Theory of Thermal Remagnetization of Permanent Magnets

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Abstract

A self-consistent mean-field theory explaining the thermal remagnetization (TR) of polycrystalline permanent magnets is given. The influence of the environment of a grain is treated by an inclusion approximation, relating the field inside the grain to the local field outside by means of an internal demagnetization factor $n$. For the switching fields and the fluctuations of the local fields around the mean field Gaussian distributions of widths $\sigma_s$ and $\sigma_f$ resp. are assumed. The isothermal hysteresis curve, the recoil curves, and the TR in dependence on the model parameters $n$, $\sigma_s$, and $\sigma_f$ are calculated. Furthermore, the influence of the initial temperature and the strong dependence of the TR on the demagnetization factor of the sample are studied, and it is shown that for reasonable parameter sets TR effects up to 100% are possible. The theoretical results correspond well with the experimental situation.

Key words: Thermal Remagnetization, Permanent Magnets, Hysteresis, Inclusion Approximation, Switching Field Distribution

PACS: 75.50Vv, 75.50Ww, 75.60Ej

1 Introduction

It is common knowledge that increasing temperature destroys ferromagnetism, due to the related decrease of both the saturation magnetization and the coercivity. Thus an experiment, where an unmagnetized sample is heated to some hundred degrees and becomes thereby magnetized, is a challenge to our understanding of permanent magnetism. This effect was discovered in SmCo$_5$.

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about 25 years ago [1,2] and named thermal remagnetization (TR). The experimental procedure, sketched in Fig. 1, includes the saturation and isothermal dc-demagnetization via the points “1” and “2” and a subsequent heating. The largest TR-effect, i.e. more than 80 % of the saturation magnetization at the initial temperature, has been observed in well-aligned sintered SmCo$_5$ for a closed magnetic circuit [2,3]. In open circuits the TR depends strongly on the demagnetization factor $N$ of the sample [4,5]. In well texturized NeFeB magnets the TR-effect is lower, but well measurable [5,6]. In contrast to the nucleation controled SmCo$_5$- and NdFeB-magnets the effect is absent [4,7] or at least very small [8] in high coercivity Sm$_2$Co$_{17}$ sintered magnets, which are believed to be pinning controled. In spite of the good experimental situation the theoretical understanding is dissatisfying. There is agreement on the fact that the average coercivities of positively and negatively magnetized grains have to be different and that the temperature dependence of the coercivity is essential for understanding the TR. The latter was demonstrated by experiments on barium ferrite, where a sign-change of the temperature coefficient of the coercive field $H_C$ results in an “inverse” TR effect, i.e. a remagnetization upon cooling [5]. Livingston and Martin [4] argued that the TR in SmCo$_5$ is due to the change of the grains from single-domain to multi-domain state. Their mechanism can not explain TR to more than 50 %. We proposed an alternative mechanism [5], where the TR is caused by the fluctuating fields in the neighbourhood of “hard” grains. This first model aimed at a qualitative understanding of the TR solely, therefore oversimplified distributions of the switching fields and the field fluctuations were used. In a subsequent calculation [9] we introduced Gaussian distributions both for the switching fields and for the field fluctuations. Furthermore the distribution width of the latter was chosen as function of the average magnetization. This brought the resulting TR curves closer to the experimental situation, especially well below the temperature $T_{\text{max}}$, where the maximum TR occurs, but was not able to explain the vanishing TR at temperatures above. The same kind of distributions for the fluctuation fields were used by Müller et. al. [6]. They presented a mean-field theory devoted mainly to the coercivity of NdFeB, where non-magnetic phases as well as multi-domain magnetic grains are present. They also derived a formula for the maximum TR. Whereas they took the field fluctuations into account while calculating the demagnetization curve, they neglected these fluctuations in that part of their paper, which was devoted to the TR. The resulting overestimation of the TR for NeFeB they attributed to that neglect of the field fluctuations, and furthermore to the pinning of domain walls, as well as to the influence of the sample demagnetization factor. Supported by the experimental fact, that SmCo$_5$ of lower to medium coercivity in its dc-demagnetized state contains multi-domain grains [10,4,11], Lileev et al. [12,13] presented a theory, which takes into account both multi-domain grains, and field fluctuations caused by the interacting field of neighbouring grains. Since they included the mutual dipol-dipol interaction of the grains they had to carry out numerical simulations which makes it difficult to handle
their theory, especially if one is interested in the influence of basic parameters e.g. the distribution width of the field fluctuations, the influence of the initial temperature etc.. They calculated a TR curve which has a rather sharp peak. Whereas the magnitude of the maximum effect is comparable to the experiment, the shape of the theoretical curve differs from the experimental situation. Nevertheless, we share the opinion that the influence of the nearest neighbourhood of a grain is an essential for every elaborated theory of the TR. For this task the current paper extends our former theory [9] by an inclusion approximation. Furthermore we take into account the possibility that the grains may nucleate into multi-domain states. The paper is organized as follows: In the next section we present the model. Since a theory of the TR presumes a calculation of both the isothermal hysteresis loops and the recoil curves from every point of the hysteresis loop, we present the related theory and the results in subsection 3.1 and 3.2 respectively. The calculation of the TR due to heating is the topic of subsection 3.3. In the last section we discuss the results.

2 The model

The polycrystalline permanent magnet is an ensemble of high-coercive magnetic uniaxial grains. To fix our model we have to define the properties of both a single grain and the ensemble.

2.1 Properties of a single grain

Every grain is characterized by

(A1) $M_S(T)$, the temperature dependent saturation magnetization,
(A2) $H_s(T)$, the temperature dependent switching field. This is the absolute value of the field needed for reversing the magnetization of the grain in a closed circuit.
(A3) $n$, the “internal” demagnetization factor, which is determined by the shape of the grain,
(A4) $\vec{c}$, the direction of the easy axis due to the high uniaxial anisotropy
(A5) $V$, the volume of the grain.

2.2 Properties of the ensemble

Regarding the ensemble we assume
all grains exhibit the same $M_s(T)$, 
the easy axis $\vec{c}$ of the grains are completely aligned (ideal texture), 
the grains differ in their switching fields $H_s$, but the distribution of these 
switching fields should be known. For simplicity we assume a Gaussian 
normalized with respect to the region $0 < H_s < \infty$,

$$g(H_s) = \frac{2}{\sqrt{\pi}\sigma_s}\left(1 + \text{erf}\left(\frac{H_s - \bar{H}_s}{\sigma_s}\right)\right)^2,$$  
\text{with the “mean switching field” $\bar{H}_s$ and the distribution width $\sigma_s$.}

It is problematic to get direct information both on the temperature dependence of the switching fields and their distribution. What can be measured is the average magnetization in dependence on the external magnetic field, the temperature and the time. The switching field distribution can not be measured directly. In non-interacting ensembles the switching field distribution may be determined from remanence measurements [14], but if the grains interact strongly this method fails. From the Henkel plots measured for Nd-FeB [15,16] we know that most sintered hard magnets are strongly interacting ensembles. Thus, for the moment, we have to regard $\bar{H}_s$ and $\sigma_s$ as model parameters. Fortunately we will see later on that our theory delivers a possibility to relate $\bar{H}_s$ to the coercivity $H_C$ of the sample.

2.3 Inclusion approximation

Since we believe that the TR is caused mainly due to grain interactions we want to explain the approximations in detail. On a macroscopic length scale the internal magnetic field and the magnetization of the sample are homogeneous. But, if the length scale is reduced to the order of some grain diameters the magnetization becomes coarser and the inhomogeneities gain influence. Averaging over volume elements containing a few grains only, yields values, which deviate from the average. Thus, we consider every grain as an inclusion embedded in a local environment, which may differ stochastically in its magnetic field and magnetization from the related averaged values. We show this schematically in Fig. 2. For simplicity we approximate the magnetization in the vicinity of the inclusion by the mean magnetization $\langle M \rangle$, neglecting the mentioned fluctuations. Otherwise we allow for field fluctuations $\Delta H = H - \langle H \rangle$ around the mean internal field in the environment of a grain. The mean internal field is related to the external applied field $H_{ext}$ according to

$$\mu_0\langle H \rangle = \mu_0 H_{ext} - N\langle M \rangle,$$
with $N$ being the demagnetization factor of the sample. The field fluctuations are characterized by the following assumptions

(C1) The local magnetic fields (in $\vec{c}$-direction) are Gaussian distributed

$$f(H) = \frac{1}{\sqrt{\pi\hat{\sigma}_f}} e^{-\left(\frac{\Delta H}{\hat{\sigma}_f}\right)^2} \quad \text{with} \quad \Delta H = H - \langle H \rangle.$$ \hspace{1cm} (3)

(C2) The distribution width $\hat{\sigma}_f$ is itself a function of the mean magnetization. We set

$$\hat{\sigma}_f = \sigma_f \left(1 - \frac{\langle M \rangle^2}{M_S^2}\right).$$ \hspace{1cm} (4)

Assumption C1 is a consequence of the central limit theorem of probability theory. Of course the local fields fluctuate around the mean internal field also in direction, thus, strictly speaking eq. (3) accounts for the fluctuations of the $z$-component only. Furthermore the deviation of the field direction from the easy axis gives rise to rotation processes, but this is surely a small effect in ideally aligned magnets with high anisotropy constants. Assumption C2 attempts to take into account that in an ideally textured magnet the fluctuation width is zero in the saturated state and maximum in the dc-demagnetized state. If $\hat{\sigma}_f(\langle M \rangle)$ is expanded to second order and the expansion parameters are fixed by these requirements together with the demand that $\hat{\sigma}_f(\langle M \rangle)$ has to be a symmetric function one gets eq. (4).

3 Field- and temperature-dependent magnetization

3.1 Calculation of the isothermal demagnetization curve

After the saturation all grains are up ($\uparrow$) magnetized. They posses their maximum switching fields and $\langle M \rangle = M_S$ holds, due to assumption B2. If the external field is lowered to $H_{ext}^{(1)}$ (point “1” in Fig. 1) the magnetization of a grain is switched down ($\downarrow$) if its internal field $H_{i,\uparrow}^{(1)}$ is lower than $-H_s$ and the internal field after switching $H_{i,\downarrow}^{(1)}$ will be smaller than $+H_s$. Thus we have the two conditions

$$H_{i,\uparrow}^{(1)} < -H_s \quad \text{("switching condition")},$$ \hspace{1cm} (5)

$$H_{i,\downarrow}^{(1)} < +H_s \quad \text{("not-back-switching condition")}.$$ \hspace{1cm} (6)

The internal fields before and after switching are with respect to the above introduced approximations:
Here $H$ is the local field in the environment of the grain. A variation of the external field $H_{\text{ext}}$ results in a change of $\langle M \rangle$ on the one hand and on the other hand it changes the probability $f(H)dH$ that the field in the environment of a grain is between $H$ and $H + dH$ due to the relation (2) and eq. (3). If the switching condition (5) is fulfilled but the condition (6) is not fulfilled, i.e.

\[
\mu_0 H^{(1)}_{i,\uparrow} = \mu_0 H - n(M_s - \langle M \rangle_1) \quad ,
\]

\[
\mu_0 H^{(1)}_{i,\downarrow} = \mu_0 H - n(-M_s - \langle M \rangle_1) \quad .
\]

holds, the grain cannot jump into a stable state. If the grain is large enough it solves the conflict by an incomplete jump, what turns its single-domain state (SDS) into a multi-domain-state (MDS). We will call such grains “weak”. Whereas the “hard” grains can exist in states with $\pm M_s$ only, the magnetization of a weak grain $\langle M \rangle_i$ is an average over the upwards and downwards magnetized volume fractions within the grain. From phase theory of 180°-domains we get for the averaged magnetization of the $i$th grain in dependence on the local field in the environment (cf. Appendix A)

\[
\langle M \rangle_i = \langle M \rangle_+ \frac{\mu_0 H}{n} .
\]

Opposite to the hard grains the weak grains have no memory since the magnetization follows the local field immediately as long as $|\langle M \rangle_i| < M_s$ holds. Otherwise they are saturated up- or downwards. If one calculates the probability that a grain with a given $H_s$ is in a MDS the switching condition (5) limits the integration over the local field distribution from above, whereas the back-switching condition (9) limits from below. The related probability is

\[
p_w(H_s) = \int_{H_l}^{H_H} dH f(H)
\]

The two limiting fields are

\[
\mu_0 H_H = -\mu_0 H_s + nM_s - n\langle M \rangle \quad ,
\]

\[
\mu_0 H_L = \mu_0 H_s - nM_s - n\langle M \rangle \quad .
\]

The requirement that the upper limit has to be greater then the lower one yields the condition

\[
\mu_0 H_s < nM_s \quad .
\]
Thus, grains with switching fields smaller than their own internal demagnetizing field are weak. The magnetization of the volume fraction of the weak grains with a given $H_s$ is

$$\langle M \rangle_w(H_s) = M_S \int_{H_H}^{H_L} dH f(H) - M_S \int_{-\infty}^{H_L} dH f(H) + \int_{H_L}^{H_H} dH f(H) \langle M \rangle_i. \quad (15)$$

The integral may be evaluated yielding

$$\langle M \rangle_w(H_s) = -\frac{M_S}{2} (\text{erf}(y_H) + \text{erf}(y_L))$$

$$+ \frac{1}{2} \left( \frac{\mu_0 H^{(1)}_{\text{ext}}}{n S} + \langle M \rangle_1 (1 - \frac{N}{n}) \right) (\text{erf}(y_H) - \text{erf}(y_L))$$

$$- \frac{\mu_0 \sigma f}{2\pi n} \left( e^{-y_H^2} - e^{-y_L^2} \right)$$

with

$$y_H = -\frac{\mu_0 H_s + nM_S - n\langle M \rangle_1 - \mu_0 H^{(1)}_{\text{ext}} + N\langle M \rangle_1}{\mu_0 \sigma f (1 - \langle M \rangle_1^2 / M_S^2)} \quad (17)$$

$$y_L = \frac{\mu_0 H_s - nM_S - n\langle M \rangle_1 - \mu_0 H^{(1)}_{\text{ext}} + N\langle M \rangle_1}{\mu_0 \sigma f (1 - \langle M \rangle_1^2 / M_S^2)}. \quad (18)$$

The index 1 at $H^{(1)}_{\text{ext}}$ and $\langle M \rangle_1$ indicates that the calculation is for point 1 in Fig. 1.

For hard grains $H_L$ is higher than $H_H$. If these grains fulfill the switching condition (5), condition (6) will be fulfilled automatically. The probability to find a grain downwards magnetized if a field $H^{(1)}_{\text{ext}}$ is applied is

$$p_1(H_s) = \int_{-\infty}^{+\infty} dH f^{(1)}(H) \Theta(-H_s - H^{(1)}_{i,\uparrow}) \Theta(H_s - H^{(1)}_{i,\downarrow}). \quad (19)$$

with $H^{(1)}_{i,\uparrow}$ and $H^{(1)}_{i,\downarrow}$ being the fields within the grain in the upwards and downwards magnetized state resp. and $\Theta(x)$ is the Heaviside function. $f^{(1)}$ is the field distribution eq. (3) with $\mu_0 \langle H \rangle = \mu_0 H^{(1)}_{\text{ext}} - N\langle M \rangle_1$. The integration in eq. (19) is easily done yielding

$$p_1(H_s) = \frac{1}{2} (1 + \text{erf}(x_H)) \quad (20)$$
with

\[ x_H = -\mu_0 H_s + nM_s - n\langle M \rangle_1 - \mu_0 H_{\text{ext}}^{(1)} + N\langle M \rangle_1 \]

\[ \mu_0\sigma_f (1 - \langle M \rangle_1^2/M_s^2) \]

For the magnetization of such a fraction of hard grains with a given \( H_s \) we find

\[ \langle M \rangle_1(H_s) = M_S (1 - 2p_1(H_s)) = -M_S \text{erf}(x_H) \]  

(21)

The total magnetization of the sample one gets from averaging with respect to \( H_s \)

\[ \langle M \rangle_1 = \int_0^{nM_s/\mu_0} dH_s g(H_s) \langle M \rangle_{\text{w},1}(H_s) + \int_{nM_s/\mu_0}^{\infty} dH_s g(H_s) \langle M \rangle_1(H_s) \]  

(22)

Here the integration has been splitted due to the different contributions of the weak and hard grains. For the weak grains we have to use eq. (17) with \( H_{\text{ext}}^{(1)} \) being \( H_s \) and \( \langle M \rangle \) being \( \langle M \rangle_1 \). Since the right hand side of eq. (22) depends on the mean magnetization \( \langle M \rangle_1 \) itself, we have to solve this implicit equation numerically. This is done by fixing \( \langle M \rangle_1/M_S \) and searching the related \( H_{\text{ext}}^{(1)} \).

Fortunately there is always exactly one solution for \(-1 < \langle M \rangle_1/M_S < 1\).

The aim of the present paper is to clarify, how the TR depends on the model parameters \( \sigma_s, \sigma_f \) as well as on the internal demagnetization factor \( n \). We vary these parameters around the values \( M_S = 1T, \sigma_s = 1.5T/\mu_0, \sigma_f = 0.5T/\mu_0, n = 0.333, \) and a mean value of the switching field distribution \( \mu_0 H_s = 3.0T \).

The values of \( M_S, H_s, \) and \( \sigma_s \) resemble SmCo (VACOMAX 200), where we measured at T=300K e.g. \( M_S = 0.972T, \mu_0 H_C = 2.87T, \sigma_s \) is a rough estimate of the width of the differentiated experimental demagnetization curve. The results for the calculated demagnetization curve are plotted in Fig. 3, where a spherical sample was assumed \((N = 1/3)\).

### 3.2 Calculation of the isothermal recoil curve

Next, as shown in Fig. 1 (Point “1” to point “2”), the external field is changed from \( H_{\text{ext}}^{(1)} \) to \( H_{\text{ext}}^{(2)} \) with \( H_{\text{ext}}^{(2)} > H_{\text{ext}}^{(1)} \) via the recoil curve. Some of the hard “down”-grains are switched back, whereas the weak grains shift their magnetization reversible. The related mean magnetization is \( \langle M \rangle_2 \). The contribution of the weak grains results from eq. (17), if we insert \( H_{\text{ext}}^{(2)} \) for \( H_{\text{ext}} \) and \( \langle M \rangle_2 \) for \( \langle M \rangle \) resp. followed by an integration from zero to \( nM_s/\mu_0 \) with respect to \( H_s \). The contribution of the hard grains is more difficult to calculate, due to the memory effect. Let us consider the probability \( w_{\downarrow \uparrow}(H_s) \) that a hard grain with a given \( H_s \) was switched by \( H_{\text{ext}}^{(1)} \) and switched back by \( H_{\text{ext}}^{(2)} \) afterwards.
To calculate this probability it is inevitable to make an assumption on the correlation between the fluctuation of the local field $H_1$ which acts on a grain if $H_{ext}^{(1)}$ is applied and the fluctuation of $H_2$ according to the external field $H_{ext}^{(2)}$. Of course, if $H_{ext}^{(2)}$ is only slightly different from $H_{ext}^{(1)}$ it is unlikely that the neighbourhood of a grain changes considerably, so that $H_1$ and $H_2$ should be strongly correlated. With increasing distance between $H_{ext}^{(1)}$ and $H_{ext}^{(2)}$ this correlation will vanish, due to the multitude of switching processes. The reasoning is as follows: The thermodynamical potential has surely a lot of nearly degenerated local minima. Applying $H_{ext}^{(1)}$ selects one of them. A small change of the applied field is not enough to overcome the barrier between adjacent minima, but larger changes generate energy surfaces, which are completely different in their topological structure, thus making states accessible, which may be very different from the state at $H_{ext}^{(1)}$. The change to such a state will be accompanied by a lot of correlated switchings. Even if the mean magnetization is changed only slightly, the neighbourhood of an individual grain may be completely different, hence we can average with respect to $H_1$ and $H_2$ independently, if the difference $H_{ext}^{(2)} - H_{ext}^{(1)}$ is not to small. Thus the probability $w_{↓↑}$ decouples accordingly:

$$w_{↓↑}(H_s) = p_1(H_s)q_2(H_s)$$  \hspace{1cm} (23)$$

with $p_1(H_s)$ from eq. (21) and

$$q_2(H_s) = \int_{-\infty}^{\infty} dH f_{(2)}(H) \Theta(H_{i,↓}^{(2)} - H_s) \Theta(H_{i,↑}^{(2)} + H_s)$$

$$= \frac{1}{2} \left( 1 - \text{erf}(y_L) \right)$$  \hspace{1cm} (24)$$

with

$$y_L = \frac{\mu_0 H_s - nM_s - n\langle M \rangle_2 - \mu_0 H_{ext}^{(2)} + N\langle M \rangle_2}{\mu_0 \sigma f \left( 1 - \langle M \rangle_2^2 / M_s^2 \right)}.$$  \hspace{1cm} (25)$$

Again the switching conditions are included by help of related Heaviside functions. Due to the mentioned magnetization changes in the environment of a grain it may happen that grains which resisted $H_{ext}^{(1)}$ are switched down by $H_{ext}^{(2)}$ (“up”-“down”-contribution). The related probability factorizes also

$$w_{↑↓}(H_s) = \left( 1 - p_1(H_s) \right) p_2(H_s)$$  \hspace{1cm} (26)$$

with $p_1(H_s)$ again from eq. (21) and
\[
p_2(H_s) = \int_{-\infty}^{\infty} dH f_{(2)}(H) \Theta(-H_s - H_{c2}^{(2)}) \Theta(H_s - H_{c1}^{(2)})
\]

\[
= \frac{1}{2} \left( 1 + \text{erf}(z_H) \right)
\]

with

\[
z_H = \frac{-\mu_0 H_s + nM_s - n\langle M \rangle_2 - \mu_0 H_{ext}^{(2)} + N\langle M \rangle_2}{\mu_0 \sigma_f \left( 1 - (\langle M \rangle_2^2/M_s^2) \right)}.
\]

There are two further probabilities corresponding to the remaining two histories, which a grain may experience, i.e. the probability that it resisted both \(H_{ext}^{(1)}\) and \(H_{ext}^{(2)}\) ("up"-"up" contribution) and the probability that a grain was switched by \(H_{ext}^{(1)}\) but resisted back-switching by \(H_{ext}^{(2)}\) ("down"-"down" contribution). We find

\[
w_{\uparrow\uparrow}(H_s) = \left( 1 - p_1(H_s) \right) \left( 1 - p_2(H_s) \right)
\]

\[
w_{\downarrow\downarrow}(H_s) = p_1(H_s) \left( 1 - q_2(H_s) \right)
\]

Thus hard grains with a given \(H_s\) contribute to the magnetization

\[
M_2(H_s) = M_s \left( w_{\uparrow\uparrow}(H_s) - w_{\uparrow\downarrow}(H_s) + w_{\downarrow\uparrow}(H_s) - w_{\downarrow\downarrow}(H_s) \right)
\]

Averaging with respect to \(H_s\) yields the recoil curve for the mean magnetization \(\langle M \rangle_2\) in state 2 in dependence on both magnetic fields \(H_{ext}^{(1)}\) and \(H_{ext}^{(2)}\):

\[
\langle M \rangle_2 = \frac{nM_s/\mu_0}{nM_s/\mu_0} \int dH_s g(H_s) \langle M \rangle_{w_2}(H_s) + \int_{nM_s/\mu_0}^{\infty} dH_s g(H_s) \langle M \rangle_2(H_s)
\]

In Fig. 3 we show additional to the recoil curve a set of minor loops starting from different values of \(\langle M \rangle_1\).

3.3 Calculation of the thermal remagnetization

In the preceding sections we calculated the demagnetization and the recoil curves with the implicit understanding that the magnetization changes isothermally at a temperature \(T_0\). The actual TR occurs if a dc-demagnetized RE-TM-magnet is heated while \(H_{ext}^{(2)}\) is kept constant (cf. Fig. 1 from point "2" to point "T"). Raising the temperature has usually two effects. On the one hand
the saturation magnetization decreases with increasing temperature and on the other hand the switching fields \( H_s \) are changed. Whereas the temperature dependence of the saturation magnetization can be measured easily, it is impossible to measure it for the switching fields. What can be measured directly is the temperature dependence of the coercive field. In appendix B we show that in our model \( \bar{H}_s \) is related to \( H_C \) and \( M_S \) by the simple formula

\[
\mu_0 \bar{H}_s(T) = \mu_0 H_C(T) + n M_S(T)
\]  

(34)

if the influence of the weak grains is negligible. Otherwise we have to solve eq. (22) for \( \bar{H}_s \) with \( H_c^{(1)} = H_C \) and \( \langle M \rangle_1 = 0 \). \( H_C(T) \) and \( M_S(T) \) are taken from measurements [17]. Due to the lack of better information we adopt the temperature dependence of \( \bar{H}_s \) for arbitrary values of the switching fields

\[
H_s(T) = H_s(T_0) \frac{\bar{H}_s(T)}{\bar{H}_s(T_0)}
\]  

(35)

The calculation of the magnetization \( \langle M \rangle_2^T \) at an enhanced temperature \( T \) is similar to the calculation of the magnetization \( \langle M \rangle_2 \) by help of eq. (33), whereby the deformation of the switching field distribution has to be regarded. That should be done by indicating the changed parameters with an index \( T \), whereas the related parameters at the temperature \( T_0 \) will not be marked. From eq. (35) and the normalization of the switching field distribution follows that \( \sigma_s \) has the same temperature dependence like \( H_s(T) \). The number of weak grains increases with temperature, since \( H_s(T) \) is usually much more reduced than \( n M_S(T) \) if the temperature is raised. The magnetization of a weak grain with given \( H_s \) at the temperature \( T \) results from eq. (17) with the appropriate \( H_s(T), M_S(T) \), and \( \langle M \rangle_2^T \) inserted. For the hard-grain contribution we have to calculate the probabilities according to eqs. (26,23,30,31), however for the elevated temperature. We find for the magnetization of the hard-grain fraction with given \( H_s \)

\[
M_2^T(H_s) = M_S^T \left( w_{↑↑}^T(H_s) - w_{↑↓}^T(H_s) + w_{↑↑}^T(H_s) - w_{↑↓}^T(H_s) \right)
\]  

(36)

It is obvious that the probability \( p_1(H_s) \) remains unchanged, since the grains were switched downwards at the initial temperature \( T_0 \). Therefore we have

\[
w_{↑↑}^T(H_s) = (1 - p_1(H_s))^2
\]  

(37)

\[
w_{↑↓}^T(H_s) = (1 - p_1(H_s)) p_2^T(H_s)
\]  

(38)

\[
w_{↑↑}^T(H_s) = p_1(H_s) q_2^T(H_s)
\]  

(39)

\[
w_{↓↓}^T(H_s) = p_1(H_s) \left( 1 - q_2^T(H_s) \right)
\]  

(40)
The probabilities

\[ q_2^T(H_s) = \frac{1}{2} \left( 1 - \text{erf}(y_L^T) \right) \] (41)

and

\[ p_2^T(H_s) = \frac{1}{2} \left( 1 + \text{erf}(z_H^T) \right) \] (42)

become temperature dependent both directly due to \( H_s(T) \), \( M_s(T) \), and \( \sigma_f(T) \) and indirectly via \( \langle M \rangle_T^2 \), since we have now

\[ y_L^T = \frac{\mu_0 H_s^T - nM_S^T - n\langle M \rangle_T^2 - \mu_0 H_{ext}^{(2)} + N\langle M \rangle_T^2}{\mu_0 \sigma_f^T \left( 1 - (\langle M \rangle_T^2 / M_S^2)^2 \right)} \] (43)

and

\[ z_H^T = \frac{-\mu_0 H_s^T + nM_S^T - n\langle M \rangle_T^2 - \mu_0 H_{ext}^{(2)} + N\langle M \rangle_T^2}{\mu_0 \sigma_f^T \left( 1 - (\langle M \rangle_T^2 / M_S^2)^2 \right)} \] (44)

The temperature dependence of \( \sigma_f^T = \sigma_f(T) \) was assumed to be that of \( M_s \), since the fluctuations are caused by the inhomogeneities of the magnetization. From the numerical solution of the implicit self-consistent equation we calculated the TR in dependence on the parameters for the above mentioned parameter set. Here the starting temperature was fixed to 300 K and we assumed a spherical sample. The influence of the switching field distribution width is studied in Fig. 4 A. To get noticeable TR-effects the distribution width \( \sigma_s \) should not be too small. The dependence of the TR on the fluctuation width \( \sigma_f \) is shown in Fig. 4 B. The TR peak becomes broader and shifts to lower temperatures with increasing field fluctuations. The strong dependence of the TR on the internal demagnetization factor is depicted in Fig. 4 C. For small demagnetization factors, i.e. for elongated grains, the TR is small, whereas effects up to 100 % are possible for platelet-shaped grains. Up to now there are no experimental results available regarding the influence of the internal demagnetization factor \( n \) onto the TR. Regarding the external demagnetization factor \( N \) it is known that in closed-circuit measurements TR-effects up to 100% are possible [2] (related to \( M_S(T_{max}) \) !) in the case of SmCo\(_5\) sintered magnets. For samples measured in a VSM, where the demagnetization factor is between 0.1 and 0.6, a significant lowering of the TR with increasing demagnetization factor \( N \) is reported. This is exactly what the calculated curves in Fig. 5 demonstrate. Another feature, which has been studied experimentally, is the dependence of the TR on the initial temperature \( T_0 \). The maximum
4 Discussion

The above presented theory explains the observed features of the TR-experiments well, whereby we focused especially to the shift of the temperature $T_{\text{max}}$, where the maximum TR occurs, as well as to the height of the maximum itself. Especially the strong dependence on the external demagnetization factor and on the starting temperature can be explained. Furthermore the theory is able to explain TR effects of more than 50%. To proceed to the description of the TR it was inevitable to develop a theory for the hysteresis and the recoil curves. Whereas there exist a multitude of micromagnetic calculations trying to explain the coercivity by assuming special microstructures which give rise to either pinning of domain walls or nucleation, our theory starts a little above this level, since we do not judge about the reason for a switching field, but simply accept its existence. In this point our theory resembles the Preisach [18] model, which was created to take into account the interaction between different grains (bistable magnetic units). The main disadvantage of the Preisach model is due to the fact that the Preisach distribution function is not dependent on the magnetization state [18,19]. It is easy to prove that a theory of the TR based on this model is not able to explain TR effects of more than 50%. The reason is the lack of feedback. Nevertheless, we share the opinion that the difference between the local fields and the mean field, which is due to grain interactions, is the crucial point. In this work the width of these fluctuations is given by $\sigma_f$. Our results demonstrate that without local field fluctuations there will be no TR effect above 50%, e.g. in a normal mean-field theory. To take into account the feedback of the neighbourhood of a grain the inclusion approximation, albeit at a very rough level, was introduced. This provides us with a new parameter $n$, i.e. the internal demagnetization factor. Assigning the same internal demagnetization factor $n$ to all grains seems to be a severe simplification, but since we have no reliable information on the switching field distribution, we probably account, at least partly, for different grain shapes and/or exchange coupling via the grain boundaries by adjusting the switching field distribution. Thus, $n$ is more a feedback parameter than a real demagnetization factor. The most difficult problem is the determination of the temperature dependence of the switching fields. All the former theories used the temperature dependence of the coercivity for this task, due to the lack of better information. But it is evident that this forces the TR magnitude to drop to zero at the temperature $T_{H_C=0}$ where the coercivity vanishes. Contrary, investigations on SmCo$_5$ demonstrated that the TR manifestly remains
above $T_{H_C=0}$ \cite{17}. This is due to the fact that for this compound the difference between $T_{H_C=0}$ and the Curie temperature $T_C$ is about 250 K \cite{20}. Since our theory relates the mean switching field to the coercivity, it was possible to extract the temperature dependence $\bar{H}_s(T)$ from the measured $H_C(T)$ curves. The derived formula is in congruence with the empirical formula used in \cite{21,22} if one relates $cH_A (c \ll 1)$ to the switching field and $n$ to the corresponding effective demagnetization factor $N_{eff}$. This holds as long as the mean switching field is greater than the internal demagnetizing field. Otherwise the simple rule given in eq. (34) fails. Another interesting point is whether the weak grains are essential for the description of the TR. The calculated relative volume fraction of weak grains in dependence on the temperature is depicted in a dashed fashion in Fig. 8. At $T_{max}$ the $\bar{H}_s$ equals roughly $nM_S/\mu_0$, i.e. the maximum TR is observed shortly before the majority of the grains become weak. At temperatures higher than $T_{max}$ the magnetization decreases. Sm-Co-magnets undergo irreversible changes in their microstructure at higher temperatures. That is why measurements above $T_{max}$ are rather difficult and, therefore, seldom reported. For the parameter set used in Fig. 7 the influence of weak grains is small as long as the temperature is below $T_{max}$. For $T > T_{max}$ the neglect of the weak grains would result in a qualitative different behaviour, since we get a sign change of the TR before it finally vanishes, as shown in Fig. 7 in dotted manner. The reason for that peculiarity is made clear in Fig. 8, where we have plotted the temperature dependence of the probabilities $w_{\uparrow\uparrow}$, $w_{\uparrow\downarrow}$, $w_{\downarrow\uparrow}$, and $w_{\downarrow\downarrow}$ regarding the hard grain fraction. It is obvious that at first $w_{\uparrow\downarrow}$ increases and when it drops down $w_{\uparrow\downarrow}$ rises up, thus explaining the sign change. This behaviour is not observed if the probability $p_2$, i.e. that a grain which resisted $H^{(1)}_{ext}$ will be switched by $H^{(2)}_{ext}$, is neglected, as it was done in former theories, but then an overestimation of the TR occurs, as visible in Fig. 7. For the description of the TR at higher temperature it is necessary to take into account, that above $T_{max}$ even the switching fields of the hardest grains are lowered enough to be switched by the still existing stray fields. The latter do not vanish until the magnetization breaks down. It is worth to point out, that weak grains are not necessary to get effects of more than 50 %. Such a large TR one gets due to the feedback of the magnetization via the internal demagnetizing fields. In some sense this models the well known avalanche effects observed during hysteresis measurement, which should also occur to a certain extent if the sample remagnetizes. In consideration of the various approximations one can not expect that all magnetic measurements are explained quantitatively by the presented theory. Nevertheless in Fig. 9, where we present a fit of the theory to our measurement of the TR in a SmCo$_5$ sample (VACOMAX 200), the agreement is rather well, especially in comparison with Fig. 2 in ref. \cite{13}. If one calculates the hysteresis curve with the parameters fitted to the TR curve the agreement is not as well as for the TR. This is not astonishing, since the theory was designed mainly for the qualitative understanding of the TR, which is very sensitive to the field fluctuations but less sensitive to the switching field distribution. For the hysteresis the situation is vice versa. To
get a good description of the hysteresis the $H_s$ spectrum has to be determined more accurately. Another reason may be the simplification in the description of the grain interaction. Correlated switching of a multitude of grains is taken into account in a simplified manner via $n$ only, as discussed above. Surely this is more important for the hysteresis than for the TR. The neglection of the angular fluctuations of the field and the reversible rotation processes are in this regard of less importance, since this will influence both the hysteresis and the TR similarly. Since the present paper is focused on the theory, we save the detailed analysis of experimental results, as well as the modifications necessary to describe multi-phase magnets and the inverse TR in Ba-ferrites to forthcoming papers. In conclusion we may say that the presented theory is able to describe the rather complex findings by help of only three internal parameters, i.e. $\sigma_s$, $\sigma_f$, and $n$, which may be helpful in characterizing different hard magnetic materials more effectively.

Acknowledgement

We would like to thank V. Ivanov and K.H. Müller for useful discussions. One of us (L.J.) acknowledges the support by the Deutsche Forschungsgemeinschaft, Grant LO 293/1-1.

Appendices

A The magnetization of a weak grain

We want to find the mean internal magnetization of a grain in a MDS. For that purpose we assume, that the grain contains a lot of domains separated by $180^\circ$-domain walls. The dimension in field direction is large with respect to the dimension in a perpendicular direction. Thus the demagnetizing factor of such a slap-shaped domain is approximately zero. The average internal magnetization is then

$$\langle M \rangle_i = (1 - \lambda)M_S - \lambda M_S$$  \hspace{1cm} (A.1)$$

with $\lambda$ being the relative volume of the downwards magnetized domains. The magnetic energy density contains both the Zeeman energy

$$w_H = -\langle M \rangle_i H$$  \hspace{1cm} (A.2)$$
and the energy of the stray fields

$$w_{\text{stray}} = -\int H_{\text{dem}} \, d\langle M \rangle_i \quad (A.3)$$

Substituting for the demagnetization field

$$\mu_0 H_{\text{dem}} = -n (\langle M \rangle_i - \langle M \rangle) \quad (A.4)$$

an integration of eq. (A.3) yields the total energy density $w_{\text{tot}}$ in dependence on $\lambda$ to be

$$w_{\text{tot}} = -\frac{M_s^2}{\mu_0} \left( \frac{\mu_0 H}{M_S} + n \frac{\langle M \rangle}{M_S} \right) (1 - 2\lambda) + \frac{nM_s^2}{2\mu_0} (1 - 2\lambda)^2 \quad (A.5)$$

If the $\lambda$, which minimizes the energy, is inserted into eq. (A.1) one gets eq. (10), as long as $\lambda$ is between zero and one. Otherwise the grain is saturated in forward or backward direction. Thus the MDS grains show no memory effect, but follow the local field immediately.

**B Relation between $H_C$ and $\bar{H}_s$**

The influence of the weak grains is negligible, if $\mu_0 H_s \gg nM_S$ holds and the distribution width $\sigma_s$ is small enough. The first integral in eq. (22) may be neglected and in the second one we can extend the lower integration limit to minus infinity. If the external field $H_{\text{ext}}^{(1)}$ equals $-H_C$ we have $\langle M \rangle_1 = 0$ simultaneously. Therefore we find now

$$0 = -\int_{-\infty}^{\infty} dH_s \, g(H_s) \, \text{erf}(x_H) \quad (B.1)$$

Substituting $x = (H_s - \bar{H}_s)/\sigma_s$ the switching field distribution becomes symmetrical with respect to $x = 0$, thus the monotonous function $\text{erf}(x_H)$ has to be asymmetrical to reduce the integral to zero. Otherwise we find from eq. (21) that the erf-function is shifted, since we have $x_H = (-\bar{H}_s - \sigma_s x + nM_S/\mu_0 + H_C)/\sigma_f$. To make the erf-function asymmetrical, the shift has to be zero. This yields eq. (34).
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Captions of figures

Fig. 1. Scheme of the TR-experiment. From the saturated state the sample is isothermally driven to point “1” and afterwards along the recoil curve to point “2”. The TR happens, if the sample is heated (point “2” to point “T”), while the external field is kept constant (enhanced insert on the right hand side). For the external fields at point “1” and “2” the sheared remanence coercivity $H_{R}^{ext}$ and $H_{ext} = 0$ zero resp. were chosen, since the most TR-experiments start from the dc-demagnetized state.

Fig. 2. The grain is considered as an inclusion with magnetization $\pm M_{S}$ (here we assumed $-M_{S}$, symbolized by the long white arrow) embedded in an environment differing in its magnetic field $H$ from the mean magnetic field $\langle H \rangle$ within the sample due to the fluctuation $\Delta H$. Fluctuations of the magnetization in the environment are neglected, thus the magnetization in the environment is equal to the mean magnetization of the sample. This is symbolized by the two shorter white arrows of equal length.

Fig. 3. The demagnetization curve, the recoil curve, and a set of minor loops starting from different values of $\langle M \rangle_{1}$ calculated for a representative parameter set close to $SmCo_{5}$-magnets at 300K. Both the sample and the grains are adopted as spheres.

Fig. 4. The TR in dependence on (A) the width of the switching field distribution $\sigma_{s}$, (B) on the width of field fluctuations $\sigma_{f}$, and (C) on the internal demagnetization factor $n$.

Fig. 5. The TR for different external demagnetization factors $N$. For simplicity this figure was calculated with $\bar{H}_{s}(T) = H_{C}(T)$.

Fig. 6. The TR for different values of the initial temperature $T_{0}$ (parameters on the curves).

Fig. 7. The TR for different levels of approximation. The solid line gives the results of the presented theory. The dot-dashed line results from neglecting the weak grain fraction and for the calculation of the dashed line, both the weak grains and $p_{T}^{(2)}$, i.e. the probability, that a grain which resisted $H_{ext}^{(1)}$ will be switched while heating at $H_{ext}^{(2)} = 0$, were neglected.

Fig. 8. The probabilities $w_{\uparrow\uparrow}^{T}$, $w_{\uparrow\downarrow}^{T}$, $w_{\downarrow\uparrow}^{T}$, and $w_{\downarrow\downarrow}^{T}$ (cf. eqs. (37-40)) in dependence on the temperature. The dashed line shows the ratio of the volume of weak grains to the total volume.

Fig. 9. The TR and $H_{C}(T)$ measured for a SmCo$_{5}$ sample (VACOMAX 200) together with the calculated TR, where the parameters $\sigma_{s}$, $\sigma_{f}$, and $n$ have been adjusted accordingly.
Figure 1
Inclusion (grain)\n\[ H_{i+} = H - n(-M_S - \langle M \rangle)/\mu_0 \]

Environment\n\[ H = \langle H \rangle + \Delta H \]

Sample\n\[ \langle H \rangle = H_{\text{ext}} - N\langle M \rangle/\mu_0 \]

Figure 2
\( H_s = 3.0 T/\mu_0 \)
\( \sigma_s = 1.5 T/\mu_0 \)
\( \sigma_i = 0.5 T/\mu_0 \)
\( n = 0.333 \)
\( N = 0.333 \)
Figure 4
\[ \bar{H}_s = 3.0T/\mu_0 \]
\[ \sigma_s = 1.5T/\mu_0 \]
\[ \sigma_f = 0.5T/\mu_0 \]
\[ n = 0.333 \]

Figure 5
Figure 6

\[ \frac{\langle M \rangle}{T} \]

- \( M_S = 1.0 \, \text{T} \)
- \( H_s = 3.0 \, \text{T} / \mu_0 \)
- \( \sigma_s = 1.5 \, \text{T} / \mu_0 \)
- \( \sigma_f = 0.5 \, \text{T} / \mu_0 \)
- \( n = 0.333 \)
- \( N = 0.333 \)
\[
\begin{align*}
\bar{H}_s &= 3.0 T/\mu_0 \\
\sigma_s &= 1.5 T/\mu_0 \\
\sigma_f &= 0.5 T/\mu_0 \\
N &= 0.333 \\
n &= 0.333
\end{align*}
\]

Figure 7
with $H_s(T) = H_c(T)$

$M_s = 1.0 T$
$H_c = 2.667 T/\mu_0$
$N = 0.333$

$\sigma_s = 1.5 T/\mu_0$
$\sigma_i = 0.5 T/\mu_0$
$n = 0.333$

Figure 8
\[ M_s(300K) = 0.972 \text{T} \]
\[ H_C(300K) = 2.87 \text{T}/\mu_0 \]
\[ N = 0.53 \]
\[ \sigma_s(300K) = 1.60 \text{T}/\mu_0 \]
\[ \sigma_f(300K) = 0.91 \text{T}/\mu_0 \]
\[ n = 0.56 \]