Effect of exchange interaction on superparamagnetic relaxation

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We use Langer’s approach to calculate the reaction rate of a system of two (classical) spins interacting via the exchange coupling $J$ in a magnetic field $H$, with uniaxial anisotropy of constant $K$. We find a particular value of the exchange coupling, that is $j \equiv J/K = j_c \equiv 1 - h^2$, where $h \equiv H/2K$, which separates two regimes corresponding to a two-stage and one-stage switching. For $j \gg j_c$ the Néel-Brown result for the one-spin problem is recovered.

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I. INTRODUCTION

Due to their high coercivity, single-domain nanoparticles exhibit long-range stability of the magnetization and thereby that of the information stored in recording media. The storage density may be increased by using very small particles, but then surface effects become dominant and affect the magnetization relaxation. Therefore, one of our aims is to include surface effects in the calculation of the relaxation time of the particle magnetization. However, this requires a microscopic approach to account for the local environment inside the particle, and thus include microscopic interactions such as spin-spin exchange, in addition to anisotropy and applied field. Unfortunately, this leads to a rather difficult task owing to the large number of degrees of freedom which hinders any attempt to analyze the energiescape. For this reason, inter alia, calculations of the reversal time of the magnetization of fine single-domain ferromagnetic particles, initiated by Néel [1], and set firmly in the context of the theory of stochastic processes by Brown [2], have invariably ignored all kind of interactions and included only the internal anisotropy of the particle, the random field due to thermal fluctuations, and the Zeeman term.

In 1968 Langer [3] came up with a general approach to the calculation of the reaction rate of a system with $N$ degrees of freedom, which is a powerful method of attack on the problem of interaction. Within this approach the problem of calculating the relaxation time for a multi-dimensional process is reduced to solving a steady-state Fokker-Planck equation (SSFPE) in the immediate neighborhood of the saddle point that the system crosses as it goes from a metastable state to another state of greater stability. The idea is that a steady-state situation can be set up by continuously replenishing the metastable state at a rate equal to the rate at which it is leaking across the activation energy barrier. A brief review of the main steps and application to the problem at hand are given after Eq. (8), see also [4] for uniform and non-uniform magnetization, and [5] for comparison with Kramers’ theory. However, Langer’s approach is only valid in the limit of intermediate-to-high damping (IHD) because of the inherent assumption that the potential energy in the vicinity of the saddle point may be approximated by its second-order Taylor expansion. The result for small damping fails because the region of deviation from the Maxwell-Boltzmann distribution set up in the well extends far beyond the narrow region at the top of the barrier.

The problem associated with the generalization of Brown’s theory to include interactions is rather difficult and can in general be solved only numerically. But before attacking this problem, one needs to understand the effect of exchange interaction on the relaxation time of the minimal system, i.e., for a pair of atomic spins coupled via exchange interaction, including of course the usual magneto-crystalline anisotropy and Zeeman terms. Besides, in the case of exchange interaction, this is the unique non-trivial step towards the above-mentioned generalization, where analytical expressions can be obtained for the relaxation time. It is the purpose of this work to solve this problem within Langer’s approach and to compare with the Néel-Brown result for the one-spin problem. As a byproduct, we will show that Langer’s quadratic approximation at the saddle point fails when the exchange coupling assumes a “critical value” even in the IHD limit. In this case, finiteness of the relaxation rate requires a higher-order expansion of the energy near the saddle point. The generalization to a multi-spin particle will be briefly discussed at the end.

We finally mention that the effect of dipolar interactions on the relaxation time has been considered by a few
FIG. 1: Energyscape from Eq. (1) ($\varphi = 0$) for $j = 0.25, 0.8, 1.5$ and $h = 0.1$. The arrows indicate the switching paths.

authors. In Refs. [6] approximate analytical expressions were obtained for the relaxation time in various situations of assemblies of magnetic moments. In Ref. [7] a pair of coupled dipoles was dealt with using the (numerical) Langevin approach.

II. STATEMENT OF THE PROBLEM AND ANALYSIS OF THE ENERGYSCAPE

We consider a system of two exchange-coupled spins with the Hamiltonian

$$
\mathcal{H} = -\frac{j}{2} \vec{S}_1 \cdot \vec{S}_2 - \frac{1}{2} \left[ (\vec{S}_1 \cdot \vec{e}_1)^2 + (\vec{S}_2 \cdot \vec{e}_2)^2 \right] - \vec{h} \cdot (\vec{S}_1 + \vec{S}_2),
$$

where $J > 0$ is the exchange coupling, $K > 0$ the anisotropy constant, $H$ the applied magnetic field, and $h$ the reduced field, i.e., $0 \leq h < 1$. $\vec{e}_i$ are uniaxial anisotropy unit vectors. Here we restrict ourselves to the case $\vec{h} \parallel \vec{e}_i, i = 1, 2$. Owing to the symmetry of this system with respect to rotations around the easy axis, the number of variables reduces to three, $\theta_1, \theta_2, \varphi \equiv \varphi_1 - \varphi_2$.

Now we apply Langer’s approach to the energy (1) and study the relaxation rate as a function of the exchange coupling $j$. We first analyze the energyscape in Fig. 1. The absolute minimum of the energy (1) corresponds to the ferromagnetic order along the easy axis,

$$(0, 0, \varphi); \quad e^{(0)}_{ss} = -\frac{j}{2} - 2h - 1,$$

where henceforth $e^{(0)}$ denotes the energy of the state. One metastable state corresponds to a ferromagnetic order opposite to the field

$$(\pi, \pi; \varphi); \quad e^{(0)}_{m1} = -\frac{j}{2} + 2h - 1.$$

There is also the metastable state of anti-ferromagnetic order,

$$(0, \pi; \varphi) \text{ or } (\pi, 0; \varphi); \quad e^{(0)}_{m2} = \frac{j}{2} - 1.$$  

As to saddle points, we find that their number and loci crucially depend on the exchange coupling constant $j$. More precisely, for $j > j_c \equiv 1 - h^2$, there is a single saddle point given by

$$\left( \cos \theta_1 = \cos \theta_2 = -h; \varphi \right); \quad e^{(0)}_{s} = -\frac{j}{2} + h^2,$$

whereas for $j < j_c$ there are two saddle points given by

$$\cos \theta_{1,2} = \frac{1}{2} \left( -h - a^2 \pm \sqrt{\Delta^2} \right) \equiv X^\pm_2; \varphi,$$
where $\varepsilon = \pm$ and $a^c = \varepsilon \sqrt{1 - j}$, $b^c = \varepsilon \sqrt{1 + (j/2)^2/(1 - j)}$, $\Delta^c = (h + a^c)^2 + 2j - 4b^c h$, with energy,

$$
e^{(0)}_c = -\frac{j}{2} \sqrt{(1 + b^c h - \frac{j}{2})^2 - (a^c + h)^2 - \frac{j}{2} (1 + b^c h - \frac{j}{2}) + \frac{1}{2} (h^2 - (a^c)^2 + 2b^c h).}
$$

(7)

At $j = j_c$, the saddle point is merged with $\varepsilon = +$ merges with the saddle point, while that with $\varepsilon = -$ merges with the metastable state, see Fig. 1 central panel.

Starting with both spins aligned in the metastable state with $\theta_1 = \pi$, if $j < j_c$ one of the two spins crosses the saddle point ($\varepsilon = +$) into the stable state ($\varepsilon = -$) and the second spin follows through the stable state ($\varepsilon = -$), of lower energy due to the exchange coupling (see Fig. 2(left)), ending up in the stable state ($\varepsilon = -$). In Fig. 1(left) the path is indicated by a pair of curved arrows. There are actually two such paths corresponding to the two-fold symmetry of the problem owing to the full identity of the two spins. Note that when the first spin starts to switch and arrives at $\theta_1 \sim \pi/2$, the second spin has $\theta_2 \lesssim \pi$ (hence the curved arrows in Fig. 1), which suggests that in the switching process of the first spin, the position of the second spin undergoes some fluctuations creating a small transverse field, and when $\theta_1 = 0$ the second spin goes back to the position $\theta_2 = \pi$ before it proceeds to switch in turn. The successive switching of the two spins through the corresponding saddle points is a sequential two-step process, so the relaxation rates for $j < j_c$ add up inverse-wise. In the case $j > j_c$, the two spins cross the unique saddle point to go from the metastable state into the stable one in a single step, see Fig. 1(right) where the path is indicated by a single straight arrow. There is the symmetry $\pm \theta^{(s)}$, which leads to a factor of 2 in the relaxation rate. Therefore, if we denote by $\Gamma^{+}_{j \leq j_c}$, $\Gamma^{-}_{j \leq j_c}$, and $\Gamma^{\pm}_{j \geq j_c}$ the respective relaxation rates, the relaxation rate of the two-spin system is given by

$$
\Gamma = \begin{cases} 2\Gamma^{+}_{j \leq j_c}, & \Gamma^{-}_{j \leq j_c} / (\Gamma^{+}_{j \leq j_c} + \Gamma^{-}_{j \leq j_c}) \text{ if } j \leq j_c \\ 2\Gamma^{\pm}_{j \geq j_c}, & \text{if } j \geq j_c. \end{cases}
$$

(8)

In Langer’s approach the $\Gamma$’s are obtained from the SSFPE linearized around each saddle point, using the fluctuating variables $\eta_i = (t_i, p_i)$ where $t_i = \theta_t - \theta_t^i$, $p_i = \varphi_i - \varphi_i^t$, $i = 1, 2$, or more adequately the “canonical” variables $\psi_i = (\xi_\pm, \zeta_\pm)$ with $\xi_\pm = (t_1 \pm t_2)/\sqrt{2}$, $\zeta_\pm = (p_1 \pm p_2)/\sqrt{2}$, in which the energy Hessian is diagonal. The deterministic dynamics of the system is governed by the Landau-Lifshitz equations, which upon linearization near the saddle point, read

$$
\begin{align*}
\partial_t \xi_i &= -\partial_{\xi_i} \mathcal{H}_2 - \alpha \partial_{\xi_i} \mathcal{H}_2 \\
\partial_t \zeta_i &= -\alpha \partial_{\xi_i} \mathcal{H}_2 + \partial_{\xi_i} \mathcal{H}_2, & i = 1, 2
\end{align*}
$$

(9)

where $\mathcal{H}_2$ is the quadratic approximation of the energy near the saddle point and $\alpha$ is the damping parameter. In matrix form Eqs. (9) become $\partial_t \eta_i = \sum_j M_{ij} \partial_{\xi_j} \mathcal{H}_2$, where $M$ is the dynamic matrix containing the precessional and dissipative parts. Rather than investigating the stochastic trajectories $\eta_i(t)$ that arise by adding a noise term in Eqs. (9), Langer concentrates on the distribution function $\rho(\eta_i(t), t)$ as the probability that the system is found in the configuration $\{\eta_i\}$ at time $t$. The time evolution of $\rho$ is governed by the Fokker-Planck equation (FPE) $\partial_t \rho + \sum_i \partial_{\eta_i} J_i = 0$, which is a continuity equation with the probability current $J_i$ given, the saddle point, by $J_i = -\sum_j M_{ij} (\partial_{\eta_i} \mathcal{H}_2 + 1 / k_B T \partial_{\eta_i} \rho) \rho$. In order to calculate the nucleation rate, one must solve the FPE. A particular solution is obtained at equilibrium and is given by the Maxwell-Boltzmann distribution $\rho_{eq} = \exp(-\beta \mathcal{H}(\eta)/2)$, which corresponds to zero current, $J_i = 0$. However, what is really needed is a finite probability current flowing across the saddle point. In fact, instead of solving the time-dependent FPE, Langer solves the SSFPE $\partial_t \rho = 0$ near the saddle point. The steady-state situation may be realized by imposing the boundary conditions: $\rho \simeq \rho_{eq}$ near the metastable state and $\rho \lesssim 0$ beyond the saddle point. Then, the problem of calculating the escape rate reduces to the calculation of the total current by integrating the probability current $J_i$ over a surface through the saddle point. This led Langer to his famous expression for the relaxation rate which is valid in the HLD limit.

However, since the escape rate is simply given by the ratio of the total current through the saddle point to the number of particles in the metastable state. Langer’s result for the escape rate can in fact be achieved by only computing the energy-Hessian eigenvalues near the saddle points and metastable states, from which one then infers the partition function $Z_\text{m}$ of the system restricted to the region around the saddle point where the energy-Hessian negative eigenvalue is (formally) $\lambda_\text{n}'s$ taken with absolute value, and the partition function $Z_\text{m}$ of the region around the metastable state. When computing these partition functions, one has to identify and take care of each Goldstone mode, that is a massless mode or zero-energy fluctuation associated with a continuous unbroken global symmetry. Finally, one computes the unique negative eigenvalue $\kappa$ of the SSFPE corresponding to the unstable mode at the saddle point as the negative eigenvalue of the dynamic matrix $M_{mn} = -\lambda_\text{n}(DMD^t)_{mn}$, where the $\lambda_\text{n}'s$ are the eigenvalues of the Hessian at the saddle point and $D$ is the transformation matrix from $\eta_i$ to $\psi_i$. 
Consequently, Langer’s final expression for the escape rate is rewritten in the following somewhat more practical form

$$\Gamma = \frac{|\kappa|}{2\pi} \tilde{Z}_m,$$  (10)

where $|\kappa|$ is the attempt frequency which contains the damping parameter $\alpha$.

### III. RELAXATION RATE OF THE TWO-SPIN SYSTEM

Now, we give the different relaxation rates for $j > j_c, j < j_c$, and $j \approx j_c$.

#### Relaxation rate for $j > j_c$:

The quadratic expansion of the energy $H^{(2)}$ at the saddle point $|s|$ reads,

$$H^{(2)} = e_s^{(0)} - \frac{j_c}{2} \xi^2 + \frac{j - j_c}{2} \xi^2 + \frac{jj_c}{2} \xi^2,$$  (11)

with zero eigenvalue for the $\zeta$ mode, that is the Goldstone mode associated with the rotation around the easy axis. $e_s^{(0)}$ is given in Eq. (5). The negative eigenvalue of the SSFPE, corresponding to the unstable mode, is $\kappa = -\alpha j_c$.

The partition function at the saddle point $Z_s = \int d\Omega_1 d\Omega_2 e^{-\beta H_s}$, where $d\Omega_i = \sin \theta_i d\theta_i d\phi_i$, $i = 1, 2$ is calculated by changing to the variables $\xi, \zeta$, setting $\sin \theta_i \simeq \sqrt{j_c}$ (see Eq. (5)) in the integration measure, and finally computing the Gaussian integrals. Hence,

$$\tilde{Z}_s = 2\pi \left(\frac{2\pi}{\beta}\right)^{3/2} \frac{1}{\sqrt{1 - j_c/j}} e^{-\beta e_s^{(0)}},$$  (12)

where we have formally replaced the negative eigenvalue $(-j_c)$ by its absolute value. The partition function $Z_m$ at the metastable state $|m|$ is computed by expanding the energy up to 2nd order, leading to

$$Z_m = e^{-\beta e_m^{(0)}} \left(\frac{2\pi}{\beta}\right)^2 \frac{1}{(1 - h)(j + 1 - h)}.$$  (13)

Using Eq. (10) and inserting the symmetry factor of 2, the relaxation rate for $j > j_c$ reads

$$\Gamma_{j > j_c} = 2\alpha \sqrt{\frac{\beta}{2\pi}} (1 - h^2)(1 - h) \frac{1 + (1 - h)/j}{\sqrt{1 - j_c/j}} \times e^{-\beta \Delta e^{(0)}}, \quad \Delta e^{(0)} = (1 - h)^2.$$  (14)

For $j \to \infty$, (14) tends to the Néel-Brown result,

$$\Gamma_{j > j_c} \to 2\alpha \sqrt{\frac{\beta}{2\pi}} (1 - h^2)(1 - h) e^{-\beta (1 - h)^2},$$  (15)

for the relaxation rate of one rigid pair of spins with a barrier height twice that of one spin. Note, however, that this convergence is very slow so that $\Gamma_{j > j_c}$ remains above the Néel-Brown result and only merges with the latter for $j \gtrsim 10$.

#### Relaxation rate for $j < j_c$:

Here we are faced with a semi-analytical case since the attempt frequencies $|\kappa^\varepsilon|$ (for $\varepsilon = \pm$) cannot be obtained in a closed form and are thus computed numerically. On the other hand, following the same procedure as for $j > j_c$ we obtain the relaxation rate for $\varepsilon = \pm$,

$$\Gamma^\varepsilon_{j < j_c} = \sqrt{\frac{\beta}{\pi}} |\kappa^\varepsilon| \left(\frac{P^\varepsilon}{j}\right)^{\frac{N^\varepsilon}{\sqrt{R^\varepsilon_+ R^\varepsilon_- + 2Q^\varepsilon(R^\varepsilon_+ + R^\varepsilon_-)}}} e^{-\beta \Delta e^{(0)}},$$  (16)

where $\Delta e^{(0)}$ is the barrier height given by the energy $H^{(2)}$ measured with respect to $|s|$ or $|m|$ for $\varepsilon = +, -$ respectively, and (see Eq. 4 et seq. for notation) $P^\varepsilon = \sqrt{(1 + b^\varepsilon h - j/2)^2 - (a^\varepsilon + h)^2}$, $Q^\varepsilon = b^\varepsilon h - j/2 + P^\varepsilon$, $R^\varepsilon_\pm = -1 + X^\varepsilon_\pm (2X^\varepsilon_\pm + h)$, $N^+ = (1 - h)(j + 1 - h)$, $N^- = j_c - j$. The limit of the relaxation rate (16) when $j \to 0$ is just the Néel-Brown result for one spin. Indeed, the product of the last two factors in the prefactor tend to $(1 - h)/\sqrt{2}$, the attempt frequencies tend to $\alpha j_c = \alpha (1 - h^2)$, and the energy barriers $\Delta e^{(0)} \to (1 - h)^2/2$. 


Note that in the present regime of $j < j_c$, the large value of anisotropy has not changed the temperature dependence of the individual relaxation rates, i.e., $\Gamma_{j < j_c}^+$, with $\varepsilon = \pm$. This is due to the fact that $1/\sqrt{T}$ appears in the prefactor each time there is a continuously degenerate class of saddle points [5], which is indeed the case for $j > j_c$ and $j < j_c$ with $\varepsilon = +$ and $\varepsilon = -$. However, anisotropy does affect the temperature dependence of the relaxation rate of the two-spin system, since for $j < j_c$ there are two saddle points bringing each a factor $1/\sqrt{T}$, see the first line in Eq. [8].

The case of $j \approx j_c$: When $j$ approaches $j_c$ either from above or from below, more Hessian eigenvalues (in addition to $\lambda_{\xi+}$) vanish, rendering the saddle point rather flat and thus leading to a divergent relaxation rate. Indeed, for $j \approx j_c$ the relaxation rate [14] diverges, which clearly shows that Langer’s approach which uses a quadratic approximation for the energy at the saddle point, e.g., Eq. [11], fails in this case. The remedy is to push the energy expansion to the 6th order in the variable $\xi$, (since $\lambda_{\xi-} = j - j_c \rightarrow 0$ as $j \rightarrow j_c$), i.e.,

$$\delta e_s = e_s - e_s^{(0)} \simeq \frac{\lambda_\xi - \xi^2}{2} + \frac{c}{4} \xi^4 + \frac{d}{6} \xi^6,$$

(17)

where $c = (1 - j - 7h^2/4)/3 < 0$, $d = (j - 1 + 31h^2/16)/30 > 0$. Then, the contribution $\sqrt{2\pi/\beta\lambda_{\xi-}}$ of the mode $\xi_-$ to the relaxation rate [14] must be replaced by $\int_{-\infty}^{\infty} e^{-\delta e_s} d\xi$, upon which the divergence of $\Gamma_{j > j_c}^+$ is cut off (see Fig. 2 (right)). Similarly, when $j$ approaches $j_c$ from below, for $\varepsilon = -$, the eigenvalue $\lambda_{\xi-}$ vanishes at $j = j_c$, upon which the saddle point [12] ($\varepsilon = -$) merges with the state [11] and thereby the partition functions $\tilde{Z}_s$ and $Z_s$ tend to infinity. However, as $Z_m \propto 1/(j_c - j)$ increases much faster than $\tilde{Z}_s$, and $|\kappa^-| \rightarrow 0$, $\Gamma_{j < j_c}^+$ tends to zero as $j \rightarrow j_c$.

On the other hand, for $\varepsilon = +$, both $\lambda_{\xi+}$ and $\lambda_{\xi-}$ vanish leading to a divergent relaxation rate, since now $\tilde{Z}_s$ diverges but $Z_m$ in Eq. [14] remains finite for the metastable state [3] is well defined. Indeed, as $j \rightarrow j_c$, the relaxation rate $\Gamma_{j < j_c}^+$ goes over to the result in Eq. [14] upon making the change $j \leftrightarrow j_c$, and taking account of the symmetry factor. In this case, the divergence at the point $j = j_c$ cannot be cut off by expanding the energy beyond the 2nd order and $\Gamma_{j < j_c}^+$ is simply cut off at the point where it joins $\Gamma_{j > j_c}$ taking account of Eq. [17].

IV. DISCUSSION AND CONCLUSION

In Fig. 2 (right) we plot (in of) the relaxation rate of the two-spin system as defined in Eq. [8], under the condition of IHD, in which Langer’s approach is valid, that the reduced barrier height $\beta \Delta e^{(0)} \gg 1$ and $\alpha \beta \Delta e^{(0)} > 1$ [5]. We also plot separately both relaxation rates for $j \leq j_c$. We see that the relaxation rate of the two-spin system contains two unconnected branches corresponding to the two regimes, $j < j_c$ and $j > j_c$, the bridging of which would require a more sophisticated approach. Fig. 2 (right) also shows that as $j$ increases, but $j \ll j_c$, the relaxation rate $\Gamma_{j \leq j_c}^+$
decreases because the switching of the first spin is hindered by the (ferromagnetic) exchange coupling. While $\Gamma_{j \leq j_c}$ is an increasing function of $j$ with a faster rate, since now the exchange coupling works in favor of the switching of the second spin. This is also illustrated by the evolution of the energy barrier height in Fig. 2 (left). As $j$ approaches $j_c$ from below, the relaxation rate $\Gamma_{j \leq j_c}$ tends to $\Gamma_{j \geq j_c}$ because the respective saddle points merge at $j = j_c$. Whereas $\Gamma_{j \leq j_c}$ goes to zero since the corresponding saddle point merges with the antiferromagnetic state that is no longer accessible to the system. For $j \geq j_c$, as $j$ increases the minimum 2, the metastable state 3 and the saddle point 5 merge together, which means that the system is found in an “energy groove” along the direction $\theta_1 = \theta_2$ because the eigenvalue $\lambda_{\xi}$ corresponding to the mode $\theta_1 - \theta_2$ becomes very large, and thus the escape rate decreases and eventually reaches the Nél-Brown value at large $j$.

The present study has helped us understand the effect of exchange coupling on the relaxation rate of the two-spin system, and will be very useful for the generalization to multiple-spin small particles at least for small deviations from collinearity, where it has been shown 9 that the surface contribution to the macroscopic energy has a simple cubic anisotropy. However, this generalization can only be performed using numerical techniques. This is now attempted by the help of the ridge method 10 for probing the energy landscape and locating the saddle points, and by the (Onsager-Machlup) path integrals 11 for determining the most probable paths connecting a metastable state to a more stable state.

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[13] See Ref. 9 for a rigorous derivation
[14] Indeed, if the saddle point is to describe the nucleating fluctuation, there must be exactly one direction of motion away from the saddle point in which the solution of the equations of motion of the modes $\psi_n$ is unstable 9.