Proximity-enhanced magnetocaloric effect in ferromagnetic trilayers

M Persson*, M M Kulyk1, A F Kravets2 and V Korenivski3

Nanostructure Physics, Royal Institute of Technology, 10691 Stockholm, Sweden

E-mail: miltonp@kth.se

Received 13 September 2022, revised 17 October 2022
Accepted for publication 1 November 2022
Published 14 December 2022

Abstract

The demagnetization and associated magnetocaloric effect (MCE) in strong-weak-strong ferromagnetic trilayers, upon a reorientation of the strong ferromagnets from parallel to antiparallel (AP) magnetization, is simulated using atomistic spin dynamics. The simulations yield non-trivial spin distributions in the AP state, which in turn allows entropy to be calculated directly. The influence of longer-range spin–spin interactions and of variable strength of the external switching field are investigated. Finally, we find that the MCE in the system can be significantly improved by allowing the local exchange to vary through the spacer, which in practice can be implemented by spatially tailoring the spacer’s magnetic dilution.

Keywords: magnetocaloric effect, magnetic multilayers, atomistic spin dynamics, mean field entropy

(Some figures may appear in colour only in the online journal)

1. Introduction

The magnetocaloric effect (MCE) relates to the energy exchange between the phonon and magnon subsystems in a material, manifesting as a decrease in temperature upon demagnetization and, correspondingly, an increase in temperature upon magnetization. The effect was first observed in nickel by Weiss and Piccard in 1917 [1, 2] and is studied today for cooling applications where it offers more efficiency [3, 4] and can be more environmentally friendly than today’s vapor-compression technology, with greater systems’ longevity and without high global warming potential refrigerants [5]. Many exotic materials exhibiting giant MCE have been found and studied, but when used in their bulk form the fields required to produce substantial values of MCE are well above 1 T [6]. With cooling of electronics being one of the most interesting application, it is vital to reduce these operating fields, and the search has turned to novel nanostructured materials [7]. These may additionally reduce or eliminate the need for rare-earth metals, which are environmentally unfriendly [8].

In a nanostructured trilayer consisting of one free and one pinned ferromagnetic layer, separated by a weakly ferromagnetic spacer, a field of the order of 10 mT is sufficient to switch the outer ferromagnetic layers from parallel (P) to antiparallel (AP) alignment and thereby reduce the effective exchange field in the spacer, which in turn leads to its demagnetization [9, 10]. Such trilayer systems have been rather intensively studied recently, both in experiments and theoretically [11–19]. As regards MCE, first discussed by Fraerman and Shereshevskii [13], these studies have mostly been indirect experimentally (via magnetometry) and phenomenological in terms of modeling [13, 15–19], which introduces model-dependent uncertainties in the conclusions drawn.

In this article, the above trilayer system is simulated using atomistic spin dynamics and the magnetic entropy (MCE) is computed directly from the resulting atomic spin distribution at various temperatures. The results are in good qualitative

---

1 Also at Institute of Physics, NASU, 03028 Kyiv, Ukraine.
2 Also at Institute of Magnetism, NASU, 03142 Kyiv, Ukraine.
* Author to whom any correspondence should be addressed.

Original Content from this work may be used under the terms of the Creative Commons Attribution 4.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
agreement with direct MCE measurements on a prototype system [20]. In addition and quite informatively, our atomistic simulations reveal the profile of the magnetization inside the spacer. In the limit of hard outer ferromagnets these profiles differ qualitatively from the functional forms used in the literature. Finally, possible nanostructure optimizations are investigated by allowing the exchange to vary through the spacer (in practice via tailoring the concentration of magnetic material), and a significant improvement is found for a specific gradient-spacer design.

2. System description

The system under study is a trilayer F/f/F with two strongly ferromagnetic layers, F, surrounding a weakly ferromagnetic spacer, f. One of the strongly ferromagnetic layers is pinned, by intrinsic anisotropy or exchange, while the other is free, so that an external magnetic field can oppose the effect of exchange coupling through the spacer and produce an AP configuration. The idea is that the magnetization of the spacer should be higher (ideally much higher) in the P as compared to the AP configuration, so that a significant change in the magnetization could be produced when switching between the two states (P-to-AP and AP-to-P). Furthermore, while for bulk MCE the external field competes directly with thermal disorder and therefore has to be very large, a small field is sufficient to switch the strongly ferromagnetic layers in F/f/F, which is then effectively amplified by the intrinsic exchange competing with thermal disorder within the spacer.

Atomistic spin dynamics simulations are carried out using the VAMPIRE [21] software package, based on the Landau–Lifshitz–Gilbert (LLG) equation with Langevin Dynamics. The simulated system is a BCC crystal with 28 × 100 × 100 unit cells, periodic exchange in the lateral directions, and the iron lattice parameter of a = 2.87 Å. This translates to roughly 8 × 29 × 29 nm³ with 1 nm for each of the strong ferromagnetic layers and 6 nm for the weak ferromagnetic spacer; see figure 1. Magnus et al [14] found that long-range spin–spin interactions need to be taken into account in order to properly reproduce the experimentally observed coupling through thick paramagnetic or weakly ferromagnetic spacers. Here, we test this premise for MCE by comparing the results of our atomistic simulations with the 1st, 2nd, 3rd, and up to 4th nearest neighbors included in the spin–spin exchange, to see if longer range interactions yield a qualitative difference in interpretation. Similar to [14] we take the longer-range exchange as decaying algebraically [22]:

\[ J_{\text{thnn}} = \left( \frac{J_{\text{1stnn}}}{\sigma} \right)^{d+\sigma} J_{\text{1stnn}}, \]

with \( d = 3 \) and \( \sigma = 0.5 \). To study the effect of only the range of the spin–spin interactions, the values of the exchange terms are rescaled so that the total effective exchange field, when all spins are parallel, remains the same. The respective scaling factor, \( X_n \), for interactions including up to the \( n \)th nearest neighbors, satisfies

\[ J_{\text{1stonly}} N_{\text{thnn}} = X_n \sum_{j=1}^{n} J_{\text{jth}} N_{\text{jth}}, \]

where \( N_{\text{th}} \) is the \( n \)th coordination number. For the interface exchange between the F and f materials, with the internal exchange values equal to \( J_1 \) and \( J_2 \), the harmonic mean was used [23]:

\[ J_{1+2} = \frac{2 J_1 J_2}{J_1 + J_2}. \]

Deviations from this mean value of the order of 1% were forced by the rescaling (2). Strong easy-plane anisotropy was used for all layers to emulate the demagnetizing fields in the thin film geometry. The time step used is 1.0 fs which should be adequate for our system with a relatively weak effective exchange in the spacer, even in the vicinity of the Curie temperature, since the resulting spin precession is not exceedingly fast [21]. Furthermore, the spin precession is slowed down and the relaxation sped up by the use of critical damping—setting the phenomenological Gilbert damping constant to 1 in the LLG equation [24].

3. Results

3.1. MCE versus temperature

Before considering realistic external fields for the P-to-AP switching, we investigate the maximum possible MCE in the system by using large intrinsic anisotropy to pin the outer layers P or AP at all temperatures. The first nearest-neighbor exchange is set to \( J_{\text{1st}} = 7.05 \times 10^{-21} \text{J/lnm} \) in the outer ferromagnetic layers for iron [21], and the spacer is given 15% of this value to match the experimentally known \( T_C \) for Fe₈₀Cr₂₀ spacers [20, 25, 26]. The atomic moments are then set to the iron value of 2.22 \( \mu_B \) in the outer layers and 30% of this, 0.666 \( \mu_B \), in the spacer.

Figure 1. Multilayered system under study with two 1 nm thick outer strongly ferromagnetic layers separated by a 6 nm thick weakly ferromagnetic spacer. Arrows show simulated spatial distribution of normalized magnetic moment vectors of each monolayer in the antiparallel F-F magnetization state at a temperature close to \( T_C \). Upper black curve shows magnitude of magnetic moments, while similar curves on the left and bottom show projections onto (x, y) and (y, z) planes, respectively.
The system is relaxed at different temperatures from 0 K to well above the Curie temperature of the spacer (200–214 K, depending on the interaction range), to find the maximum magnetization difference between the P and AP states. The temperature change is sequential with a step size of ∆T = 2 K and at each temperature the system is equilibrated for 20 ps, followed by 50 ps of averaging. An example of the simulated magnetization profile for the AP state is illustrated in figure 1 and the spacer’s magnetization versus temperature is shown in figures 2(a) and (b). This magnetization is not a projection as one would obtain in magnetometry, nor is it the magnitude of the total magnetization vector of the spacer. Instead it is the order parameter, m, extracted by taking the average normalized magnetization of all monolayers in the spacer.

The magnetization of the spacer in the P and AP states versus temperature follow one another quite closely until the AP magnetization abruptly drops approximately 10 K below the Curie transition in the P state. The longer exchange range can at this point only be seen to have one clear effect in that it increases the effective Curie temperature. This effect is only quantitative and does not change the overall behavior.

The magnetization difference on P to AP switching, ∆m, is shown in figure 3 as a function of temperature (as −∆m), for the same four cases of the spin–spin exchange range. In order to translate ∆m into a difference in entropy, the magnetization in each monolayer is considered before using the additive property of entropy, ∆S = \sum_i S(m_{i,n}) − S(m_{i,0}). For the entropy of a (2J + 1)-state ferromagnet, where J is the total angular momentum, a mean field model was presented by Tuszyński and Wierzbicki [27], in which

\[
\frac{S_J(m)}{Nk_B} = \ln \frac{\sinh[(2J+1)x]}{\sinh(x)} + 2Jm,
\]

where x = g_{SLJ}\mu_B\nu/2, proportional to the Lagrange coefficient \nu, is found by numerically solving the equation

\[
2Jm = \coth(x) - (2J+1)\coth[(2J+1)x].
\]

The above model is used with S = 3/2, L = 0, J = S + L, and g_{SLJ} = 1.478 for low temperature bulk iron [28], and when converting from entropy per-atom to per-mass of the active material, the spacer is considered to have 30% magnetic (Fe) atoms, see figure 3. With −∆m and ∆S plotted on the left and right axis, respectively, a difference in the shape of the curves is easily seen. ∆S is above −∆m at low temperature and below it at high temperatures, since a given change in magnetization causes a larger entropy change near saturation compared to that close to zero magnetization. The corresponding S-vs-m relation is shown in the inset. A consequence is that the peak in ∆S is at slightly lower temperature than the peak in −∆m.

Regarding the effect of the exchange range, it may appear that the peaks in figure 3 become wider (quantified via the full width at half maximum (FWHM)) as the Tc increases, however the FWHM values are affected by this shift in temperature. After rescaling the temperature axes such that the four entropy peaks align, the widths become 11.3, 11.6, 12.1, and 10.9 K, with no clear trend present. Vertical rescaling of (∆S) does not affect the FWHM, but if a peak is truncated, then the FWHM is increased. This is why the FWHM value for the 3rd n.n. case sticks out—the 2 K increments of the temperature sweep ‘missed’ the respective peak somewhat.

The effects of increasing the spin–spin interaction range up to 4th n.n. are rather intuitive and found above to be relatively weak, such that the qualitative behavior of the system, when it relates to MCE, does not change. In order to reduce the computational complexity of the atomistic modeling of the large phase space of the studied multilayer, in what
follows the spin–spin interaction range is restricted to the 1st nearest neighbors. Long-range exchange, going far beyond the 4th n.n., and its relevance for MCE, calls for a somewhat less rigorous than spin-atomistics, but computationally much more compact phenomenological numerical model to be discussed elsewhere [20].

3.1.1. Magnetization profiles. Looking closely at the AP-state magnetization profiles at the temperatures of maximum MCE, $T^*$, shown in panels (c)–(f) of figure 2, one can see a cusp-like shape, being slightly concave on either side of the center. This is not predicted by the linear [13] or hyperbolic [10, 19] functions $m(z)$ commonly used in the literature. Better fitting functional forms, derived by taking into consideration the domain wall present at low temperatures, are presented and discussed in the appendix. As the next section reveals, however, this profile is affected by the strong pinning of the outer layers used to study the maximum possible MCE.

3.2. Switching with fields

In order to model the experiment [20], the pinning anisotropy in the free layer is reduced to a more realistic 55 mT (5.65 × 10⁻²³ J/atomic-volume) for iron [21] and an external field is applied to the free layer to achieve the AP state (even though the field may not be strong enough to turn the outer layers fully AP at the lowest temperatures). Three different field strengths are tested: 1 T, 0.5 T, and 0.25 T (lower reversing fields would be appropriate for thicker free layers).

The $m$-$T$ and the magnetization profiles for the pinned case and the three fields are shown in figure 4. The peak of MCE is found at progressively higher temperatures ($T^*$) as the applied field is decreased in strength, which is natural since additional thermal softening is needed to allow rotation of the free layer (toward the AP state) in an weaker field. The increase is not large, however, because $T^*$ is limited from above by the $T_C$ of the spacer in the P state. A moderate decrease in $\Delta m$ (P-vs.-AP) is revealed by the magnetization profiles as the reversing field strength is reduced. Additionally, a qualitative difference in the shape of the AP profiles is seen in the data, with the cusp-like shape for the pinned case (discussed in the appendix) being progressively replaced by a fully concave shape that is more similar to what is commonly used in the literature [10, 19].

The decrease in $\Delta m$ and the upper limit on $T^*$ is seen more clearly in figure 5 where $-\Delta m(T)$ and $\Delta S(T)$ are plotted. Another significant difference is the complete absence of MCE at low temperatures for the weak fields. The explanation for this can be seen in figure 6 where the internal magnetization is shown at low temperatures and around $T^*$. The Bloch-type domain wall present at all temperatures $T \lesssim T^*$ in the pinned case, cannot be created by the weaker fields before the temperature has increased sufficiently. The mechanism of the exchange-enhanced MCE is modified for lower reversing fields, with a gradual rotation of the free layer into an incomplete AP-state (as when an increasing field is applied at fixed temperature). The result is a more gradual ‘collapse’ of the Bloch-type domain wall in the spacer, compared to the ideal hard-pinned case.

Even with a 0.25 T external field in a 1 nm thick free layer, the reduction in MCE is no more than 41% of the absolute maximum. Smaller fields are problematic in simulations as a less well-defined energy minimum leads to increased superparamagnetic behavior. In fact, for 0.25 T at 132 K (figure 6(d), light blue) $m_z < 0$ but was multiplied by $-1$ for clarity. It is clear, however, that the use of realistic external fields must be included in the investigation for a complete picture of the system’s MCE. It should also be pointed out, that the approximation of neglecting the direct effect of the external field on the spacers magnetization, is better justified at lower fields.
in \( \text{JK}^{-1} \text{kg}^{-1} \)-active-material. These quantitative changes are due to the fact that it is more difficult (costs more exchange energy) for a smaller number of monolayers in the spacer to support a full domain wall. Additionally, the proximity effect is greater for a thinner spacer, which increases the spacer’s effective \( T_C \) in the P state. The rather large increase in the FWHM is expected only in the pinned case, as the outer layers would stay parallel at low temperatures in realistic fields; see figures 5 and 6. Nevertheless, the effect of the gradient magnetic dilution within the spacer is expected to be qualitatively the same for 3 and 6 nm spacers.

At each point \((J_{\text{in}}, J_{\text{out}})\) a temperature sweep is performed in order to find the maximum entropy change, which represents the spacer in the resulting map \( \Delta S(J_{\text{in}}, J_{\text{out}}) \). For the calculation of the entropy at any point \((J_{\text{in}}, J_{\text{out}})\) one must have not just the order parameter \( m = m(z) \), but also the saturation magnetization, which should depend on the exchange values since they represent concentration of magnetic material. The relations between the concentration, saturation magnetization, and Curie temperature have been well studied for alloys such as FeCr, and could be used to find \( M_s(J_{\text{in}}, J_{\text{out}}; z) \). However, this would make the results less general, and, for consistency and simplicity, a linear approximation is used: 15% exchange corresponds to 30% iron atoms, as for the system above with the uniform 6 nm spacer, and 100% exchange corresponds to 100% iron atoms. The saturation magnetization used for the calculation of entropy, is then set to be proportional to the magnetic concentration, just as the atomic moments are in the simulations.

We start by investigating the maximum possible MCE, by hard-pinning the outer ferromagnetic layers. The resulting map \( \Delta S(J_{\text{in}}, J_{\text{out}}) \) can be seen in figure 7(a), and the largest values lie just below the line \( J_{\text{in}}/J_{\text{out}} = 1 \), i.e. the best spacer is nearly uniform. However, the external fields required to saturate \( \Delta S \) to these values may vary with the exchange values. Looking at the magnetization profiles, in figure 7(a), it is clear that the magnetization is larger in spacers with \( J_{\text{in}}/J_{\text{out}} < 1 \) and that the domain wall is concentrated to a smaller region than in spacers with \( J_{\text{in}}/J_{\text{out}} > 1 \). Smaller fields should therefore be required by the latter. To investigate this, the anisotropy in the free layer is reduced, as before, together with the introduction of an external field.

With a field of 1 T, there is a clear change in the optimal ratio \( J_{\text{in}}/J_{\text{out}} \) in favor of the original idea of lifting up the magnetization in the center of the spacer, i.e. \( J_{\text{in}}/J_{\text{out}} > 1 \); see figure 7(b). The largest \( \Delta S \) is at \( (20\%, 17\%) \) with a 13% improvement compared to the ‘equivalent’ uniform spacer at \( (18\%, 18\%) \), which has the same amount of Fe, or the same average exchange, \( (2 \cdot 17 + 20)/3 = 18 \). The change in the \( \Delta S \) surface suggests that 1 T is below the saturation field for \((J_{\text{in}}, J_{\text{out}})\) that are above the ridge of maximum \( \Delta S \), but still above the saturation field for points below this ridge. Therefore, the field is further reduced by 2 and 4 times, to 0.5 T and 0.25 T; figure 7(b). At 0.5 T, the largest \( \Delta S \) is observed at \( (20\%, 15\%) \) and is 23% greater than for the equivalent uniform spacer, while for 0.25 T these numbers are \( (19\%, 13\%) \) and 72%—a significant enhancement of nearly a factor of two. The non-uniformity \( J_{\text{in}}/J_{\text{out}} \), and the improvement in using

![Figure 6. Internal magnetization for the pinned case (a) and three different fields (b)–(d). The colored curves trace the monolayers’ magnetic moments, represented by gray lines. The external field is directed along the negative x direction.](image-url)
non-uniform spacers $\Delta S_{\text{max}}/\Delta S_{\text{unif}}$, are plotted in figure 7(c). Furthermore, the improvement of 72% is not just an improvement at 0.25T, but a significant improvement overall in terms of MCE per unit field, as seen on the right axis of figure 7(c).

Units of entropy per field are not meant to imply that $\Delta S$ scales linearly with the field, but if a smaller field can reorient the ferromagnetic layers, then more re-orientations can be performed per unit time, and $(\Delta S/\Delta t)/(\Delta H/\Delta t) = \Delta S/\Delta H$. The 0.25T switching can be performed at four times the frequency compared to the 1T switching, with an alternating field of equal T/s-magnitude. This is important as high-amplitude alternating fields are difficult to arrange in device applications and problematic in electronics.

For large ratios $J_{\text{in}}/J_{\text{out}}$, the maximum $\Delta m$ occurs when the magnetization is near zero in the outer sections of the spacer, and the difference is concentrated in the central section. Most of the domain wall will then be located in the outer sections, while the center is more uniform. This is problematic as it means that an external field, which on the experiment is not localized to the free layer, could have a significant effect on the central magnetization in the AP state, and thereby counteract the MCE. Using a thicker free layer would reduce this problem and increase the MCE per unit field, but it would also reduce the fraction of active material (spacer vs total volume). Hence, the best way to further improve the system would be to look for more efficient exchange (magnetic dilution) profiles $J(z)$. For any ratio $J_{\text{in}}/J_{\text{out}}$, there should be an optimal shape of the profile $J(z)$ with $J(0) = J_{\text{in}}$ and $J(|z| = d/2) = J_{\text{out}}$.

4. Conclusions

Highly parallelized computation applied to atomistic spin dynamics, in contrast to algorithmically serial Monte-Carlo methods, makes it possible to quickly find the spatially resolved magnetization (microscopic spin distribution) in realistic and computationally rather large nanosystems. The systems’ entropy can be calculated in a straightforward way, directly from the spin distribution using a mean field model for ferromagnets with identical spins. Extending the exchange interaction range from the 1st to the 4th nearest neighbors results in quantitative changes of the simulated properties, with the overall qualitative picture intact. Much longer range spin–spin interactions have been suggested by some of the experiments for the investigated metallic F/f/F system, where the conduction band is polarized and can mediate exchange over several nanometers, in addition to the more localized lattice-exchange. Such long range interactions, however, are computationally too heavy for spin-atomistic numerics and call for specialized phenomenological modeling.

The simulated magnetization profiles in the P and AP states are studied in detail. While the P state is well described by hyperbolic cosines, frequently used in the literature, it is found that the profiles of the AP state depend qualitatively on the strength of the external field used to switch the free outer layer. More specifically, when pinning the outer layers AP at all temperatures to study the maximum possible MCE in the system, it is found that the profiles of the AP state cannot be

![Figure 7](image-url)
described by the linear functions or hyperbolic sines used in the literature. Different functional forms are found by taking into consideration the domain wall present at low temperatures. More importantly, however, it becomes clear that realistic external fields must be used in order to obtain an agreement with experiments.

Finally, the effects of using a nonuniform spacer are studied, with the exchange, or concentration of magnetic material, allowed to vary through the spacer. With the spacer divided into three equal sections it is found that the entropy change per unit field is significantly improved when the inner section is given a larger exchange than the outer sections. Further improvements should be possible with more sophisticated exchange (concentration) profiles.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Acknowledgments

Support from the Swedish Research Council (VR 2018-03526) and the Olle Engkvist Foundation (Project 2020-2022) are gratefully acknowledged. The computations were enabled by resources provided by the Swedish National Infrastructure for Computing (SNIC), partially funded by the Swedish Research Council through Grant Agreement No. 2018-05973. We thank Lars Viklund (HPC2N) and Tor Kjellsson (PDC) at SNIC for their assistance with installation, use and modification of software, which was made possible through application support provided by SNIC.

Appendix

Magnetization profiles

Different functional forms have been used to describe the profile of magnetization through the spacer in F/F/F tri-layers, including linear functions and hyperbolic sines and cosines, whose parameters are found by minimizing the free energy [10, 13, 19]. In these cases, the spacer was assumed paramagnetic and its proximity-induced magnetization to be collinear with the magnetizations of the outer layers, in both the P and AP states. The hyperbolic-cosine form employed for the P state, fits our numerical results rather well. However, when using strong pinning of the outer layers to investigate the maximum possible MCE in the system, the magnetization profiles of the AP state differ qualitatively from hyperbolic sines, as shown in figure 8. Around the peak in magnetic disorder, $\Delta S_{\text{max}}$, and at lower temperatures, $m(z)$ has a cusp in the center of the spacer corresponding to a local maximum in the derivative, $m'(z)$, instead of a minimum in $m'(z)$ seen with hyperbolic-sine profiles. The form of $m(z)$ in our simulations approaches a hyperbolic sine as the temperature is increased, but not before $\Delta m$ and $\Delta S$ (MCE) have essentially vanished. Better fitting expressions for $m_{\text{ap},x}(z)$ and $m_{\text{ap},y}(z)$ are presented here, followed by a minimization of the free energy in an attempt at finding the fitting parameters analytically.

With the outer layers pinned by strong anisotropy there is a Bloch type domain wall present in the AP state even at low temperatures, uniformly distributed across the spacer at 0 K. As the temperature increases and the magnetization magnitude in the center decreases due to thermal disorder, the domain wall shortens since it is less costly energetically in regions of lower magnetization. A shorter domain wall means a larger angle between the adjacent moments (monolayers’ magnetizations), which in turn means a lower effective exchange field and, hence, additional reduction in the domain wall length. This eventually leads to a ‘collapse’ of the domain wall and a peak in $-\Delta m$ and $\Delta S$. There is still a domain wall near the center of the spacer after this peak temperature, but it is much less pronounced as its baring magnetization essentially vanishes under high thermal agitation. These characteristics of the simulated data suggest that the magnetization in the AP state at a given temperature is better described by a product of the hyperbolic cosine of the P state at that same temperature, and the magnetization of a domain wall whose strength and length decreases with increasing temperature:

$$m_{\text{p}}(z) = m_{\text{p},s}(z) = \alpha_1 \cosh(\alpha_2 z) + \alpha_3,$$

$$m_{\text{ap},s}(z) = \begin{cases} -m_{\text{p}}(z), & z \leq -1/2, \\ m_{\text{p}}(z) \sin \frac{\pi z}{2}, & |z| < 1/2, \\ m_{\text{p}}(z), & z \geq 1/2, \end{cases}$$

$$m_{\text{ap},y}(z) = \begin{cases} 0, & |z| \geq 1/2, \\ Xm_{\text{p}}(z) \cos \frac{\pi z}{2}, & |z| < 1/2, \end{cases}$$

(A6)
a paramagnetic spacer that was considered in these articles. The problem of finding \( \Delta m \) is then reduced to finding \( m_{ap}(z) \), i.e. the parameters \( X \) and \( l \). For fixed values of \( X \) the domain wall length can be found by minimizing the exchange energy,

\[
F_{ex} = \int_0^{d/2} \left( \frac{dm}{dz} \right)^2 \, dz,
\]

where \( d \) is the spacer thickness and where no exchange parameter is included since it does not affect \( dF/dl = 0 \). Zeman energy need not be considered as no external field is applied within the spacer. Shape anisotropy is not a factor either since the functional form \( m(z) \) already restricts the magnetic moments to lie in the plane of the spacer. Specifics of exchange at the interfaces are excluded as they are not important, due to the sharp growth of the hyperbolic cosine function near the interfaces, which helps restrict the domain wall to the spacer.

Splitting integral \( (A7) \) in two, for the intervals \((0, l/2)\) and \((l/2, d/2)\), and taking the derivative with respect to \( l \) leads to, for a fixed \( X \in (0, 1) \),

\[
\frac{l}{2} m_p^2 \left( \frac{2}{l} \right) = 2 \int_0^{l/2} m_p^2(z) \left[ \sin^2 \left( \frac{\pi z}{l} \right) + X^2 \cos^2 \left( \frac{\pi z}{l} \right) \right] \, dz
+ (1 - X^2) \int_0^{l/2} \sin \left( \frac{\pi z}{l} \right) \left[ \frac{l}{2} m_p^2(z) + X m_p^2(z) \right] \, dz
\]

(\(A8\)) (which simplifies significantly in the case \( X = 1 \)). The equation can be solved numerically for \( l \), given \( X \) and \( l \), and the results are displayed in figure 10(b), denoted \( l_1 \). The \( X = m_{ap}(0)/m_p(0) \) ratio was obtained from the simulated spin profiles (non-trivial to obtain analytically). To achieve better agreement at low temperatures, a boundary term of the form \( c_b(m_1 - m_2) \) was added to the free energy [13], which, with a large enough coefficient \( c_b \), can represent the high cost of the domain wall extending into the magnetically hard outer \( F \)-layers. The improved results are displayed in figure 10(b), denoted \( l_2 \).

Although the above analysis gives the domain wall length, \( X \) needs to be known beforehand, and with this simple model for the free energy \( (F = F_{ex}) \) the surface \( F(X, l) \) is smallest at the boundary \( X = 0 \). The relation between \( X \) and \( l \) needs to be known both for \( l \) and \( X \) can be found by minimizing \( F \).

**ORCID iDs**

M Persson https://orcid.org/0000-0002-9310-6183

M M Kulyk https://orcid.org/0000-0001-7568-656X

A F Kravets https://orcid.org/0000-0001-8754-3152

V Korenivski https://orcid.org/0000-0003-2339-1692

**References**

[1] Weiss P and Piccard A 1917 Le phénomène magnétocalorique

\[ J. \text{Phys. Theor. Appl.} \textbf{7} \text{103–9} \]
[2] Smith A 2013 Who discovered the magnetocaloric effect? *Eur. Phys. J. H* **38** 507–17

[3] Zimm C, Jastrab A, Sternberg A, Pecharsky V, Gschneidner K, Osborne M and Anderson I 1998 Description and performance of a near-room temperature magnetic refrigerator *Advances in Cryogenic Engineering* (New York: Springer) pp 1759–66

[4] Tishin A M and Spichkin Y I 2003 *The Magnetocaloric Effect and Its Applications* (Bristol: IOP Publishing) p 380

[5] Metz B, Kuijpers L, Solomon S, Andersen S O, Davidson O R and Meyer L (IPCC Response Strategies Working Group, IPCC Working Group I and Technology and Economic Assessment Panel—UNEP) 2005 *Safeguarding the Ozone Layer and the Global Climate System* ed Intergovernmental Panel on Climate Change (Cambridge: Cambridge University Press)

[6] Ram N R, Prakash M, Naresh U, Kumar N S, Sarmash T S, Subbarao T, Kumar R J, Kumar G R and Naidu K C B 2018 Review on magnetocaloric effect and materials *PhD Thesis*

[7] Miller C W, Belyea D D and Kirby B J 2014 Magnetocaloric effect in nanoscale thin films and heterostructures *J. Vac. Sci. Technol.* **32** 040802

[8] Monfared B, Furborg R and Palm B 2014 Magnetic vs. vapor-compression household refrigerators: a preliminary comparative life cycle assessment *Int. J. Refrig.* **42** 69

[9] Kravets A F, Timoshevskii A N, Yanchitsky B Z, Bergmann M A, Buhler J, Andersson S and Korenivski V 2012 Temperature-controlled interlayer exchange coupling in strong/weak ferromagnetic multilayers: a thermomagnetic curie switch *Phys. Rev. B* **86** 21

[10] Kravets A F, Dzhezherya Y I, Tostolytkin A I, Kozak I M, Gryshchuk A, Savina V O, Pashchenko V A, Gnatchenko S L, Koop B and Korenivski V 2014 Synthetic ferrimagnets with thermomagnetic switching *Phys. Rev. B* **90** 10

[11] Kravets A F, Tostolytkin A I, Dzhezherya Y I, Polishchuk D M, Kozak I M and Korenivski V 2015 Spin dynamics in a curie-switch *J. Phys.: Condens. Matter* **27** 446003

[12] Kravets A F, Polishchuk D M, Dzhezherya Y I, Tostolytkin A L, Golub V O and Korenivski V 2016 Anisotropic magnetization relaxation in ferromagnetic multilayers with variable interlayer exchange coupling *Phys. Rev. B* **94** 6

[13] Fraerman A A and Shereshevskii I A 2015 Magnetocaloric effect in ferromagnet/paramagnet multilayer structures *JETP Lett.* **101** 9

[14] Magnus F, Brooks-Bartlett M E, Mouhah R, Procter R A, Andersson G, Hase T P A, Banks S T and Hjörvarsson B 2016 Long-range magnetic interactions and proximity effects in an amorphous exchange-spring magnet *Nat. Commun.* **7** 1

[15] Vdovichev S N, Polishchuk N I, Rodionov I D, Prudnikov V N, Chang J and Fraerman A A 2018 High magnetocaloric efficiency of aNiFe/NiCu/CoFe/MnIr multilayer in a small magnetic field *Phys. Rev. B* **98** 1

[16] Polushkin N, Fashenkin I, Fadeev L, Lã¼deranta E and Fraerman A 2019 Magnetic and magnetocaloric properties of pygdf/CoFe/IrMn stacks *J. Magn. Magn. Mater.* **491** 165601

[17] Kuznetsov M A, Pashenkin I Y, Polushkin N I, Sapozhnikov M V and Fraerman A A 2020 Magnetocaloric effect in exchange-coupled strong/weak/strong ferromagnet stacks *J. App. Phys.* **127** 183904

[18] Fraerman A A 2021 Exchange enhancement of the magnetocaloric effect in ferromagnetic nanostructures (brief review) *JETP Lett.* **113** 5

[19] Kuznetsov M A, Drovoskov A B and Fraerman A A 2021 Magnetocaloric effect in nanosystems based on ferromagnets with different curie temperatures *J. Exp. Theor. Phys.* **132** 1

[20] Kulyk M, Persson M, Polishchuk D and Korenivski V 2022 Magnetocaloric effect in multilayers studied by membrane-based calorimetry (arXiv:2208.07761)

[21] Evans R F L, Fan W J, Chureemart P, Ostler T A, Ellis M O A and Chantrell R W Atomic spin model simulations of magnetic nanomaterials *J. Phys.: Condens. Matter* **26** 10

[22] Fisher M E, keng Ma S and Nickel B G 1972 Critical exponents for long-range interactions *Phys. Rev. Lett.* **29** 917

[23] Vansteenekiste A, Leljaert J, Dvornik M, Helsen M, Garcia-Sanchez F and Waeysenberge B V 2014 The design and verification of MuMax3 *AIP Adv.* **4** 107133

[24] Gilbert T 2004 Classics in magnetics a phenomenological theory of damping in ferromagnetic materials *IEEE Trans. Magn.* **40** 3443–49

[25] Kumar B R and Kaul S 2015 Magnetic order-disorder phase transition in Cr$_{55}$Fe$_{25}$ thin films *J. Alloys Compd.* **652** 479–95

[26] Kumar B R and Kaul S 2018 Thickness as a control parameter for magnetocaloric effect in Cr$_{55−x}$Fe$_{25+x}$ $(x = 0, 5)$ thin films *J. Magn. Magn. Mater.* **460** 312–9

[27] Tuszyński J A and Wierzbicki W 1991 The mean field entropy for an ensemble of identical spins with arbitrary magnitude *Am. J. Phys.* **59** 555–61

[28] Köbler U, Englisch J, Hupe O and Hesse J 2003 Effective spin quantum numbers in iron, cobalt and nickel *Physica B* **339** 156–63

[29] Burke S K and Rainford B D 1983a The evolution of magnetic order in CrFe alloys. I. Antiferromagnetic alloys close to the critical concentration *J. Phys. F: Met. Phys.* **13** 441

[30] Burke S K, Cywinski R, Davis J R and Rainford B D 1983 The evolution of magnetic order in CrFe alloys. II. Onset of ferromagnetism *J. Phys. F: Met. Phys.* **13** 451

[31] Burke S K and Rainford B D 1983b The evolution of magnetic order in CrFe alloys. III. Ferromagnetism close to the critical concentration *J. Phys. F: Met. Phys.* **13** 471