

Modeling the Training Effect in Exchange-Biased Bilayers for Large Numbers of Magnetization Reversal Cycles

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The exchange bias (EB) is a unidirectional anisotropy that occurs, e.g., upon field-cooling ferromagnet/antiferromagnet systems. In many material systems, the EB field is reduced from one hysteresis loop to the next measurement. This so-called training effect (TE) has been investigated in experiments and by means of theoretical efforts by many research groups. The reduction of the EB field as a result of subsequent magnetization reversal processes is often fitted by a power law, usually with the exception of n = 1, or with an equation based on the discretized Landau–Khalatnikov equation, as first suggested by Binek. Few other models, usually with more fitting parameters, have been proposed yet. Herein, it is shown that for large numbers of subsequent magnetization reversal processes in Co/CoO thin film samples, a modified power law or a logarithmic fit can model the TE in most cases as well as the abovementioned, commonly used models.

1. Introduction

The exchange bias (EB) is a unidirectional anisotropy field, which was first observed in Co/CoO core/shell nanoparticles.^[1,2] It occurs typically in ferro-/antiferromagnetic (FM/AFM) thin film systems or nanostructures after cooling in the presence of magnetic field (so-called field-cooling) the system through the Néel temperature of the AFM, while there are also several systems containing ferrimagnets as one of the components, or even single-phase materials, in which surface effects result in an EB.^[3,4] The most prominent feature is a horizontal shift of

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/pssb.202300435.

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DOI: 10.1002/pssb.202300435

the hysteresis loop, usually opposite to the cooling field direction. However, there are also reports about a broadening of the hysteresis loop, a vertical loop shift, or an asymmetry of the loop.^[5,6]

Another often reported feature, associated with the EB phenomenon, is the training effect (TE), i.e., a reduction of the EB shift upon repeated measurements of hysteresis loops without new field cooling.^[7-10] While this effect can be very strong in some systems, there are other material combinations where it is quite weak^[11] or even completely absent.^[12]

Depending on the system, varying explanations for the TE have been proposed. For $Co/Co_{1-\gamma}O$, Beschoten et al. concluded from the existence of a TE that

magnetization reversal in the FM layer was neither spatially homogeneous nor reversible and that with each reversal cycle, the AFM domain structure was rearranged, resulting in a partial loss of the AFM net magnetization with each measured hysteresis loop, which led to a reduction of the EB field with each measurement cycle.^[7] A slightly different explanation for the TE in Co/CoO was given by Radu et al. who assumed the AFM to be in a single domain state with AFM and FM spins being oriented (anti)parallel directly after field cooling, while a stable AFM multidomain state was reached after the first hysteresis measurement, without changing the alignment of the FM domains.^[13,14] Ali et al. also attributed the TE in Co/CoO to irreversible changes in the AFM domain state magnetization during hysteresis measurements.^[15] Guo et al. described the TE more generally as to stem from relaxation processes of AFM spin configurations, where cycling reduces the number of beneficial interface spin.^[16]

In the same material system, Popova et al. used off-specular neutron reflectometry measurements to investigate magnetization reversal and found that the reversal mechanism along the descending branch changed after the first hysteresis loop.^[17] For the descending branch, they measured domain wall nucleation and propagation for the first loop, while parts of the magnetization were reversed by rotation for subsequent loops. Dias et al. ascribed the TE to variations of the rotatable anisotropy parameters.^[18]

Some authors differentiated between athermal and thermal training, where a large TE from the first to the second hysteresis loop is usually interpreted as athermal and subsequent smaller TE as thermal training. Kaeswurm and O'Grady interpreted the athermal TE as stemming only from the interface spin configuration, while the thermal TE is attributed to thermal instability in the AFM bulk.^[19] Biternas et al. distinguished athermal training

at low temperatures and thermal training at high temperatures^[20] and found athermal effects for large AFM dilutions (i.e., large numbers of defects) and thick AFM layers, while low AFM dilutions and thin AFM layers resulted in thermal training in their experiments.^[21] Hussain and Reddy reported a change from athermal to thermal training for increasing FM layer thickness in Co/CoO bilayers.^[22]

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Corresponding to the different reports about athermal and thermal training and the varying interpretations of the TE, there are a few models, which are commonly used to fit the TE, i.e., the dependence of the EB field on the number of cycles *n*. An early suggestion from Paccard et al. is the so-called power-law, resulting in a fitting equation proportional to the square root of *n*, based on the assumption of fluctuations in the FM/AFM coupling in different interface regions, which change the magnetic interactions with each reversal^[23]

$$H_{\rm EB}^n = H_{\rm EB}^\infty + A \cdot n^{-1/2}$$
 (1)

where *A* is the fitting parameter and H_{EB}^{∞} is the saturated EB field after infinite number of cycles.

For a relatively large number of 50 cycles, they reported a good agreement between measurement and theory in Co/CoO, NiFe/NiFeMn, and NiFe/Cr₂O₃ systems.

This power law has since then been used in many studies and usually showed good agreement with the measurement of data for n > 1. This is why recently a small modification of the power law was suggested, based on asymmetric changes in descending and ascending branches^[24]

$$H_{\rm C1(2)}^n = H_{\rm C1(2)}^\infty + k_{1(2)} \left(n + n_0^{1(2)}\right)^{-1/2} \tag{2}$$

with the coercive fields $H_{C1(2)}^n$ (index 1 for descending and 2 for ascending), the values for infinite magnetization reversal processes $H_{C1(2)}^\infty$, fitting parameters $k_{1(2)}$, and initial states $n_0^{1(2)}$ for the respective branches.

Rui et al. showed for up to 20 measured field cycles in Ta(4 nm)/Fe₁₁Au₈₉(50 nm)/FeNi(5 nm)/Ta(2 nm) that the modified power law resulted in a much better fit for n = 1 than the original power law.^[24] Shi et al. tested NiFe/FeMn, Fe_{0.51}Cr_{0.49}/IrMn, and epitaxial Fe/CoO for 30–150 cycles and also found the modified power law to give a better fit to the measured values than the original power law, while the $1/n^{1/2}$ plot still showed systematic deviations between measurements and fit function.^[25]

Another approach was suggested by Binek who derived an implicit formula for the dependence of the EB field on the number of loops from the discretized Landau–Khalatnikov equation under the assumption that the energy landscape controlling the relaxation process depends only on the difference of the AFM interface magnetization from its equilibrium^[26]

$$\mu_0[H_{\rm EB}(n+1) - H_{\rm EB}(n)] = -\gamma \{\mu_0[H_{\rm EB}(n) - H_{\rm EB}^{\infty}]\}^3$$
(3a)

with the magnetic permeability μ_0 , the EB field H_{EB} for *n* and (n + 1) as well as the equilibrium EB field H_{EB}^{∞} for an infinite number of reversal processes, and a temperature-dependent fitting parameter γ . In subsequent publications, Binek et al. showed

Since implicit fit functions are usually harder to implement and more time-consuming than explicit ones, here, we suggest Equation (3b) as an explicit estimation of the implicit Equation (3a). The derivation of Equation (3b) is given in the Supporting Information.

$$\Rightarrow H_{\rm EB}(n) = H_{\rm EB}^{\infty} \pm \sqrt{\frac{1}{2(\gamma \mu_0^2 n - c)}}$$
(3b)

with an integration constant *c*. Equations (3a) and (3b) are a little further apart in the single-digit range for n due to the transformation of a discrete equation into a continuous one. As n > 10increases, the equations become more equal. It should be mentioned that this formula is guite similar to the modified power law (Equation (2)) and thus can be expected to give fits of similar accuracy. In his derivation of Equation (3a), Binek himself showed that already at n = 3, his equation was approximated very well by the power law (Equation (1)).^[26] However, the explicit Equation (3b) cannot be expected to work as well as the implicit Binek formula (Equation (3a)) for systems in which large changes can be found between the first and the second data point in a TE measurement, i.e., Equation (3b) can only be used as a substitute for Equation (3a) in case of relatively small changes between the first and the second hysteresis loop measured after field cooling.

The Binek model has been used in diverse publications since. Mostly, 5–20 field cycles are measured, while the systems vary in a broad range of material combinations, such as Co/NiO/[Co/Pt],^[29] La_{0.7}Sr_{0.3}MnO₃/SrRuO₃,^[30] Ba₂ScRuO₆,^[31] Co/CoO,^[9,32] or NiFe₂O₄/NiO.^[33] A slight modification of the Binek model, with an additional higher-order term, was proposed by Wu et al. who fitted 10 cycles of magnetization reversal in Co/CoO.^[34]

Generally, more fitting parameters give more degrees of freedom and thus may result in easier fitting of measurement data. Nevertheless, the aforementioned idea of combined physical effects being responsible for the TE may require more than one or two fitting parameters. Mishra et al. suggested a new formula based on the assumption that frozen and rotating AFM interface spins interact with the reversed FM magnetization, and that mixed FM and AFM coupled components show different contributions to the TE, resulting in 5 fit parameters^[35]

$$H_{\rm EB}^n = H_{\rm EB}^\infty + A_{\rm f} \exp\left(-\frac{n}{P_{\rm f}}\right) + A_i \exp\left(-\frac{n}{P_i}\right) \tag{4}$$

with the EB field of the *n*th hysteresis loop H_{EB}^n , the equilibrium EB field H_{EB}^∞ reached after infinite magnetization reversal processes, parameters related to the change of the frozen spins A_f and P_f , and parameters related to the change of the interface magnetic frustration A_i and P_i .

This equation worked well for fitting the TE in NiFe/IrMn (14 cycles),^[35] phase-separated polycrystalline $Sm_{0.1}Ca_{0.7}Sr_{0.2}MnO_3$ (12 cycles),^[36] or $La_2Cu_{0.9}Fe_{0.1}IrO_6$ (5 cycles)^[37]; however, it has the aforementioned disadvantage of five fitting parameters.

Thus, a much simpler idea shall not be ignored, based on an assumed wide distribution of energy barriers resulting in an ln(t) law regarding the time dependence of the magnetization.^[38]

The thermomagnetic relaxation is often modeled logarithmically.^[12,39–41] For the TE, however, such a logarithmic relaxation has rarely been applied, e.g., for 50 cycles of magnetization reversal in FeNi/Cu/Co/FeMn^[42]

$$H_{\rm EB}^n = A + B \cdot \ln(n) \tag{5}$$

with fitting constants *A* and *B*. Besides n = 1, this model was found to fit the measured values better than the power law or an exponential function.

Here, we compare most of the aforementioned models for two different Co/CoO thin film samples. Equation (4) was not included in our tests due to the large number of fitting parameters (five), and due to the problem, that both exponential functions have a symmetric form, i.e., a differentiation between frozen and interface frustrated spins is not possible according to this fit function. The Binek function was modeled according to the original Equation (3a) as well as according to Equation (3b) which can easier be implemented into a fit than the implicit function in Equation (3a). We show that the modified power law, the implicit and explicit Binek functions as well as the logarithmic fit provide good fits of the experimental data for up to 100 or 1000 cycles, respectively.

2. Results and Discussion

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The measurement of the TE of the MgO(110) sample showed a strong difference between the first and the second value (**Figure 1**), suggesting fitting according to the Binek formula (Equations (3a) and (3b)). Besides this formula, Figure 1 shows the EB values of all 100 magnetization reversal cycles with the aforementioned fitting equations, without Equation (4) due to its large amount of necessary fitting parameters. The coefficients of determination are $R^2 = 0.973$ (Equation (1)), 0.978 (Equation (2)), 0.991 (Equation (3a)), 0.978 (Equation (3b)), and 0.993 (Equation (5)), respectively. These values show that the modified power law is not advantageous compared to the original power law. The implicit Binek fit (Equation (3a)) and the logarithmic function fit the measurement values best. It must be mentioned that, in contrast to ref. [42], here a slightly modified



Figure 1. EB of the MgO(110) sample measured during 100 magnetization reversal cycles with power law fit (Equation (1)), modified power law (Equation (2)), implicit Binek fit (Equation (3a)), explicit Binek fit (Equation (3b)), and logarithmic fit (Equation (5)).

function $H_{EB}^n = A + B \cdot \ln(n - n_0)$ was used with $n_0 = -0.995$ to enable fitting the first parameter properly, similar to the modification of the original power law given in Equation (2). In this way, the first EB value can be chosen arbitrarily, since the logarithm function diverges when the argument approaches 0.

Similarly, **Figure 2** shows measurements performed up to 1000 magnetization reversal cycles of the sapphire(0001) sample. The coefficients of determination are $R^2 = 0.880$ (Equation (1)), 0.977 (Equation (2)), 0.994 (Equation (3a)), 0.977 (Equation (3b)), and 0.992 (Equation (5)), respectively. Again, the logarithmic model (Equation (5)) and the implicit Binek model (Equation (3a)) fit the measurements best, while the modified power law (Equation (2)) and the explicit Binek fit (Equation (3b)) are identical, both only slightly lower values. Here, in contrast to the MgO(110) measurement series, the modified power law works much better than the original power law.

On the one hand, these results show that a logarithmic fit should be taken into account when the TE is modeled for large numbers of magnetization reversal processes. On the other hand, the different goodness of fit for both examined Co/Co_3O_4 samples suggests further investigations with more samples, measuring the TE for large numbers of magnetization reversal cycles. This could be used to evaluate whether the agreement of the experimental results with either model allows for the determination of physical differences between them, equivalent to the aforementioned different theories for the TE in different material systems.

Due to some authors' remarks about different effects in the ascending and descending branches of the hysteresis loop,^[17,36] we also investigated the left and right coercive fields for these samples to find out whether different effects are superposed in the measured EB. **Figure 3** depicts both coercive fields of the sapphire (SAP) sample with the corresponding fits. The coefficients of determination are for the left coercive field: $R^2 = 0.924$ (Equation (1)), 0.989 (Equation (2)), 0.995 (Equation (3a)), 0.989 (Equation (3b)), and 0.983 (Equation (5)), respectively; for the right coercive field: $R^2 = 0.973$ (Equation (1)), 0.980 (Equation (3a)), 0.995 (Equation (3b)), and 0.941 (Equation (5)), respectively. While the modified power law (Equation (2)) and the implicit (Equation (3a)) and explicit



Figure 2. EB of the sapphire(0001) sample measured during 1000 magnetization reversal cycles with power law fit (Equation (1)), modified power law (Equation (2)), implicit Binek fit (Equation (3a)), explicit Binek fit (Equation (3b)), and logarithmic fit (Equation (5)).







Figure 3. a) Left coercive field H_{CL} and b) right coercive field H_{CR} of the SAP sample with corresponding fits: power law fit (Equation (1)), modified power law (Equation (2)), implicit Binek fit (Equation (3a)), explicit Binek fit (Equation (3b)), and logarithmic fit (Equation (5)).



Figure 4. a) Left coercive field H_{CL} of the MgO sample with power law fit (Equation (1)), modified power law (Equation (2)), implicit Binek fit (Equation (3a)), explicit Binek fit (Equation (3b)), and logarithmic fit (Equation (5)); b) right coercive field H_{CR} of the MgO sample.

Binek fits (Equation (3b)) show very good agreement with the measured data in both cases, the common power law is better suitable than the logarithmic fit for the right coercive field and vice versa for the left coercive field. However, in both cases, the dependence of the coercive fields on the number of loops is quite similar.

Unexpectedly, this is completely different for the MgO(110) sample, as shown in Figure 4. The left coercive field can be fitted similarly to the previous graphs, with the coefficients of determination $R^2 = 0.965$ (Equation (1)), 0.980 (Equation (2)), 0.987 (Equation (3a)), 0.980 (Equation (3b)), and 0.985 (Equation (5)), respectively, where the logarithmic fit, the modified power law model, and both Binek fits are best suited to fit the measured data. The right coercive field, however, is nearly constant, and even slightly increasing, as opposed to the strongly decreasing value of H_{CR} in case of the SAP sample. According to Radu et al.^[13,14] this may be interpreted as magnetization reversal via domain rotation on the right side of the loop in the MgO sample, while a strong change of a coercive field from the first to the second loop was often attributed to a change from domain wall processes.^[17] This suggests that the left branch of the MgO sample and both sides of the hysteresis loop of the SAP sample show a change from domain wall processes to an increasing amount of magnetization rotation with each cycle.

The comparison of both Co/Co_3O_4 samples under examination shows that not only the TE of the EB should be investigated further, but also evaluating both coercive fields separately may also give more information about magnetization reversal processes, although it is only scarcely shown in the literature.^[36]

3. Conclusion

To conclude, we have measured the TE of the EB in two epitaxial Co/Co_3O_4 systems of different crystal orientations. In both cases, the best fits were given by the implicit Binek function as well as a logarithmic function, which has so far only rarely been suggested for the TE. This finding and the varying quality of the other tested fit functions for both samples under investigation suggest performing further TE measurements with 100 or more cycles for different material systems to evaluate whether the quality of the different fit functions can be correlated with different TE theories.

Besides, separate evaluation of left and right coercive fields shows that no TE occurs on the right side of the MgO(110)/ $Co_3O_4(110)/Co(110)$ sample, while the Al₂O₃(0001)/twinned $Co_3O_4(111)/$ twinned Co(111) shows a pronounced TE on both sides of the loop, indicating a different magnetization reversal

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process on the right side of the MgO(110)/Co₃O₄(110)/Co(110) sample than in the other cases.

In general, phenomena described by logarithmic relationships may occur in the case of irreversible and noncyclic changes. The TE considered in the investigated magnetic EB system is an example of such situation where magnetization scales logarithmically with the number of magnetization reversal processes in an imperfect sample.

4. Experimental Section

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Two samples were grown epitaxially by molecular beam epitaxy (MBE) on different substrates with lateral dimensions 10 mm \times 5 mm: 1) MgO(110) substrate/Co₃O₄(110) (20 nm)/Co(110) (6 nm)/Cu cap and 2)Al₂O₃ sapphire(0001) substrate/ twinned Co₃O₄ (111) (20 nm)/ twinned Co(111) (6 nm)/Cu cap.

The Co₃O₄ layer was grown at an oxygen partial pressure of $p(O_2) = 5 \times 10^{-6}$ mbar in the MBE chamber. Samples containing Co₃O₄ can be expected to show a larger EB and also a larger TE than samples with CoO as AFM.^[7]

Superconducting quantum interferometry measurements were performed by field cooling the samples in an external magnetic field of 1 T in the sample plane to T = 20 K.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

T.B. acknowledges the partial support from the local SUT (grant no. 14/030/RGJ23/0221).

Open Access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

All data used for this paper are given in the paper and its annex.

Keywords

Binek fit, Co/CoO, exchange bias, Landau–Khalatnikov equation, power law, training effects

Received: September 25, 2023 Revised: December 23, 2023 Published online:

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