Thermally activated switching at long time scales in exchange-coupled magnetic grains

Ahmad M. Almudallal,1 J. I. Mercer,2 J. P. Whitehead,1 M. L. Plumer,1 J. van Ek,3 and T. J. Fal4

1Department of Physics and Physical Oceanography, Memorial University of Newfoundland, St. John’s, NL, A1B 3X7, Canada
2Department of Computer Science, Memorial University of Newfoundland, St. John’s, NL, A1B 3X7, Canada
3Western Digital Corporation, San Jose, California 95158, USA
4Department of Physics and Energy Science, University of Colorado Colorado Springs, CO, 80918, USA

(Dated: April 16, 2015)

A micromagnetic algorithm is presented for the treatment of thermally activated magnetic moment reversal of two exchange coupled grains over long time scales based on the Arrhenius-Néel relation and numerical solutions to the rate equation. The energy landscape and energy barriers are calculated numerically using a variant of the nudged elastic band method called the minimum energy path technique. This information is used in a formulation of the attempt frequency which depends on the energy path between minima. Applications are made to the calculation of magnetic hysteresis loops and compared with approximate forms of the energy landscape and $f$. An evaluation of the merits of exchange-coupled-composite magnetic recording media is made.

I. INTRODUCTION

After many decades, the Landau-Lifshitz-Gilbert (LLG) equation continues to provide the foundation for micromagnetic modeling of the dynamic evolution of granular magnetic material, with an increasing number of applications devoted to the study of the effects of thermal fluctuations.1,2 The LLG equation is commonly used to study granular recording media but it is limited to the study of phenomena over relatively short time scales (ms). Modern exchange-coupled composite (ECC) recording media is composed of high anisotropy ‘hard’ layer exchange coupled to one or more lower anisotropy ‘soft’ layers.3–9 One of the most important applications of micromagnetic modeling is the characterization of recording media through $MH$ hysteresis loops. Often, model parameters are determined by fitting results to experimental data. Inconveniently, experimental hysteresis loops typically require minutes to hours to complete, time scales that are outside the range of standard LLG simulations. In addition, the thermally activated decay of recorded bits requires a micromagnetic model that is valid over much longer times scales (years). Various scaling arguments, based on the Arrhenius-Néel law, have been proposed as a means to extrapolate LLG results to longer times which appear useful for older single-layer type recording media.10 Thermally activated processes in ECC media, however, are more complex and the simple scaling arguments appear to break down in this case.11

For the purpose of studying long-time scale micromagnetics that are governed by thermally activated processes, Kinetic Monte Carlo (KMC) methods have proven useful.12–16 In Ref. 17, a KMC algorithm to study long-time scale thermally activated grain reversal of single-layer recording media was described. The Arrhenius-Néel expression for the transition rates between the minimum energy states of the individual grains was used to calculate the time between successive reversals. The minimum energy states and the energy barriers separating them were calculated using a modified version of the Wood analytic expression for single Stoner-Wohlfarth particles18 (SWPs) which includes the effective exchange and magnetostatic fields from neighboring grains. For weakly interacting recording media, the effective field approximation appears valid. For the attempt frequency, the temperature and field dependent formula based on a single energy barrier given in Ref. 19 was used. This algorithm was subsequently used to study the magnetic $MH$ loops of high anisotropy magnetic recording media at both short and long time scales over a wide range of temperatures relevant to heat-assisted magnetic recording.20 Good agreement between the KMC results and those from LLG simulations at relatively short time scales was demonstrated.

In Ref. 21, this KMC algorithm was applied to study $MH$ loops of dual-layer ECC recording media at finite temperature and long time scales. In this application, the effect of ECC inter-layer exchange coupling was approximated through the effective field acting on its coupled neighbors, as with single-layer media. ECC media for practical applications has a relatively strong exchange coupling and it is not evident that treating the energy barriers in the Wood formulation is a good approximation6 for this purpose. In addition, the expression used for the attempt frequency is based on a single energy barrier, which is unlikely to be valid for multi-layer ECC media at relevant (moderate) coupling strengths. The absence of simple Arrhenius-Néel-type scaling between thermal and temporal effects for ECC media supports these conclusions.11

In this present work, a method is described to accurately treat the thermally activated reversal of two exchange-coupled grains at long time scales. This approach is based on using a modified version of the nudged elastic band (NEB) method to calculate the minimum energy paths (MEPs) between different energy minimum states22–24 which is used to determine energy barriers as well as a numerical formulation of the attempt frequency. The MEP method is implemented for two ECC coupled grains to calculate magnetic hysteresis loops over
a range of temperature and time scales and the results are validated by comparison with LLG simulations. This work compliments and extends previous studies of dual-grain-reversal energy landscapes \cite{3,4} and formulations of the dual-grain attempt frequency.\cite{5,6} It also serves as a prelude to our formulation and application of the combined MEP-KMC algorithm to study interacting N \times N \times 2 ECC thin films with all magnetostatic and intra-layer exchange interactions also included.\cite{7}

The remainder of this paper is organized as follows: In Sec. II, the NEB approach and our variant of it, the MEP, are described and our model of ECC media formulated. In Sec. III we show and discuss illustrative examples of calculated energy landscapes. In Sec. IV we describe the numerical calculation of the attempt frequency and MH hysteresis loops. Results are presented at various sweep rates and inter-layer coupling strengths. Approximations to the MEP are proposed and evaluated in Sec. V. In Sec. VI we compare MH loop results from our method with those obtained from (exact) LLG simulations at fast sweep rates. A figure of merit to assist in the evaluation of the benefits of ECC media is proposed in Sec. VII, and our conclusions are presented in Sec. VIII. Finally, some details of the formulation of the attempt frequency are provided in an appendix.

II. MINIMUM ENERGY PATH AND ECC MEDIA

The NEB method is a numerical algorithm for finding the energy landscape of a system with multiple energy minima. In the present case, we use a modified version of the NEB, the MEP, which follows the previously developed string method.\cite{22–24} The NEB connects two energy states by a chain of nodes that are influenced by a force field to help evenly space the nodes along the path. The MEP method optimizes the placement of nodes along the path, with a higher density of nodes as needed. The force field acting on the nodes is a sum over the perpendicular component of the potential force and the tangential component of the spring force connecting the nodes. We calculate the gradient of the energy at each node and the differences between the current node and its neighboring nodes, which will be taken as a multi-dimensional vector which is "parallel" to the path.\cite{22} The nodes are then moved along the component of the gradient which is "perpendicular" to the \( \hat{\tau} \) vector, except for end nodes which remain fixed. The expression for the perpendicular component of the gradient is,\cite{22}

\[
(\nabla V(n))_\perp = \nabla V(n) - (\nabla V(n) \cdot \hat{\tau})\hat{\tau},
\]

where \( \hat{\tau} \) is the unit vector tangential to the curve between neighboring nodes. The effect is that all of the nodes defining the energy path are shifted towards a valley in the energy landscape, but not necessarily towards an absolute minimum. After the nodes are moved, they are then evenly redistributed along the new path. This process is repeated until the perpendicular gradient is zero,\cite{22}

\[
(\nabla V(n))_\perp = 0.
\]  

ECC media consists of magnetic grains with different layers of varying anisotropy strength and moderate exchange interactions between these layers.\cite{3,4} The desired effect is to be able to use the very strong anisotropy of a hard layer to keep the grains thermally stable. The hard layer is then exchange coupled to a layer with lower anisotropy, the soft layer. The soft layer will respond more readily to a switching field and the exchange interaction will make switching the hard layer easier. The result is a thermally stable grain that can be reoriented by using an applied field at lower magnitudes than would be required if just the hard layer was used. In this work, we focus on only two coupled grains.

The soft grain has magnetization \( M_1 = 4 \times 10^5 \) A/m and anisotropy constant \( K_1 = 1.5 \times 10^6 \) J/m\(^3\), while the hard grain has \( M_2 = 5.4 \times 10^5 \) A/m and \( K_2 = 3.0 \times 10^5 \) J/m\(^3\). Grains are cubic with a side length \( a = 6 \) nm stacked along the \( z \) axis. For simplicity, and to be able to support the numerical results with analytic calculations, we study the system without magnetostatic interactions. The energy of a grain pair is therefore written in terms of the normalized magnetization vectors \( \hat{n}_i = M_i / M_i \),

\[
E = - K_1 v_1 (\hat{n}_1 \cdot \hat{\tau}_1)^2 - K_2 v_2 (\hat{n}_2 \cdot \hat{\tau}_2)^2 - I A (\hat{\dot{n}}_1 \cdot \hat{\dot{n}}_2) - \mu_0 \hat{H} \cdot (M_1 v_1 \hat{n}_1 + M_2 v_2 \hat{n}_2),
\]

where \( \hat{H} \) denotes the applied field and \( K_i, v_i, \hat{n}_i \) and \( M_i \) denote the anisotropy constant, volume, anisotropy axis and the saturation magnetization of the \( i^{th} \) grain, respectively. Grains are coupled through the exchange term with constant \( I \), and interfacial area \( A = a^2 \). It is then convenient to write this expression in spherical coordinates with \( \hat{n}_i || \hat{\tau} \) and the field \( \hat{H} \) also aligned along \( \hat{\tau} \), giving

\[
E = v \sum_{i=1}^2 (K_i z_i^2 + H M_i z_i)
\]

\[
- I A \left( z_1 z_2 + \cos(\phi_1 - \phi_2) \sqrt{(1 - z_1^2)(1 - z_2^2)} \right),
\]

where subscript \( v \) is the polar angle, \( v = v_1 = v_2, z_i = \cos \theta_i \). Note that the exchange interaction energy between grains can be derived from a more familiar form,

\[
E_{ex} = - J_{12} \hat{M}_1 \cdot \hat{M}_2,
\]

where \( J_{12} \) is the usual exchange coupling between magnetic moments and \( I = J_{12} M_1 M_2 / a \).

III. ENERGY LANDSCAPES

To implement the MEP method, we determine an initial path with a set of nodes spaced evenly between two
states. By calculating the gradient at each node and moving it downhill to lower energy, the node can then be brought to a state with the lowest local minimum, and therefore the whole initial path will be driven to settle along the MEP. The energy landscape for a single grain has two minimum energy states, one when M is parallel (↑) to H and another when M is anti-parallel (↓) to H, and a maximum (saddle point) when M is perpendicular to H. The difference between one of the energy minima and the maximum energy defines the energy barrier that a state needs to overcome to transform to another state. For two weakly coupled grains, the energy landscape has a more complex structure with four possible states, the two grains are parallel to H (↑↑), the top grain is parallel while the bottom one is antiparallel to H (↑↓), the top grain is antiparallel while the bottom one is parallel to H (↓↑), and the two grains are antiparallel to H (↓↓), and a unique saddle point located on the path between each pair of states. However, if the coupling is strong, the two states ↑↓, ↓↑ have high energy configurations and the relevant portion of the energy landscape is effectively reduced to a one with only ↑↑, ↓↓ states analogous to the single-grain case.

Fig. 1(a) shows the energy landscape of two strongly coupled grains (I=2×10^{-3} J/m^2) at H=0 Oe. The black solid line is the initial path and the red circles trace the location of the calculated MEP. From the color code in this graph, we can easily distinguish two equal energy minima and a maximum as also illustrated when plotting the energy along the path nodes in Fig. 1(b), where the black line is the initial path and the red line is the final path with minimum energy, which for this case are nearly indistinguishable. For a strong field (H= -4 kOe) in the z-direction, the minimum for ↓↓ becomes more stable in favour of ↑↑, as illustrated in Figs. 1(c) and 1(d).

For weak coupling (I=0.5×10^{-3} J/m^2) and at H=0 Oe, the energy landscape shows two stable minima and two shallower ones as in Fig. 2(a) with four possible MEPs A, B, C and D. This suggests two different scenarios for flipping the two grains from the ↑↑ state to the ↓↓ state. The first scenario is to flip the soft grain first to bring the system to the ↑↑ state and then to flip the hard grain to the ↓↓ state. This is indicated by following path A first then path B in Fig. 2(a); Fig. 2(b) shows the energy along path nodes of A (black is initial path and red is the MEP) and B (green is initial path and blue is the MEP). The second scenario is to flip the hard grain first to bring the system to the ↑↓ state and then to flip the soft grain, as indicated by paths C and D in Fig. 2(a) with the minimum energy along path nodes in Fig. 2(c). When applying strong field -4 kOe, the energy landscape dramatically changes as in Fig. 2(d), and the saddle point along path C disappears. This will necessarily remove the second scenario and keep the first. Fig. 2(e) corresponds to the initial and minimum energy along path nodes of A and B at H= -4 kOe. By comparison with results at H=0 Oe in Fig. 2(b), we notice a decrease in the energy barrier when going from state ↑↑ to state ↑↓. However, one can think of states ↑↑ and ↑↓ as being at equilibrium due to the small energy barrier; the same idea can also be applied to states ↓↓ and ↑↓ as shown in Fig. 2(f).

We note here that the energy landscapes and MEPs for the above H=0 cases are shown more clearly in Fig. 9 in App. A where their features relevant to a numerical determination of the attempt frequency are discussed in more detail.

IV. MH HYSTERESIS LOOPS

The importance of calculating the energy landscapes arises from the necessity of calculating the escape rate r_{ij} from an initial state i to a final state j, which can be described by the Arrhenius-Néel relation,^{12–16}

\[ r_{ij} = f_{ij} \exp(-E_{Bij}/k_B T), \]

where f_{ij} is the attempt frequency, and E_{Bij} is the energy barrier between states i and j. The escape rate itself is a function of temperature and applied field (a fact which is often ignored). This implies a change in the probability p of being in one of the four energy states. In general, the rate equations describing the evolution of p_i with time (and thus implicitly with field) of being in a state i can be written as,

\[ \frac{dp_i(t)}{dt} = - \sum_{j \neq i} (r_{ij} p_i(t) - r_{ji} p_j(t)). \]
of rate equations is four as there will be four minimum energy states.

The calculation of the attempt frequency is non-trivial in the case of two coupled grains. An outline of our derivation is given in Appendix A and follows the Langer/Fokker-Plank formulation. This leads to the expression

\[ f_{ij} = \frac{\alpha \gamma \kappa}{1 + \alpha^2 M_T v} \sqrt{\frac{1}{2\pi k_B T} \left| \lambda_1 \lambda_2 \lambda_3 \right|} \]

where \( M_T = M_1 + M_2 \), \( \lambda_i \) are the eigenvalues of the Hessian matrix \( \frac{\partial^2 E(x)}{\partial x_i \partial x_j} \) at the saddle point, with \( x_i \) corresponding to four spherical-coordinate angles for the two grains, and \( \eta_i \) are the eigenvalues at the minimum. The four eigenvalues of this matrix at each minimum are positive, while at the saddle point only two are positive. At the saddle point \( \lambda_3 = 0 \), which is not included in this expression. Also in this expression, \( \kappa \) is found by solving a related eigenvalue equation. This result reduces to the Brown formula in the case of a single grain. For two coupled grains, the energy barriers and eigenvalues are calculated here numerically from the MEP method as described above.

At constant field, the set of rate equations can be solved analytically to calculate \( p_i \). However, for time dependent fields, these equations require a numerical solution to calculate \( p_i(t(H)) \). Interpolation can be then used to find the \( MH \) hysteresis loops as a function of sweep rate \( R = dH/dt \) in units of Oe/s. The range of sweep rates chosen (also used below) corresponds approximately to time scales involved in experimental measurements \( R \sim 10^9 \text{ Oe/s} \), to recording rates, \( R \sim 10^{10} \text{ Oe/s} \). Fig. 3 shows the calculated \( MH \) loops at \( T=300 \text{ K} \) for different exchange coupling values \( I=2.0, 1.5, 1.0, 0.5, 0.25 \) and \( 0.1 \times 10^{-3} \text{ J/m}^2 \), respectively, and each \( MH \) loop is calculated at the different sweep rates but only the range \( (R = 10^9-10^{10} \text{ Oe/s}) \) shown in these figures.

From these results it can be seen that the general trend of the coercivity \( H_c \) decreasing at slower sweep rates, as expected. In addition, there is little difference between the strong coupling cases of \( I=2.0 \times 10^{-3} \text{ J/m}^2 \) and \( I=1.5 \times 10^{-3} \text{ J/m}^2 \). Moderate coupling \( I=1.0 \times 10^{-3} \text{ J/m}^2 \) and \( I=0.5 \times 10^{-3} \text{ J/m}^2 \) represents a crossover regime between the two grains acting as a single grain, and the two grains responding quasi-independently. Here the loops are quite sensitive to the coupling \( I \). Weak coupling is clear in the case of \( I = 0.1 \times 10^{-3} \text{ J/m}^2 \) at the fast sweep rate where the plateau indicates that the soft grain switches first.

![Fig. 2: The energy landscape](image)

**Fig. 2:** The energy landscape (a) for weak coupling \( I=0.5 \times 10^{-3} \text{ J/m}^2 \) at \( H=0 \text{ Oe} \) with four minimum paths between the four possible energy states, (b) shows the initial and minimum energies along A and B path nodes, and (c) for the initial and minimum energies along C and D path nodes. The energy landscape for \( H=-4 \text{ kOe} \) is shown in (d), where (e) shows the initial and minimum energies along A and B path nodes, and (f) for the initial and minimum energies along the D path nodes.

![Fig. 3: MH hysteresis loops](image)

**Fig. 3:** \( MH \) hysteresis loops at \( T=300 \text{ K} \) and different sweep rates \( R \). (a) \( I=2.0 \times 10^{-3} \text{ J/m}^2 \), (b) \( I=1.5 \times 10^{-3} \text{ J/m}^2 \), (c) \( I=1.0 \times 10^{-3} \text{ J/m}^2 \), (d) \( I=0.5 \times 10^{-3} \text{ J/m}^2 \), (e) \( I=0.25 \times 10^{-3} \text{ J/m}^2 \), and (f) \( I=0.1 \times 10^{-3} \text{ J/m}^2 \).

From the hysteresis loops, we can extract the nucleation field \( H_n = H(M/M_s = 0.95) \), the coercive...
field $H_c = H(M/M_s = 0.0)$, and the saturation field $H_s = H(M/M_s = -0.95)$. Fig. 4 shows these extracted values of $H_n$, $H_c$, and $H_s$ as a function of $I$ for different sweep rates. Although the nucleation field exhibits monotonic decrease with increasing $I$ and $R$, both $H_c$ and $H_s$ shows clear minima at weak to moderate coupling values in the cases of the faster sweep rates. This feature will appear later in the discussion of a figure of merit, Sec. VII.

\[ R = 10^x \text{ (Oe/s)} \]

\[ I (10^{-3} \text{ J/m}^2) \]

\[ H_n \text{ (kOe)} \]

\[ H_c \text{ (kOe)} \]

\[ H_s \text{ (kOe)} \]

\[ (a) \]

\[ (b) \]

\[ (c) \]

FIG. 4: (a) The nucleation field $H_n$, (b) coercivity $H_c$, and (c) saturation field $H_s$ extracted from Fig. 3 as a function of $I$ at different sweep rates.

V. APPROXIMATE MODELS

1. Brown approximation

The Brown expression for the attempt frequency was formulated for a single magnetic grain with uniaxial anisotropy in an externally applied field.\textsuperscript{2,17} We consider here using this approximate formula for the coupled-grain reversal. Within this approximation, we consider a direct path between each pair of minima, which may not be a path of minimum energy. For strong coupling this approximation may be valid since the two magnetic moments will flip coherently, i.e. $\theta_1 = \theta_2$ and $\phi_1 = \phi_2$. For weak coupling, we must consider the two possible scenarios for going from the $\uparrow\uparrow$ state to the $\downarrow\downarrow$ state. With the first scenario, the system falls first into the $\uparrow\downarrow$ state before ending at $\downarrow\downarrow$, and to use Brown approximation we consider $\theta_2 = 0$ while $\theta_1$ varies for path $A$, and $\theta_1 = \pi$ while $\theta_2$ varies for path $B$. On the other hand, the second scenario assumes that the system falls first into the $\uparrow\downarrow$ state, and in this case we consider $\theta_1 = 0$ while $\theta_2$ varies for path $C$ and $\theta_2 = \pi$ while $\theta_1$ varies for path $D$ (see Fig. 2). For the two scenarios, we always set $\phi_1 = \phi_2$. The attempt frequencies for Brown approximation between $i$ and $j$ states are,

\[ f_{ij} = \frac{\alpha \gamma}{1 + \alpha^2} \left( 1 - \frac{H^2}{H_K^2} \right) \left( H_K - H \right) \sqrt{\frac{K_T}{\pi k_B T}} \]  

\[ f_{ji} = \frac{\alpha \gamma}{1 + \alpha^2} \left( 1 - \frac{H^2}{H_K^2} \right) \left( H_K + H \right) \sqrt{\frac{K_T}{\pi k_B T}}, \]  

where $\alpha$ is the damping constant, $\gamma$ is the gyromagnetic ratio, $H_K = 2K_T/M_T$, $K_T = K_1 + K_2$, and $M_T = M_1 + M_2$. The escape rates can be expressed as,

\[ r_{ij} = f_{ij} \exp \left( -\frac{E_s - E_i}{k_B T} \right) \]  

\[ r_{ji} = f_{ji} \exp \left( -\frac{E_s - E_j}{k_B T} \right), \]

where $E_s$ is the energy at the saddle point. With these escape rates, a set of rate equations can be also solved to calculate $MH$ hysteresis loops. In Fig. 5, we show a comparison of loops calculated by the MEP method (solid) and Brown approximation (dashed) for $f_{12}$, with $\alpha = 0.1$, (a) $I=2.0\times10^{-3}$ J/m$^2$ and (b) $I=0.5\times10^{-3}$ J/m$^2$, both at different sweep rates. In Fig. 5(c), the coercive field is shown as a function of sweep rate for $I=2.0$, 0.5, and $0.1\times10^{-3}$ J/m$^2$ using both methods. As can be seen, the differences between the MEP (exact) formulation and this Brown approximation are generally small and dependent on $I$ and $R$.

2. Three-state and two-state models

For moderate and weak coupling and over a particular range of $H$, one of the four minima might either vanish or
FIG. 5: (a) shows the $MH$ loops at different sweep rates for $I=2.0 \times 10^{-3}$ J/m² and (b) for $I=0.5 \times 10^{-3}$ J/m². Solid lines are obtained from the MEP method and the dashed lines are from the Brown approximation. The extracted coercivity as a function of the sweep rate for $I=2.0, 0.5$, and $0.1 \times 10^{-3}$ J/m² is shown in (c).

become very shallow, as in the case for the energy state $\uparrow \downarrow$ in Fig. 2(f). Therefore, we can assume that the minimum is in thermal equilibrium with other stable minimum, and hence the system can be reduced from a four-state to a three-state model. If we consider the case in Fig. 2(f) as an example, then we can assume that $p_0 = p_3 + p_4$, where $p_3$ is the probability of being in state $\uparrow \downarrow$ and $p_4$ is the probability of being in state $\downarrow \downarrow$. In thermal equilibrium, the ratio $r_{34}/r_{43}$ will be equal to $p_4/p_3$, where $r_{34}$ is the escape rate from $\uparrow \downarrow$ state to $\downarrow \downarrow$ and $r_{43}$ is the escape rate from $\downarrow \downarrow$ to $\downarrow \uparrow$. With this approximation, the set of rate equations in Eq. 6 will be reduced from four equations to three as,

\[
\begin{align*}
\frac{dp_{1}}{dt} &= -(r_{12} + r_{13})p_{1} + r_{21}p_{2} + r_{01}p_{6} \\
\frac{dp_{2}}{dt} &= -(r_{21} + r_{24})p_{2} + r_{12}p_{1} + r_{02}p_{6} \\
\frac{dp_{6}}{dt} &= -(r_{b1} + r_{b2})p_{6} + r_{13}p_{1} + r_{24}p_{2},
\end{align*}
\]

where,

\[
\begin{align*}
r_{b1} &= \frac{r_{31}r_{43}}{r_{34} + r_{43}} , \\
r_{b2} &= \frac{r_{21}r_{43}}{r_{34} + r_{43}}.
\end{align*}
\]

In addition to thermal equilibration between states $\uparrow \downarrow$ and $\downarrow \downarrow$, states $\uparrow \uparrow$ and $\downarrow \uparrow$ can also be brought to equilibrium. The system in this case becomes a two-state model with the following rate equations,

\[
\begin{align*}
\frac{dp_{a}}{dt} &= -r_{ab}p_{a} + r_{ba}p_{b} \\
\frac{dp_{b}}{dt} &= -r_{ba}p_{b} + r_{ab}p_{a},
\end{align*}
\]

where $p_{a} = p_{1} + p_{2}$ and,

\[
\begin{align*}
r_{ab} &= \frac{r_{13}r_{21} + r_{12}r_{24}}{r_{12} + r_{21}}, \\
r_{ba} &= \frac{r_{31}r_{43} + r_{34}r_{42}}{r_{34} + r_{43}}.
\end{align*}
\]

Fig. 6(a) shows a comparison of the $MH$ loops for $I=0.5 \times 10^{-3}$ J/m² between the original four-state model (solid curves) and the three-state approximation (open circles) at different sweep rates. Fig. 6(b) shows the comparison between the four-state model and the two-state approximation (open circles). While the three-state model shows a very high agreement at any sweep rate, the two-state model shows agreement at only slow and moderate sweep rates ($R < 10^9$ Oe/s). The discrepancy of the two-state model from the four-state model at fast sweep rates can be understood by the fact that the two states $\uparrow \uparrow$ and $\downarrow \uparrow$ are not at thermal equilibrium at strong applied field ($H < -2$ kOe) unless $R$ is very large.

VI. COMPARISON WITH LLG

As in our previous implementations of the KMC method, we can verify the approach by comparing the calculated $MH$ hysteresis loops with those obtained from 'exact' finite temperature LLG simulations.\textsuperscript{17,20} This comparison can be made only at the faster sweep rates where the LLG results are accessible within a reasonable amount of simulation time. As noted previously, if the sweep rate is too large we expect the Arrhenius-Néel approach to break down as it does not capture the spin dynamics of the reversal process. For the LLG simulations, we calculate the $MH$ loops for an exchange coupled double-layer system consisting of $16 \times 16$ non-interacting grains and using the same parameters mentioned in Sec. II. The larger system size is required in order to achieve good
FIG. 6: (a) shows a comparison of the $M$-$H$ loops for $I=0.5 \times 10^{-3}$ J/m$^2$. Solid curves are obtained by the four-state model while open circles are by the three-state model. (b) shows the comparison between the four-state model and the two-state model.

statistics from this stochastic simulation. The time step used was 2 ps, the damping parameter set at $\alpha = 0.1$, and we perform the simulations at $T=300$ K. The comparison shown in Fig. 7(a) for $I=2.0 \times 10^{-3}$ J/m$^2$ and in Fig. 7(b) for $I=0.5 \times 10^{-3}$ J/m$^2$ indicates a very good agreement at all sweep rates.

VII. FIGURE OF MERIT FOR ECC MEDIA

The benefit of coupling hard and soft layers can be quantified in a figure of merit (FOM) which is the ratio of a measure of the thermal stability and the field required to switch the grain magnetizations. This can be defined as the ratio between the energy barrier (thermal stability) and saturation field (switching energy) at a particular sweep rate as,

$$FOM = \frac{E_B(R)}{H_a(M_1V_1 + M_2V_2)} \quad (14)$$

$E_B$ for strong coupling is the energy barrier between the minimum energy of state $\uparrow\uparrow$ and the saddle point along the path to the minimum energy of state $\downarrow\downarrow$, while for weak coupling $E_B$ is the energy barrier between the minimum energy of state $\uparrow\uparrow$ and the saddle point along the path to the minimum energy of state $\downarrow\downarrow$. A larger FOM is the goal for ECC-type media. The results shown in Fig. 4(c) indicate that increasing $I$ in the range $[0, I_{max}(R)]$ will decrease the saturation field which makes switching the magnetic moment easier. On the other hand, increasing $I$ will increase the energy barrier which enhances the thermal stability (results are not shown). Fig. 8 shows the FOM at three sweep rates ($R = 10^6$, $10^8$, and $10^{10}$ Oe/s), and the optimal value of $I$ can be easily obtained from the graph: $I_{op}(10^6\text{ Oe/s})=0.2 \times 10^{-3}$ J/m$^2$, $I_{op}(10^8\text{ Oe/s})=0.35 \times 10^{-3}$ J/m$^2$, and $I_{op}(10^{10}\text{ Oe/s})=0.50 \times 10^{-3}$ J/m$^2$. These results suggest that weak to moderate coupling is preferred and that there is a strong dependence on sweep rate. Large FOM values at smaller $R$ may not be realized at larger sweep rates, and optimal coupling strengths estimated on the basis of experimental $MH$ loops obtained at slow sweep rates may thus not be the best value at recording time scales.

VIII. DISCUSSION AND CONCLUSIONS

This work represents an extension to two coupled magnetic grains, the previously described algorithms that
treat thermally activated single-grain magnetic moment reversal in an applied field based on the Arrhenius-Néel relation. At the heart of the present method is the numerical calculation of the energy landscape for the two-grain problem, based on the string method (MEP). This facilitates a calculation of the attempt frequency which requires knowledge of the saddle points within a Langer formulation of the transition rates. The probability of transitions between energy minima are calculated using numerical solutions to the rate equations. This allows for the calculation of $MH$ hysteresis loops that depend on temperature and specified field sweep rates.

While strongly coupled grains provide a simple and direct solution for calculating the $MH$ hysteresis loops, more care has to be taken when dealing with the weakly coupled grains. Strong coupling of two grains results in coherent rotation with the applied field, and this in principle suggests having only two stable energy minima in the energy landscape, the first one for $\uparrow\uparrow$ and the second for $\downarrow\downarrow$. In the moderate-to-weak coupling cases, there are four distinct energy minima, two metastable minima ($\uparrow\downarrow$, $\downarrow\uparrow$) in addition to ($\uparrow\uparrow$, $\downarrow\downarrow$). In these cases, obtaining the $MH$ hysteresis loops requires solving two rate equations for strong coupling and four rate equations for the weak coupling. The results show that strong coupling occurs when $I \geq 1.5 \times 10^{-3}$ J/m².

Comparison of our results with the Brown single-grain analytic formula for the attempt frequency, which considers only a direct path between energy minima, shows remarkably good agreement with the exact MEP/Langer calculation. Some discrepancy is observed at low sweep rate for the small values of $I$ which is a consequence of the more complicated energy landscape in the weak coupling case.

For better understanding and faster calculations that can be useful in future applications in the simulations of interacting $N \times N \times 2$ models of thin films, we consider reducing the four energy minima (four-state model), for the weak coupling case, to three- and two-state models. These approximations arise for the fact that the two metastable minima ($\uparrow\downarrow$ and $\downarrow\uparrow$) are poorly populated and at some applied field values, a thermal equilibrium (very low energy barrier) occurs between one of these metastable minima and one of the other stable minima ($\uparrow\uparrow$ and $\downarrow\downarrow$). While the agreement seems to be excellent between the four-state and three-state models, a discrepancy is observed between the four-state and two-state model at fast sweep rates ($R > 10^8$ Oe/s). This discrepancy occurs because the energy barrier between the two energy states ($\uparrow\downarrow$ and $\uparrow\uparrow$) increases with the applied field and this will necessarily break the thermal equilibrium.

Verification of our model results for $MH$ hysteresis loop was achieved through comparison with LLG simulations on a dual-layer system, each layer with a $16 \times 16$ non-interacting grains. The high degree of agreement confirms the accuracy of our algorithm and its implementation.

The results of this work serve as a prelude to the extension of our previous KMC approach to study thermally activated magnetic grain reversal in dual-layer ECC media that includes magnetostatic and intra-layer exchange interactions. The essentially exact treatment of moment reversal for the two-grain problem as outlined in this work, serves as the foundation for this extension. This will allow for the direct comparison of experimentally determined slow-sweep-rate $MH$ loops for ECC media with corresponding modelled results. This capability is essential for the estimation of model parameters which characterize recording media such as anisotropy and exchange couplings. Such a direct comparison is not possible with traditional LLG simulations where long time scales are inaccessible. This dual-layer KMC algorithm will also be especially useful in applications to dual-layer media for heat assisted magnetic recording (HAMR) where thermally activated moment reversal is pronounced. In addition, the investigation of magneto-static and intra-layer interaction effects on the FOM of Fig. 8 is of particular interest.

IX. ACKNOWLEDGMENTS

This work was supported by Western Digital Corporation, the Natural Science and Engineering Research Council (NSERC) of Canada, the Canada Foundation for Innovation (CFI), and the Atlantic Computational Excellence network (ACEnet).

X. APPENDIX A: THERMALLY ACTIVATED GRAIN REVERSAL IN ECC MEDIA

In this section we consider the non-equilibrium relaxation process that describes thermally activated reversal for an ensemble of equivalent non-interacting ECC grains in an applied field. In order to simplify the following analysis, we consider the case where the applied field and the anisotropy axes are aligned along a common axis which we choose to be the $y$-axis. The energy expression Eq.
(3) then may be written in spherical coordinates as,
\[
E = -K_1 v_1 \sin^2 \theta_1 \sin^2 \phi_1 - K_2 v_2 \sin^2 \theta_2 \sin^2 \phi_2 - \mu_0 H_y (M_1 v_1 \sin \theta_1 \sin \phi_1 + M_2 v_2 \sin \theta_2 \sin \phi_2) - IA (\cos \theta_1 \cos \theta_2 + \cos (\phi_1 - \phi_2) \sin \theta_1 \sin \theta_2).
\]

The invariance of the energy under rotation about the y axis means that the energy of the grains will have a stationary (critical) point located at each of four sites \( \{m_{y1}, m_{y2}\} \in \{\pm 1, \pm 1\} \). While the above choice of coordinates makes for a more complicated energy expression than given in Eq. (4), it has the considerable advantage that metric tensor is non-singular in the neighborhood of these stationary points. Depending on the value of the applied field and parameters some or all of these stationary points will correspond to local minima in the energy. We denote these stationary points that correspond to local minimum energy states as \( \sigma_1 \to \{1, 1\}, \sigma_2 \to \{-1, 1\}, \sigma_3 \to \{1, -1\}, \) and \( \sigma_4 \to \{-1, -1\} \) respectively.

In Fig. 9 we present surface plots of the energy of two grains in the subspace \( \theta_1 = \theta_2 = \pi/2 \) as a function of \( \phi_1 \) and \( \phi_2 \) for \( I=2.0 \times 10^{-3} \text{ J/m}^2 \) and \( I=0.5 \times 10^{-3} \text{ J/m}^2 \) respectively, at \( H = 0 \). This corresponds to the case also shown in Figs. 1(a) and 2(a), but with more detail to illustrate the concepts of this analysis. The energy landscape in Fig. 9(a) for the case \( I=2.0 \times 10^{-3} \text{ J/m}^2 \) shows two minima corresponding to two stable states with the magnetic spins aligned ferromagnetically along the \( y \)-axis. We refer to this as the strong coupling regime. Fig. 9(b) shows the landscape for the case \( I=0.5 \times 10^{-3} \text{ J/m}^2 \) which exhibits four minima consisting of the two stable ferromagnetic states and an additional two metastable states in which the spins are aligned antiferromagnetically along the \( y \)-axis. We refer to this as the weak coupling regime.

Associated with each of the local minimum energy states \( \sigma_i \) is a basin of attraction defined as that region of phase space comprising of the states that evolve asymptotically to the state \( \sigma_i \). We denote the basin of attraction associated with the state \( \sigma_i \) as \( \Omega_i \). The plots show the stable minima corresponding to each of the states \( \sigma_i \), two in the strong coupling case as in Fig. 9(a) and four in the weak coupling case as in Fig. 9(b), together with the boundaries separating each of the basins \( \Omega_i \). We denote the boundary separating two basins of attraction \( \Omega_i \) and \( \Omega_j \) by \( \Gamma_{ij} \). The boundaries \( \Gamma_{ij} \) are indicated in Fig. 9 for both the strong and the weak coupling cases by the black lines.

The following approach is based on the Langer formalism previously developed for magnetic grain reversal. If we denote by \( \rho(x,t) \) the probability density that the system is in state \( x \), where \( x \) denotes the contravariant vector \( \{x^1, x^2, x^3, x^4\} = \{\theta_1, \theta_2, \phi_1, \phi_2\} \), then we can define \( p_i \) the probability that the system is in state \( i \) as,
\[
p_i(t) = \int_{\Omega_i} \rho(x,t) d\Omega.
\]

The probability density may be calculated from the Fokker-Planck equation (FPE) which may be written in terms of the coordinates \( x^\mu \) as,
\[
\frac{\partial \rho(x,t)}{\partial t} = \nabla_{\mu} J^\mu(x,t),
\]
where the probability current density \( J^\mu(x,t) \) consists of an advective term and a diffusive term,
\[
J^\mu(x,t) = \rho(x,t) v^\mu - \frac{\gamma^2 D}{1 + \alpha^2 g^\mu\nu \nabla_\nu \rho(x,t)},
\]
with \( D = \alpha k_B T/\gamma_B m \), with \( m = M_1 v_1 + M_2 v_2 \), \( g^\mu\nu \) is the metric tensor and \( v^\mu(x) = \langle dx^\mu/dt \rangle \) is the velocity field calculated from the LLG equation and which can be written in covariant form as \( v^\mu(x,t) = -T^\mu\nu(x) \partial E(x)/\partial x^\nu \).
For the processes of interest in this study we consider steady state probability distributions \(\rho(x, t) = 0\) and define the crossover \(c(x)\) function as,

\[
\rho(x) = c(x) \exp \left( -\frac{E(x)}{k_BT} \right).
\] (19)

Substituting this expression into the FPE equation yields a differential equation for the crossover function

\[
\left( \frac{\alpha}{1 + \alpha^2} \nabla \cdot \gamma_{\mu\nu}(x) - \frac{1}{k_BT} \frac{\partial E(x)}{\partial x^\nu} \tilde{\nabla}^{\mu\nu}(x) \right) \frac{\partial c(x)}{\partial x^\nu} = 0,
\] (20)

we note that this equation admits the non-trivial solution corresponding to thermodynamic equilibrium \(c(x) = \text{constant}\).

In cases where the depth of the basins are much greater than \(k_BT\), steady state solutions exist for which the crossover function is essentially constant \(c(x) \approx c_i\) for \(x \in \Omega_i\) except in a narrow region in the neighborhood of the boundaries \(\Gamma_{ij}\) where it goes from \(c_i \to c_j\) on crossing the boundary from \(\Omega_i \to \Omega_j\). These solutions correspond to a state of ‘local’ equilibrium with thermodynamic equilibrium corresponding to the special case \(c_i = \text{constant}\) for all \(i\). The coefficients \(c_i\) may be expressed in terms of the probabilities \(p_i\), as,

\[
p_i \approx c_i \int_{\Omega_i} \exp \left( -\frac{E(x)}{k_BT} \right) d\Omega = c_i Z_i,
\] (21)

so that \(c_i = p_i/Z_i\). Since we are interested in situations where the energy scales are much greater than \(k_BT\), then dominant contribution to the integrand will come from the region \(x \approx x_i\). Approximating the energy by a quadratic,

\[
E(x) \approx E(x_i) + \sum \left. \frac{\partial^2 E(x)}{\partial x^\mu \partial x^\nu} \right|_{x=x_i} (x-x_i)^\mu (x-x_i)^\nu,
\] (22)

which may be evaluated to give

\[
c_i = \frac{(k_BT)^2}{\sqrt{\prod_k \eta_k(i)}},
\] (23)

where \(\eta_k(i)\) denote the eigenvalues of the Hessian \(E_{\mu\nu}(x_i) = \partial^2 E(x)/\partial x^\mu \partial x^\nu\) at \(x_i\).

Since the system will be in thermodynamic equilibrium if and only if \(c_i = \text{constant}\) there will be a net probability current \(\mathcal{I}_{ij} \propto c_i - c_j\) between \(\Omega_i\) and \(\Omega_j\) given by,

\[
\mathcal{I}_{ij} = \int_{\Gamma_{ij}} J^\mu dS^\mu,
\] (24)

where \(dS^\mu\) denotes the components of infinitesimal elements of the 3 dimensional hypersurface \(\Gamma_{ij}\) and \(J^\mu\) is the current density defined by Eq. 18. Omitting the divergenceless terms, this may be written in terms of the crossover function \(c(x)\) as,

\[
J^\mu = -\frac{\alpha}{1 + \alpha^2} \left( \frac{\gamma_{B} k_BT}{m} \right) \tilde{T}^{\mu\nu} \frac{\partial c(x)}{\partial x^\nu} \exp \left( -\frac{E(x)}{k_BT} \right).
\] (25)

We note that the current depends exponentially on the ratio \(E(x)/k_BT\). Since we are interested in situations where the energy scales are much greater than \(k_BT\), then the dominant contribution to the integrand will be from the section around the point \(x_s \in \Gamma_{ij}\), where \(E(x)\) is a minimum and hence the energy is stationary

\[
\left. \frac{\partial E(x)/\partial x^\mu}{} \right|_{x=x_s} = 0.
\]

Presuming that the location of the energy minima and the saddle points are known using the MEP or other techniques, the evaluation of \(\mathcal{I}_{ij}\) is straightforward. Specifying the \(c_i\) and \(c_j\) in terms of the probabilities \(p_i\) and \(p_j\) from Eq. 21 we solve for the crossover function \(c(x)\) in the region around the saddle point using a quadratic expression for the energy

\[
E(x) \approx E(x_s) + \sum \left. \frac{\partial^2 E(x)}{\partial x^\mu \partial x^\nu} \right|_{x=x_s} (x-x_s)^\mu (x-x_s)^\nu,
\] (26)

to obtain an expression for \(\mathcal{I}_{ij}\) in terms of probabilities \(p_i\) and \(p_j\) and the Hessian \(E_{\mu\nu}(x_s) = \partial^2 E(x)/\partial x^\mu \partial x^\nu\) at \(x=x_s\). While straightforward in principle the calculation is complicated by the fact that symmetry around the \(y\)-axis gives rise to a zero energy eigenvalue and the fact that \(x_s\) is not an energy minimum but a saddle point means that one of the eigenvalues, \(\kappa\), is negative. These considerations lead to the following expression for the transition rate, as used in Sec. IV

\[
\mathcal{I}_{ij} = r_{ij} p_i - r_{ji} p_j,
\] (27)

where the rate coefficients have the form Arrhennius-Néel law,

\[
r_{ij} = \frac{\alpha \gamma_B}{1 + \alpha^2} \frac{\kappa}{m} \left( \frac{1}{2\pi k_BT} \right)^{\frac{3}{2}} \exp \left( -\frac{E_{Bij}}{k_BT} \right).
\] (28)

In this expression, \(\kappa\) is determined by solving the following eigenvalue equation,\(^{28}\)

\[
\lambda_{\mu} \sum_{\nu} \tilde{M}^{\mu\nu} U^\nu = \kappa U^\nu,
\] (29)

where \(\tilde{M}^{\mu\nu}\) is defined by

\[
\frac{1}{k_BT} \frac{\partial E(x)}{\partial x^\mu} \tilde{T}^{\mu\nu} \approx \lambda_{\nu} x^\nu \tilde{M}^{\mu\nu},
\] (30)

using the expansion Eq. (25).