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Field dependence of the thermal remagnetization in sintered hard magnets

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Abstract

The dependence of the thermal remagnetization (TR) in sintered hard magnets on external magnetic fields has been investigated in order to determine more precisely the grain-interaction fields in the DC-demagnetized state. That negative external field at which the TR or its initial slope vanishes or changes its sign, corresponds well to the theoretical parameter σ_f, i.e. the distribution width of internal field fluctuations. For the "normal" TR of the metallic magnets $SmCo_5$, Sm_2Co_{17} and NdFeB these fields are of the order of −0.5, 0.25 and −0.04 T, respectively. For the "inverse" TR at the hard ferrites this external field is about −0.02 T. The systematic variation of the TR-curves with a small external field is well explained by our theoretical model and leads to a refinement of the parameters for $SmCo_5$ and barium ferrite. Susceptibility measurements with small alternating fields, carried out at different points of the TR curve, as well as repeating TR-experiments at $SmCo_5$ demonstrate clearly that the TR is mainly due to the contribution of the "weak" grains, remagnetizing in the interaction fields of the "hard" grains.

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Keywords: Permanent magnets; Thermal remagnetization; Grain interaction field; Switching probability; Switching field distribution

1. Introduction

Grain interaction in different types of polycrystalline uniaxial well-aligned hard magnets is studied by measurement and calculation of the thermal remagnetization (TR) and its dependence on small external fields. If a polycrystalline sintered RE/T hard magnet is DC-demagnetized isothermally at a low initial temperature T_0 and later on heated up to T_{max}, the remanence can increase. The TR [1–11], especially high at sintered polycrystalline $SmCo_5$-magnets [1,10], was shown to be dependent on the initial temperature [11] and the sample demagnetization factor [10].

Because the TR is strongly related to the temperature dependence of the coercivity, an "inverse" TR occurs by cooling in the case of barium ferrite [3,12,13]. The TR is mainly due to "grain" interaction, described theoretically by the

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internal "grain-demagnetization factor" n and the distribution width σ_f of the local magnetic fields [10]. Besides these two parameters a broad distribution of the switching fields with the width σ_s, a low-sample demagnetization factor N and low initial temperatures T_0 [11] result in large TR-effects, influenced essentially by the temperature dependence of the saturation magnetization [10]. Only in the case of $SmCo_5$, the latter is further characterized by a large difference $T_c - T_{H_c}$ (at T_{H_c} the coercivity vanishes) [14], which is one of the reasons for the high TR-effects in that material [10]. The relative maximum remanence enhancement for comparable low-sample demagnetization factors $N \approx 0.1$ and sufficiently low or optimal initial temperatures is found to be more than 50% for $SmCo_5$ [9–11], about 20% for sintered polycrystalline barium ferrite [9,12,13], about 10% for NdFeB [9] and only about 3% for Sm_2Co_{17} [9]. In order to prove the basic assumptions of the theory [10], we investigated in this work the dependence of the TR on a small external steady field H_{ext}, which is applied during the TR experiment. We will determine that external field, where the TR-maximum and its initial slope vanishes and changes its sign, respectively in order to find a direct measure for the grain interaction. Furthermore, we vary the sign of a small external field during heating and find that temperature, where the susceptibility begins to increase drastically. Thus, we will show the influence of the "weak" (i.e. Bloch wall containing) grains on the TR. In order to study the influence of the low coercive part of the switching field distribution on the TR, we repeated the TR-experiments by cooling down the sample to the initial temperature T_0. Afterwards, the remanence was reduced to zero by an opposite field, followed by heating once again.

2. Experimental results

We investigated well-aligned sintered standard magnets of $SmCo_5$ (Vacomax 170), Sm_2Co_{17} (Vacomax 225), NdFeB (Vacodym 510 HR) as well as sintered barium ferrite, all with high and low values of the sample demagnetization factor N (discs and rods). The magnetic measurements have been performed in an open circuit by an Oxford Mag-Lab (14 T) vibrating sample magnetometer in the high-temperature modification. The temperature was directly measured at the sample by means of a thermocouple. As usual [1–11] the normal TR-curve is measured as remanence in zero external field starting in the DC-demagnetized state at a sufficient low initial temperature T_0 by slowly heating the sample (cf. Ref. [11]).

2.1. *Influence of small external fields on the TR*

To study the influence of a small external field H_{ext} on the TR, the TR-curves were started at a defined point of the recoil curve, which was prepared at temperature T_0 as follows. After saturating the sample in a field of about 10 T it was demagnetized by help of an opposite steady field H_1. If this field equals the remanence coercivity H_R the magnetization will go to zero after switching off the field. Otherwise, if H_1 is larger than the coercivity H_C but a little bit smaller than H_R, the demagnetized state will be achieved on the recoil curve for a small negative (i.e. opposite to the initial saturation direction) residual field $H_{ext} < 0$. In case the demagnetizing field is chosen a little bit larger than H_R, a small positive field $H_{ext} > 0$ is necessary to achieve the demagnetized state along the recoil curve. The field H_{ext} was then kept constant while the sample was heated and the resulting remanence enhancement $\Delta M_{TR}(T)$ (TR-curve) was recorded. A set of such measured (points) and calculated (solid lines) TR-curves is given in Fig. 1 for $SmCo_5$. The initial temperature T_0 was 250 K. The cases with $H_{ext} = 0$, i.e. $H_1 = -H_R$, correspond to a "normal" TR-curve. We observe a systematical shift of the maximum temperature T_{max}, the maximum remanence enhancement $\Delta M_{TR,max}$ and also of the initial slope $\Delta M_{TR}(T)/\Delta T$ (Fig. 2) with increasing negative external field. We determined the three parameters used in the theory [10] by fitting the curve with $H_{ext} = 0$ (cf. Table 1). The other theoretical lines are then calculated with that parameter set.

With increasing negative external field not only the TR-maximum and the temperature of this maximum, T_{max}, but also the initial slope of the

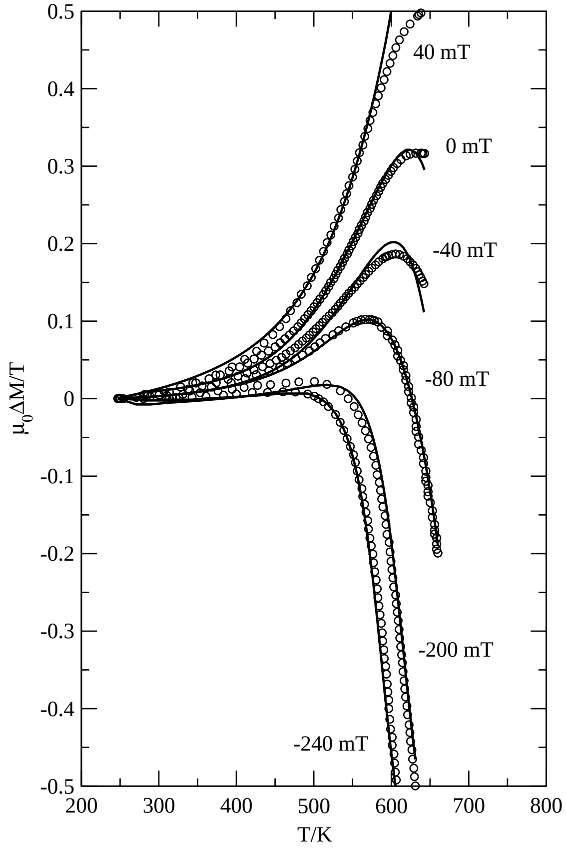

Fig. 1. Measured TR-curves $\Delta M(T)$ (circles) for a $SmCo_5$-magnet ($N = 0.1$, VACOMAX 170) for different external fields applied while heating. The solid lines are model calculations with parameters determined by the $H_{ext} = 0$-curve (cf. Table 1).

TR-curves drops down as shown in Fig. 2. The straight lines were calculated by means of the parameters of Table 1. The field dependencies of $\Delta M_{TR,max}$ and T_{max}, as derived from Fig. 1 for $SmCo_5$ and in an analogous way for NdFeB and Sm_2Co_{17}, are given in the Figs. 3 and 4. These curves depend slightly on the initial temperature T_0 and on the sample demagnetization factor N. However, the negative residual field, for which either the TR-maximum (Fig. 3) or the initial slope (Fig. 2) of the TR-curve vanish, are found to be a characteristic of the permanent magnet at the DC-demagnetized state at the temperature T_0. We call these two suppression fields H_{um} and H_{us}, respectively.

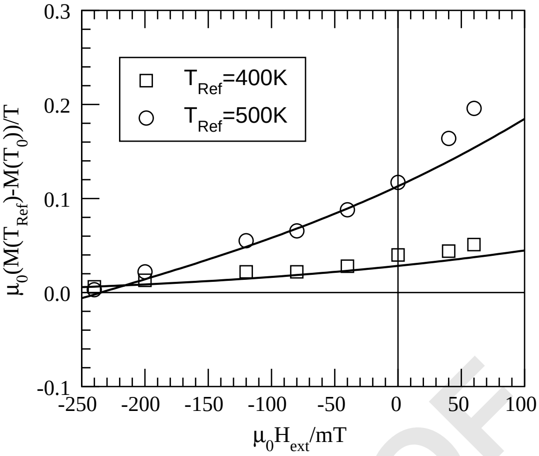

Fig. 2. Calculated field dependence (solid lines) of the initial TR-slope between the initial temperature T_0 and different reference temperatures T_{Ref} by the parameters of Table 1. The symbols represent the measured values derived from Fig. 1

Table 1
Fitted parameters σ_s (width of switching field distribution), σ_f (width of field fluctuations in the DC-demagnetized state), and n (internal mean "grain"-demagnetization factor) $SmCo_5$ ($T_0 = 250$ K; $N = 0.1$)

μ_0H_{ext}/mT	μ_0/T	$\mu_0\sigma$f/T	n
0	1.50	0.47	0.35

2.2. *Susceptibility*

For the $SmCo_5$-sample as in Fig. 1, we started once more in the DC-demagnetized state and during the registration of a TR-curve we superimposed a very small field H_{ext} of 10 mT, which we switched in sign approximately every 10 K. As shown in Fig. 5, we obtain a "normal" and two slightly shifted TR-curves. The latter became obvious above 550 K only. From the difference of the two shifted curves we derive the susceptibility, which continuously increases during heating (Figs. 5 and 6). The susceptibility χ has been obtained by back-shearing, i.e. $\chi = \chi'/(1 - N\chi')$, where $\chi' = \Delta M/\Delta H_{ext}$ and N is the sample demagnetization factor. Exactly the same shifted TR- and susceptibility-curves were calculated by means of the parameters, given in Table 2 (Fig. 5).

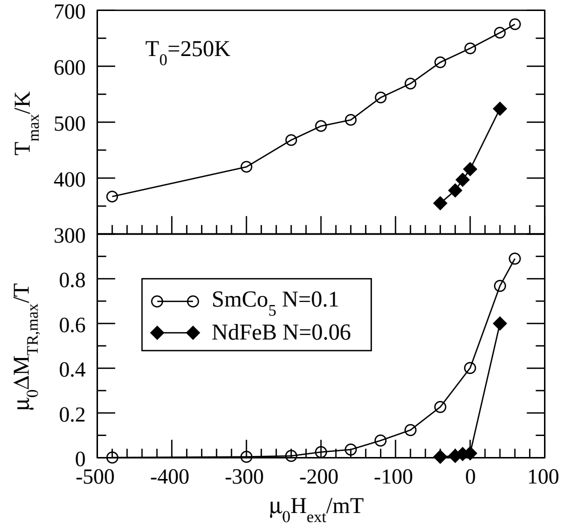

Fig. 3. Comparison of the field dependencies of the TR-maximum and the temperature T_{max} between a $SmCo_5$- and an NdFeB-magnet.

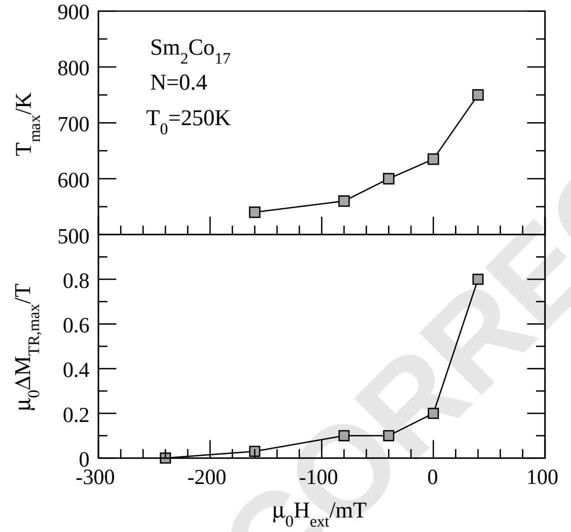

Fig. 4. The field dependencies of the TR-maximum and the temperature T_{max} of a Sm_2Co_{17}-magnet with $N = 0.4$.

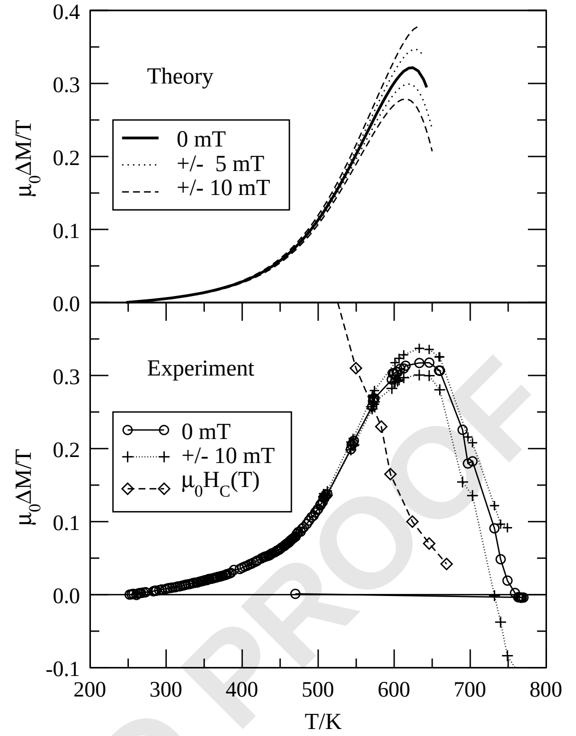

Fig. 5. Upper plot: Calculated TR curves for $\mu_0 H_{ext} = \pm 5\,mT$ and $\mu_0 H_{ext} = \pm 10\,mT$ concerning the $SmCo_5$-magnet as in Fig. 1. The solid line gives the fit curve for $\mu_0 H_{ext} = 0$. Lower plot: Measured TR curves for $\mu_0 H_{ext} = \pm 10\,mT$. The circles represent the values measured for $\mu_0 H_{ext} = 0$. Furthermore the $H_c(T)$, which was also used for the calculation, is indicated.

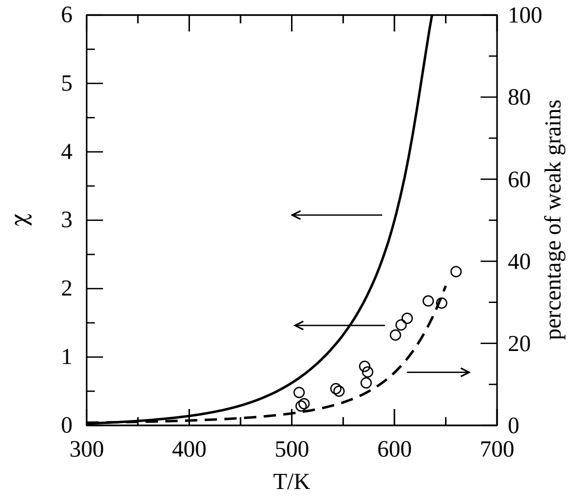

Fig. 6. Measured and calculated susceptibility (solid line) $\mu_0 H_{ext} = \pm 10\,mT$ for the $SmCo_5$-magnet from Fig. 1. Furthermore, the calculated percentage of “weak” grains is shown.

Table 2
Suppression fields H_{um} and H_{us} respectively, determined for the four types of sintered hard magnets in this work, besides other TR-characteristics [9–11,13]: Maximum TR, related to $J_s(T_0)$ for comparable external sample demagnetization factors N (≈ 0.1), starting temperature of the TR-experiment T_0, the widths of switching field- and local internal field-distribution σ_s and σ_f, respectively, and the "grain" demagnetization factor n (fitted from experiments by the theory [10])

	$SmCo_5$	$BaFe_{12}O_{19}$	NdFeB	Sm_2Co_{17}
$-\mu_0 H_{um}$ (T)	0.5		0.02	0.5
$-\mu_0 H_{us}$ (T)	0.25	0.02		
$\mu_0 M_S(T_0)$ (T)	0.97	0.1	1.5	1.15
TR_{max} (%)	33	23	12	3
σ_f (T)	0.41	0.05–0.1	—	—
n	0.33	0.5	—	—
σ_s (T)	1.9	0.1	—	
T_0 (K)	250	550	150	150

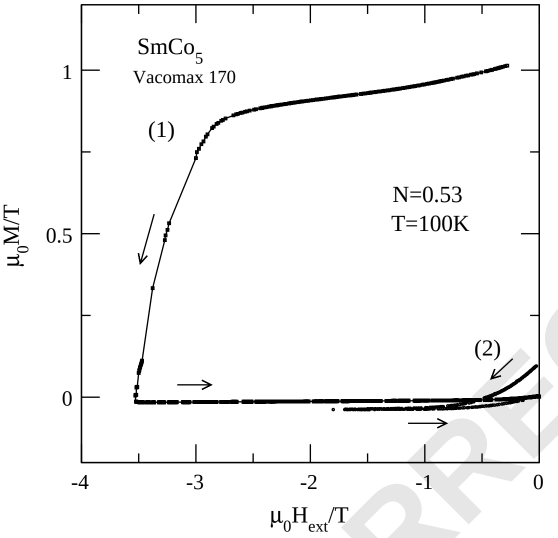

Fig. 7. Isothermal demagnetization curve: (1) and after the first TR-experiment (2), $SmCo_5$-magnet as in Fig. 1.

We mention, that by increasing the temperature far enough above the TR-maximum till the TR goes down to zero this experiment allows to control the "zero" field and the zero remanence of the superconducting magnet with an error of 1 mT, well indicated if the remanence remains zero by cooling down the sample.

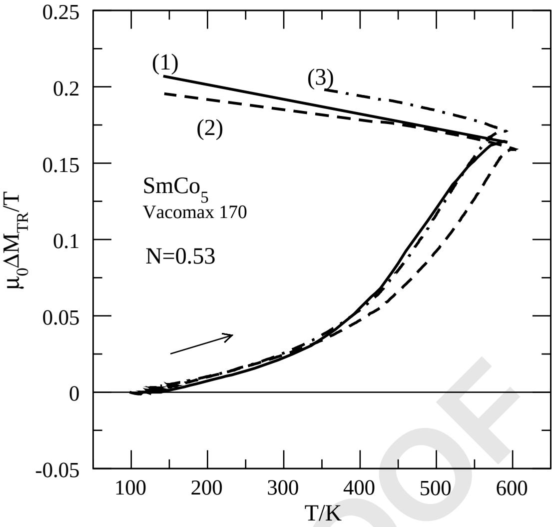

Fig. 8. $SmCo_5$ (Vacomax 170 as in Fig. 1) TR-curve and two repeated TR-curves (2) and (3).

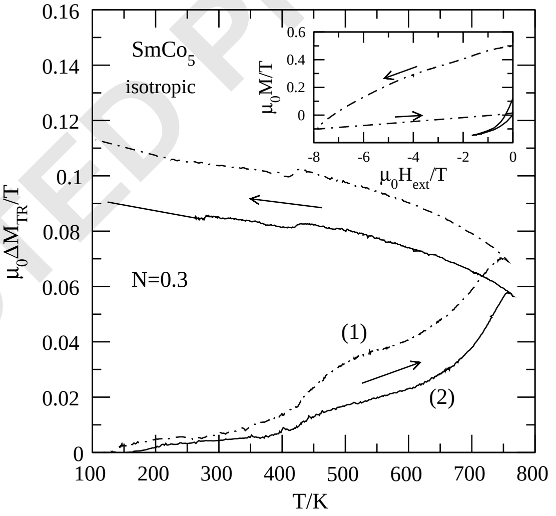

Fig. 9. First and second TR-curve for an isotropic sintered magnet ($SmCo_5$, quality as Vacomax 170 without field pressing). The inset shows the isothermal demagnetization curve before (1) an after the TR-experiment (2).

2.3. Repeated TR-experiments

Following the first TR-experiment in zero external field, as shown in Figs. 1 or 5, the sample is cooled down once more from the first TR-maximum to the initial temperature T_0 and then again the isothermal “demagnetization curve” is measured up to the remanence coercivity (DC-demagnetized state (Fig. 7), which is now essentially lower. The temperature of the sample is again increased and the TR-curve is measured a second (curve (2) in Fig. 8) and third time (curve (3)). These three TR-curves are nearly identically. A qualitative similar behaviour is observed at the isotropic $SmCo_5$-sample (cf. Fig. 9). Fig. 7 and the inset in Fig. 9 show the significant change in the demagnetization curve after the first TR-experiment. However, the differences in the two TR-curves, given in Fig. 8, are again quite small.

3. Discussion

The TR-experiments were started at T_0 in the DC-demagnetized state, where a maximum of inhomogeneity and fluctuations of the magnetization and consequently of the local internal field exist [10]. The analysis of the experiments on the field dependence of the TR-curves allows a more precise determination of the three fitting parameters (n, σ_f and σ_s). In Table 2 we give the fit values for a $SmCo_5$-sample for one external field $H_{ext} = 0$. By means of these values, we calculated the TR-curves for the other external fields, lines in Fig. 1, using the theory [10]. The agreement between the lines and the experimental points in Fig. 1 demonstrate ocularly the good quality of the fits. As shown in the Figs. 2–4 a typical negative external field, H_{um} or H_{us} respectively exists, at which the TR-effect vanishes. These suppression fields concerning the TR-maximum or its initial slope, respectively depend only slightly on the initial temperature T_0. Therefore, we can conclude that H_{um} or H_{us} are properties of the magnet and a direct measure for the characteristic interaction field in the DC-demagnetized state. We found for the more homogeneous materials $SmCo_5$ and barium ferrite, that they are of the same order as the parameter σ_f, the width of the internal field distributions (Table 3). Besides the known [9] large difference in the TR-amount (cf. the TR_{max} values in Table 3), also a high difference in the suppression field exists between $SmCo_5$ and NdFeB, which are, together with the hard ferrites, considered as “nucleation controlled”. On the other hand the TR-behaviour of the “pinning controlled” magnet Sm_2Co_{17} is out of the scope of the theory [10]. However, the field dependence of the (low) TR-maximum (cf. Figs. 3 and 4) shows qualitatively the same form as for $SmCo_5$, which hints also on local field fluctuations in the DC-demagnetized state.

Table 3

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From the TR-experiment with the alternating 10 mT field at $SmCo_5$ and the derived susceptibility we learn, that the initial TR on the first cycle is caused more by “hard grain switching” ($\mu_0 H_s > \mu_0 n M_s \gg 10\,\mathrm{mT}$; $n =$ “grain” demagnetization factor). This means for $SmCo_5$, that “weak” grains, which go into a multi-domain state, due to switching fields less than nM_s, become dominant in the range above 550 K during increasing temperature only. This also follows from the calculated temperature-dependent switching probabilities [10]. On the other hand, TR-repeating experiments from the TR-maximum show that the weak part of the switching field distribution is responsible for the second (and third) TR-cycle. This shows that small to mediate TR can be caused either by switching of single-domain grains with low switching fields due to the stray fields of the hard grain fraction or due to Bloch-wall movement due to mean field effects in Bloch-wall containing grains. This behaviour is observed qualitatively also for the isotropic sample of $SmCo_5$ in Fig. 9. The TR of the sintered sample with isotropic grain orientation is reduced roughly four times in comparison with the well aligned sample with comparable sample demagnetization factor N. This may be understood, since both the magnetic field component and the remanence are reduced by a factor 2 due to the misalignment of the grains. The large difference in the TR-characteristics between $SmCo_5$ and NdFeB, which both are called “nucleation controlled” has the following reasons:

Due to the separation of the main phase grains by a non-magnetic surrounding the magneto-static interaction is reduced in the heterogeneous NdFeB magnets. This becomes effective both in the mean field, where the non-magnetic grains have to be taken into account, as was done in Ref. [4], and in a reduction of the field fluctuations. In the more homogeneous $SmCo_5$-magnets, the observed interaction fields of approximately 0.5 T are 20 times stronger than in NdFeB, which implies that contributions of exchange interactions between adjacent grains and/or a mechanism of large inhomogeneous demagnetization fields [15,16] may be occurring. Furthermore, the broader switching field distribution, indicated by σ_s, the mentioned high difference $(T_c - T_{H_c}) \approx 250\,K$ [14] in combination with the theoretical expression for the switching field $H_s \approx H_c + nM_s$ [10] are responsible for the higher TR-maxima and surviving of a measurable TR-effect up to 100 K above T_{H_c} [9].

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