm^2. The long dimension of the sample was parallel to the sheet rolling direction (RD). The sample was placed between the pole pieces of an electromagnet and the whole ensemble was mounted in the texture goniometer. The pole figure measurement was performed with a pseudo-equal-area scanning routine with 1020 points per pole figure. The background pole figure was measured with the diffraction angle shifted 3° from the (110) Bragg angle, 2θ.

The first (110) pole figure was measured with the sample carefully demagnetized with a varitran. We assume that this pole figure (Fig. 1) is essentially the sample crystallographic texture because the contribution of the magnetic scattering in the pole figure space is approximately isotropic. In Fig. 1, the center of the pole figure represents the sheet rolling (R) direction. The transverse (T) direction of the sample lies in the sheet plane and is perpendicular to the rolling direction. The normal (N) direction is perpendicular to the sheet plane. Directions in the sample are indicated by angles χ, the angle between the diffraction vector and rolling direction, and η, the angle between the diffraction vector and the plane defined by the normal and rolling directions. Contours connect points of equal density of the [110] plane normals. The heavy contour indicates the intensity that would be obtained from a random distribution of crystallite orientations. The thinner

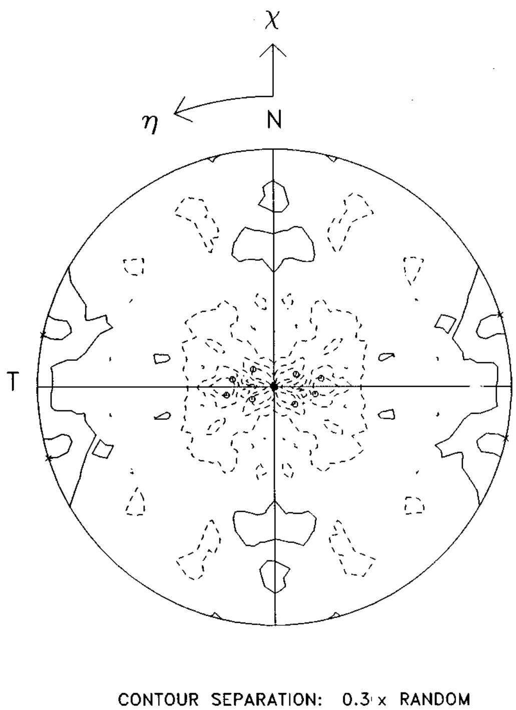

FIG. 2. Magnetic part of the total scattering for a magnetizing current of 150 mA.

continuous contours indicate intensities greater than random, in intervals shown in the caption, while the dashed contours indicate intensities less than random.

A second pole figure was measured with an electromagnet current of 150 mA, which produced an estimated internal magnetic induction of 1.1 T. The direction of the applied magnetic field was parallel to the rolling direction of the sample. All the pole figures were corrected for background (point by point subtraction), but not normalized using the random sample intensity [relation (2)].

The separation of the magnetic part from the total diffracted intensity was done by subtracting the (110) pole figure of the demagnetized sample from the (110) pole figure of the magnetized sample. The magnetic scattering is always associated with the total elastic scattering intensity. Therefore, in a textured material the pole figure representing the magnetic scattering is weighted by the pole density of the pole figure (hkl) under consideration. Thus, the theoretical pole figure $D(\chi,\eta)$ of magnetic scattering is related to the magnetic texture existing in the sample at the specific field $(H\neq 0)$ through the following equation:

$$D(\chi,\eta)=W_{(110)}(\chi,\eta)\left[\frac{1}{2\pi}\int_0^{\pi/2}\int_0^{2\pi}\sin^2\Psi M(\chi',\eta')\times\sin\chi' d\chi' d\eta'-\frac{2}{3}\right], \tag{5}$$

where $M(\chi,\eta)$ is the unknown magnetic texture and $W_{(110)}(\chi,\eta)$ is the weighting function obtained from the (110) crystallographic pole figure. The experimental magnetic scattering, which is proportional to the $D(\chi,\eta)$ pole figure, measured at a magnetizing current equal to 150 mA is plotted in Fig. 2.

The magnetic part of the total elastic scattering is a function of the reflecting plane (hkl), the neutron wavelength λ, and the magnetic scattering amplitude of atoms. Using diffraction data[13] for iron the (110) reflection and $\lambda=1.05$ Å, one can calculate that the magnetic contribution varies between extreme values of only $+6.4\%$ at $\sin\Psi=1$ and -12.7% at $\sin\Psi=0$. Because the measurable signal from the magnetic texture is a small fraction of the total intensity, long counting times are required to determine the magnetic portion of the pole figure with unpolarized neutrons.

MAGNETIC TEXTURE DETERMINATION

To obtain information about the magnetic texture, Eq. (5) has to be solved. We shall discuss the general case where the magnetic texture is described in the sample coordinate system. The sample has orthorhombic symmetry and the magnetic field is applied along the rolling direction (RD) which is one of the symmetry axes. It is reasonable to assume that the magnetic texture also has orthorhombic symmetry. Thus, the measured differences between the magnetized and demagnetized sample have been symmetrized in quadrants to produce Fig. 2.

The system of equations represented by relation (5) is therefore reduced to the first quadrant of the pole figure. In discrete representation of the pole figure using a 5° interval for both angles χ and η, the first quadrant contains $19\times 19=361$ points. A first attempt to solve Eq. (5) as a system of 361 equations with 361 unknowns $M(\chi,\eta)$ failed. The matrix of this system is nearly singular due to the slow variation of the $\sin^2\Psi$ coefficients.

For this reason, we propose a different method of solving the magnetic texture problem. In this method, the least-biased magnetic pole figure that is to be restored from experimental magnetic scattering data, has maximum entropy. The magnetic pole figure is represented as a sequence of positive values in the angular space of the sample reference frame

$$M(\chi_j,\eta_j)=M(x_j)\geqslant 0, \tag{6}$$

where the unit vector x_j denotes the direction (χ_j,η_j).

The entropy E of this set of values is defined as

$$E=-\sum_{j=1}^{J} M(x_j)\ln M(x_j). \tag{7}$$

Using the discrete representation, Eq. (5) becomes

$$D(Y_n)=\sum_{j=1}^{J} M(x_j)S(y_n,x_j)-\frac{2}{3}, \tag{8}$$

where

$$S(y_n,x_j)=\frac{\Delta\chi\Delta\eta}{2\pi}W_{hkl}(y_n)\sin^2\Psi\sin\chi_j. \tag{9}$$

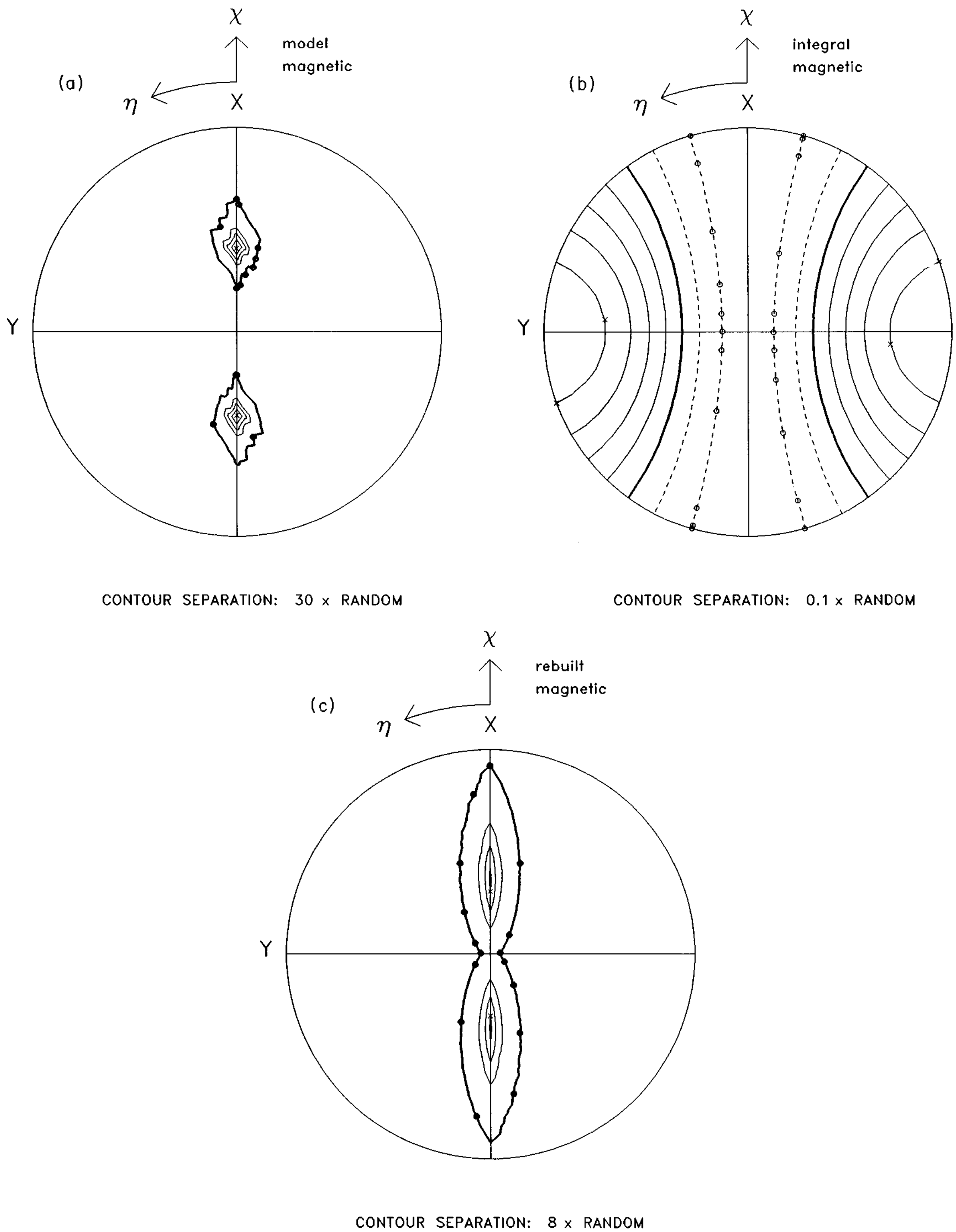

FIG. 3. Restoration by maximum entropy (c) of the magnetic scattering pattern (b), compared with the computer generated magnetic texture (a).

Following the maximum entropy formalism, we construct the variational principle for the solution set $M(\chi_j, \eta_j)$ subject to constraint equation (8):

$$-\sum_{j=1}^{J} M(x_j)\ln M(x_j) - \sum_{n=1}^{N} \lambda_n \left\{ \left[\sum_{j=1}^{J} M(x_j) S(y_n, x_j) - \frac{2}{3} \right] - D(y_n) \right\} = \max, \quad (10)$$

where λ_n are undetermined multipliers.

The magnetic texture solution is found by differentiation with respect to $M(x_j)$:

$$M(x_j) = \exp\left[-1 - \sum_{n=1}^{N} \lambda_n S(y_n, x_j) \right]. \quad (11)$$

Differentiation with respect to λ_n produces the constraint equations (8) which constitute, together with Eq. (11), the

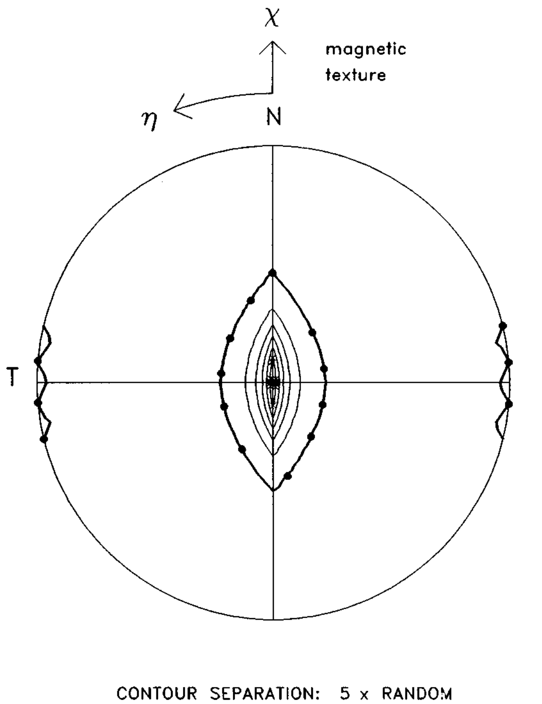

FIG. 4. Magnetic texture of the sample magnetized using a magnetizing current of 150 mA.

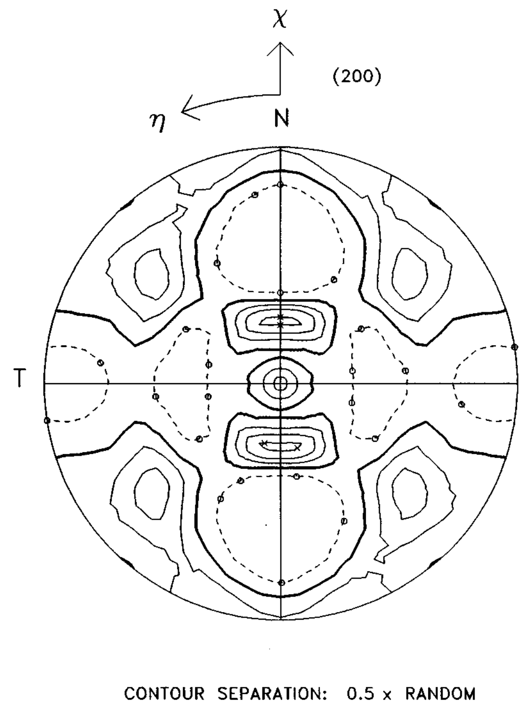

FIG. 5. Experimental (200) pole figure.

restoring formulas proposed in this paper. Since y_n and λ_n are not related to each other, Eq. (8) represents a nonlinear system of M (= number of experimental data points) equations with N (= number of undetermined multipliers) unknowns. This system was numerically solved using the Newton-Raphson method combined with the linear least-squares fitting procedure.[14]

The restoring equations (8) and (11) have been tested by computer simulation. We generated a magnetic texture, as shown in Fig. 3(a). The magnetic scattering data points have been calculated using Eq. (5) with $W_{(110)}(\chi,\eta)=1$ and they are plotted in the same stereographic projection that is used for pole figure representation in Fig. 3(b). The magnetic pole figure restored with maximum entropy as obtained using the outlined method with $N=16$, $J=360$ (step 10°) and $M=361$ is shown in Fig. 3(c). The magnetic texture is also normalized, such that

$$1=\frac{1}{2\pi}\oint M(\chi,\eta)\sin\chi d\chi d\eta. \tag{12}$$

As an example of magnetic texture calculation, the experimental data representing the magnetic scattering obtained at 150 mA magnetizing current were used to calculate the magnetic texture, as shown in Fig. 4. for comparison, the (200) pole figure, which describes the orientation distribution of the easy magnetic directions, is plotted in Fig. 5.

An interesting feature of the magnetic texture is the shape of the peak around the position of the maximum magnetic pole density which, as expected, is located in the rolling direction. However, as one can see comparing the magnetic texture with the (200) pole figure, the magnetic distribution is elongated towards the directions of maximum density of ⟨100⟩ easy directions.

SUMMARY AND CONCLUSIONS

This paper describes and demonstrates the method of magnetic texture determination by neutron diffraction. This method has the potential to become an important tool for research on magnetism of polycrystalline materials.

Information about the orientation distribution of magnetic moments in polycrystalline silicon iron in an external magnetic field can be obtained using nonpolarized neutron diffraction. The proposed method can be summarized as follows:

(1) A magnetic signal was generated using one pole figure, namely (110), measured with and without an external magnetic field. From these measurements, the magnetic scattering due to the change in orientation distribution of magnetic moments was deduced.

(2) The interpretation of experimental data was accomplished by solving the system of equations (5) using the maximum entropy method. The restoration based on this principle was illustrated by computer simulation and the use of experimental magnetic scattering data from a nonoriented silicon steel. The method revealed a distortion of the magnetic texture towards the directions of maximum density of the ⟨100⟩ poles in the material.

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