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Qualitative insight and quantitative analysis of the effect of temperature on the coercivity of a magnetic system

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The temperature dependence of the response of a magnetic system to an applied field can be understood qualitatively by considering variations in the energy surface characterizing the system and estimated quantitatively with rate theory. In the system analysed here, Fe/Sm-Co spring magnet, the width of the hysteresis loop is reduced to a half when temperature is raised from 25 K to 300 K. This narrowing can be explained and reproduced quantitatively without invoking temperature dependence of model parameters as has typically been done in previous data analysis. The applied magnetic field lowers the energy barrier for reorientation of the magnetization but thermal activation brings the system over the barrier. A 2-dimensional representation of the energy surface is developed and used to gain insight into the transition mechanism and to demonstrate how the applied field alters the transition path. Our results show the importance of explicitly including the effect of thermal activation when interpreting experiments involving the manipulation of magnetic systems at finite temperature. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4942428]

The stability of magnetic states with respect to thermal fluctuations and external perturbations is an important problem in fundamental studies of magnetism and of critical importance in the design of nanoscale magnetic devices.1 A theoretical estimate of the thermal stability of magnetization in nanoscale structures as a function of size and shape could help design devices with unprecedented capacity and functionality. The preparation of a magnetic system in a particular state can be destroyed by thermally activated transitions to other available states, contributing to the temperature dependence of various properties of magnetic materials, including hysteresis loops.2–10 If the energy barrier separating stable states is large compared to thermal energy, the thermally activated transition become a rare event and special techniques are required for the long time scale spin dynamics simulations.5,11–13 According to statistical rate theories, the rate of a thermally activated, over-the-barrier transition typically follows Arrhenius law,14,15 which contains two parameters: an energy barrier for the transition and a pre-exponential factor. While several calculations of an energy barrier for a magnetic transition have been reported,3,4,16–30 only a few calculations of a pre-exponential factor for magnetic transitions involving many degrees of freedom have been presented.26,27,29 In this article, we analyze the effect of thermal activation on hysteresis loops using HTST where both the energy barrier and the pre-exponential factor are evaluated to obtain an estimate of the time scale of the transition.

Here, we focus on spring magnets which are formed by combining a soft and hard magnetic material. The combination of the large magnetization of the soft magnetic layer and the high coercitivity of the hard magnetic layer makes these systems suitable for building permanent magnets with a large energy product (magnetic saturation times coercivity).31–35 Such systems can have various...
practical applications and have, for example, been proposed as components of thermally assisted magnetic recording devices, see for example Ref. 36. Furthermore, spring magnets have become important test systems for the study of non-collinear magnetism and the way magnetic field can affect the ordering of magnetic moments at the atomic scale.

Fig. 1 shows experimental data reported for a thin film spring magnet consisting of a 20 nm layer of Sm-Co hard magnet and 20 nm Fe soft magnet. The width of the hysteresis loop is reduced to less than a half by increasing the temperature from 25 K to 300 K. The reduction of the measured width of the hysteresis loop was ascribed to a reduction in the anisotropy parameter of the model with temperature. Similar data and interpretation have been reported for various other systems, see for example Ref. 39.

In the analysis presented here, a 2-dimensional representation of the multidimensional energy surface is developed to illustrate the effect of the applied field and to explain how temperature affects the hysteresis loop. We, furthermore, show that harmonic transition state theory (HTST) for magnetic transitions combined with an efficient way to find the minimum energy path (MEP) of a transition can in a rather straightforward way be used to reproduce quantitatively the narrowing of the hysteresis loop using the same multilayer model as in Refs. 37 and 38 but without introducing a temperature dependence of the model parameters. While MEPS are independent of temperature, transition rates estimated from MEPS and HTST are temperature dependent as in the Arrhenius law.

When a magnetic field is applied to a spring magnet and increased gradually, a non-collinear spiral structure first develops in the soft magnetic material. The film is ideally thin enough to avoid grain boundaries in the normal direction and the system can return to the initial state if the field is removed. At a large enough field strength, however, the magnetization of the hard magnetic material is irreversibly switched, as illustrated in Fig. 1.

The hysteresis loop of a bilayer spring magnet can contain two characteristic jumps in the magnetization, as seen in Fig. 1. At a magnetic field denoted as \( H_{ex} \), the spiral magnetic structures starts to form in the soft magnetic layer (see insets in Fig. 1). As the field strength increases,
the magnetic moments in the spiral rotate further towards the direction of the applied field. Eventually, at a field of \( H_{irr} \), the magnetization of the hard magnetic layer is reversed. Between \( H_{irr} \) and \( H_{irr} \), the stretching of the spiral structure is reversible.

Measured hysteresis loops of spring magnets have been reproduced successfully using a multilayer model where each atomic layer in the film is represented with a magnetic moment vector.\(^{37}\) The energy per unit area of the system is given by a sum of terms representing exchange interaction between adjacent layers, easy-axis anisotropy in the plane of the atomic layers and interaction with an external field\(^{44,45}\)

\[
E = - \sum_{i=1}^{N-1} \frac{A_i}{d} \cos(\phi_i - \phi_{i+1}) - d \sum_{i=1}^{N} K_\beta \cos^2(\phi_i) - d \sum_{i=1}^{N} H M_\beta \cos(\phi_i - \phi_H)
\]

Here, \( \phi_i \) and \( \phi_H \) the angles the magnetic moment vector in layer \( i \) and the external field, respectively, make with the anisotropy axis. In order to determine the state of the system at a given field, the energy is minimized with respect to the \( \phi_i \) for all layers. In Eq. (1), \( M_\beta \) and \( K_\beta \) are the magnetization and anisotropy constant, respectively, in the soft (\( \beta = s \)) and hard (\( \beta = h \)) magnet, \( d \) is the distance between atomic layers, and \( A_i \) the exchange coupling between adjacent layers in the soft magnet (\( \alpha = s \)), hard magnet (\( \alpha = h \)) and at the interface (\( \alpha = int \)). The values of these parameters for the hard magnet and the soft magnet as well as the coupling between the two were chosen to be the same as those used in previously reported fits to the experimental data taken at 25 K\(^{37}\) except for a 20\% increase in \( K_h \) to make the model consistent with zero temperature. Even at 25 K, thermal activation needs to be taken into account, and the fitted parameter values in Ref. 37 then implicitly contain the effect of temperature on \( H_{irr} \). We, however, explicitly include this temperature effect and need model parameters that correspond to zero temperature.

The model described by Eq. (1) defines an energy surface for the system as a function of the orientation of the magnetic moment vectors, the \( \phi_i \) variables. The non-collinear spiral state is a local minimum on this energy surface which can be found by minimizing the energy with respect to the \( \phi_i \) variables. When a significant field is applied, another deeper minimum corresponds to the collinear state where all the magnetic vectors are pointing in the direction of the field.

The energy of the system given by the multilayer model is a function of several hundred variables, one for each atomic layer. In order to visualize this as an energy surface, we use a reduced description of the model in terms of only two essential variables. This is accomplished by choosing a functional form for the orientation of the magnetic vectors that mimics a domain wall (DW)

\[
\phi_i = \phi(x_i) = \arctan \left( \frac{x_i - x_c}{w} \right) + \frac{\pi}{2}
\]

were, \( x_i \) is the location of layer \( i \), \( x_i = (i - 1)d \), while \( x_c \) is the location of the center and \( w \) the width of the DW. This functional form is consistent with detailed calculations of the MEP using the full set of variables (see below). It can also describe the collinear state by placing the center well beyond the hard magnet. With this model for the ordering of the magnetic moments, a contour graph of the energy surface can be constructed by inserting this expression for \( \phi_i \) into the expression for the energy of the system, Eq. (1). The resulting energy surface obtained for two values of the magnetic field, \( H = 0.3 \text{ T} \) and \( H = 0.6 \text{ T} \), is shown in Fig. 2. This visualization of the energy surface provides valuable insight into the mechanism of the transition.

An MEP for the transition between the two states is shown in Fig. 2. It reveals the mechanism of the transition and the displacement along the path defines a reaction coordinate. Starting from the local minimum corresponding to the spiral state, where the width of the DW is large, on the order of 25 layers, the progression along the MEP involves a gradual decrease in the width of the DW as its center moves towards the interface between the hard and soft magnets. After reaching the interface, the center of the DW remains there while the width decreases to ca. 5 atomic layers. From then on, a narrow DW moves within the hard magnet away from the interface. The maximum increase in energy along an MEP gives the activation energy for the transition. For
FIG. 2. Energy surface for the Fe/Sm-Co spring magnet corresponding to applied magnetic field of $H=0.3$ T (upper) and $H=0.6$ T (lower), constructed by assuming the magnetic moments of the atomic layers are oriented as described by Eqn. (2), mimicking a domain wall with width $w$ centered at $x_c$, and using the multilayer model of Eq. (1). The spiral state corresponds to the energy minimum to the right while the homogeneous, collinear state with parallel magnetic vectors corresponds to a minimum beyond the upper left corner. Discretization points of the minimum energy path are shown by filled circles. The saddle points are identified with a X. This graph illustrates how the magnetic field deforms the energy surface, destabilizing the spring state and decreasing the activation energy for the transition, whereby the thermal excitation needed for the transition becomes smaller. The inset shows calculated values of $H_{irr}$ as a function of temperature, as well as two experimental data points from the hysteresis loops in Fig. 1.

At a small field, this maximum (a saddle point on the energy surface) is within the hard magnet, but as the field is increased, it moves to the interface and stays there until the field reaches $H_{irr}$. This can explain the experimentally observed sensitivity of the hysteresis width to the interface quality.50
As the field strength is increased, the local minimum corresponding to the spiral state becomes shallower and the activation energy for the transition from the spiral state to the collinear state decreases. This is the primary reason for the narrowing of the hysteresis loop.

Results of calculations of the MEP for the full model (without the approximation in Eqn. (2)) using the geodesic nudged elastic band (GNEB) method, an extension of the NEB method to magnetic systems, are shown in Fig. 3. When a small field is applied, the energy of the initial, spiral state increases because the magnetization has a component pointing in a direction opposite to the field. For a stronger field, the energy of the spiral state decreases because more of the magnetic vectors in the soft magnet are aligned with the field leading to a magnetization component pointing in the direction of the field. The point of maximum energy along the MEP moves closer to the spiral state minimum and becomes lower as the field increases, consistent with the contour graphs in Fig. 2. The height of the barrier is essentially the energy cost of moving the DW to the interface. After the energy maximum has been reached, the energy changes linearly along the MEP as the center of the DW moves within the hard magnet. The width of the DW then remains unchanged (see Fig. 2) so the exchange energy is constant and the slopes of the linear segments in Fig. 3 are determined by the Zeeman energy. Since the magnetization of the system increases linearly with the reaction coordinate in this region, the energy changes linearly and the slope is larger for a stronger external field, a consequence of the stabilization of the collinear state by the applied field.

These results can now be used to estimate quantitatively how much the hysteresis loop narrows when the temperature is increased. The system will remain in the spiral state until the applied field has lowered the energy barrier sufficiently and, thereby, decreased the lifetime of the spiral state sufficiently for the transition to the collinear state to occur on the time scale of the experiment. This approach is similar to the finite temperature micromagnetic method which has been used to calculate angular and thickness dependence of the coercive field for graded perpendicular recording.
furthermore, give an estimate of the value of H
model parameters. A more detailed analysis including quantitative evaluation of the prefactor can,
ing of the hysteresis loop with temperature without any temperature dependent adjustment of the
and the multilayer model for the energy of the system, Eq. (1), can predict quantitatively the narrow-
experimental data shown in Fig. 1 (see inset). This simple analysis based only on basic assumptions
as temperature is raised from 25 K to 300 K in excellent agreement with the
of minimum energy paths (analogous to the ones shown in Fig. 3), H
irr
an implicit expression showing how H
irr
estimated from the observed H
irr
at 25 K. This analysis gives 2000 Å2,46 a value quite consistent with
atomic scale simulations of magnetization reversal in Fe islands where the width of the temporary
domain wall was found to be 30 Å.29 The area obtained here is roughly this length squared. This
gives an activation energy of E
irr
= 0.97 eV at 0.5 T and 0.13 eV at 1.3 T using saddle point energy
values from MEP calculations, as the ones shown in Fig. 3. Combined with the estimate of the
pre-exponential factor above, and assuming the time scale of the experiment is a second, H
irr
comes out to be 1.45 T at 25 K and 0.65 T at 300 K. These values are in close agreement with the experi-
mental measurements, as illustrated in the inset of Fig. 1. Therefore, the change in the hysteresis loop
with temperature can be described here solely in terms of thermal activation in the system.

The analysis and calculated results described above show how important it is to consider ther-
mally activated transitions in the analysis of the temperature dependence of magnetic hysteresis
loops. A spring magnet has been taken here as an example because it is a well defined system but
similar considerations apply more generally.

While there will be some change in the average magnetic moment, M, with temperature, which
will change the anisotropy energy,47,48 this effect is small unless the temperature is close to the Curie
temperature. For both Fe and SmCo the decrease is on the order of 3% as temperature is increased
from 25 K to 300 K.39,49 In order to reproduce the ca. factor of 1/2 narrowing of the hysteresis
loop observed experimentally (see Fig. 1) by modifying only the anisotropy constant, K, for the
hard magnet in Eq. (1), a decrease to nearly 1/3 would be needed, well beyond the magnitude that
a decrease in average magnetization would produce. The large narrowing of the hysteresis loop is of
fundamentally different origin, namely the increased rate of thermally activated transitions, as
explained above.

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