Calculations of Magnetic Skyrmion Annihilation by Quantum Mechanical Tunneling

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Theoretical analysis of the crossover temperature from over-the-barrier to quantum mechanical tunneling in multi-spin systems is presented and applied to the annihilation of a magnetic Skyrmions in a two-dimensional lattice. A remarkably strong dependence of the crossover temperature and the lifetime of the Skyrmions on the parameters in the extended Heisenberg Hamiltonian, i.e. the Dzyaloshinskii-Moriya interaction, out-of-plane anisotropy and applied magnetic field, is found and a region identified where quantum tunneling could be observed on laboratory time scale. We predict that Skyrmion tunneling could be observed in the extensively studied PdFe/Ir(111) system if an external magnetic field exceeding 6 T is applied.

Localized, non-collinear magnetic states are receiving a great deal of attention, in particular magnetic Skyrmions which have been proposed as elements in future spintronics devices [1–3]. Along with interesting transport properties, Skyrmions exhibit particle-like behavior and carry a topological charge enhancing their stability with respect to a uniform ferromagnetic ground state. A key issue is the lifetime of Skyrmions as a function of temperature and applied magnetic field. Two different mechanisms for the annihilation of a Skyrmion have been characterized by theoretical modeling of atomic scale systems: Collapse of the Skyrmion in the interior of the sample [4–7] and escape through the boundary of the magnetic domain [8]. A duplication process where a Skyrmion is transformed to a pair of identical Skyrmions has also been identified [9].

The activation energy for the various possible transitions can be calculated for a given spin Hamiltonian by finding the minimum energy path (MEP) from the local energy minimum characterizing the Skyrmion state to the final state minimum and evaluating the energy rise along the path. A maximum in energy along the path corresponds to a first order saddle point on the energy surface. The MEP can be found using the geodesic nudged elastic band method [3] and the calculation accelerated by making use of knowledge obtained about its shape in previous calculations and focusing only on a small part of the path near the maximum [10]. The pre-exponential in the Arrhenius type rate expression can be estimated using harmonic transition state theory (HTST) for magnetic systems, assuming classical over-the-barrier mechanism [11,12]. This has, for example, been done for Skyrmions in PdFe overlayer on an Ir(111) surface [7,8], a system that has been studied extensively in the laboratory [13,14]. The challenge is to design materials where magnetic Skyrmions are sufficiently stable at ambient temperature and still small enough to be used in spintronic devices. Theoretical calculations can help accelerate this development by predicting the stability of Skyrmions in various materials. Parameter values obtained from density functional theory for the PdFe/Ir(111) system [15] have been found to give results that are consistent with experimental measurements of Skyrmion lifetime [8].

In the calculations mentioned above, it was assumed that the system is thermally activated to overcome the energy barrier of the transition. It is, however, also possible that the system can quantum mechanically tunnel from the metastable Skyrmion state to the ferromagnetic ground state. Quantum mechanical tunneling of systems described by a single magnetic moment, within a macrospin approximation, has been studied extensively [16–22], in particular in the context of molecular magnets, both experimentally [23,24] and theoretically [17,21]. For example, the rate of magnetization reversals in a Mn₃ monomer and dimer molecular magnets has been calculated as a function of temperature and excellent agreement obtained with the experimentally measured rates [25,20] both in the high temperature classical regime and the onset temperature for tunneling, using a Hamiltonian parametrized using different experimental observations [20,21].

An intriguing question is whether quantum mechanical tunneling could contribute to the rate of annihilation of Skyrmions. Since Skyrmion stability is of central importance, a way to estimate whether tunneling can be a competing annihilation mechanism is needed. An experimental observation of Skyrmion tunneling would, furthermore, be an example of what is referred to as macroscopic quantum tunneling and would be of considerable interest in the study of quantum phenomena.

Recently, the quantum mechanical nature of Skyrmions has received some attention [27,29]. The quantum collapse of a magnetic skyrmions via tunneling has been estimated for the case where the external magnetic field is strong enough for the Skyrmion state...
to be close to the transition state and to be described by the Belavin-Polyakov (BP) soliton shape \[30\]. Our estimates show that the transition state is indeed well described by the BP shape for large Skyrmions of ca. 100 nm size. However calculations of mm-size Skyrmions in the FePd/Ir(111) in strong magnetic field \[31\], reveal that the BP-shape function does not describe well the Skyrmion state or even the transition state \[7, 8\]. In order to predict which materials and experimental conditions are likely to lend themselves to observations of Skyrmion tunneling, it is good to employ a detailed, multi-spin model to predict both the onset temperature for tunneling as well as the lifetime of the Skyrmion at that temperature.

In this Letter, we describe a method for estimating the onset temperature for quantum mechanical tunneling in a magnetic system of multiple spins. It represents an extension of the methodology presented earlier for systems described by a single magnetic moment \[20, 21\]. The method is then applied to estimate the onset temperature of Skyrmions in a thin layer such as a metal overlayer on a solid substrate for a wide range of parameters in the Hamiltonian: the Dzyaloshinskii-Moriya interaction, out-of-plane anisotropy, exchange coupling and external magnetic field. A region in parameter space is identified where the lifetime of the Skyrmion is on a laboratory time scale at the crossover temperature, thus identifying possible candidate materials for the observation of macroscopic tunneling of Skyrmions. For the widely studied PdFe/Ir(111) system it is estimated that Skyrmion tunneling could be observed if the external field exceeds 6 T.

Within harmonic transition state theory for over-the-barrier transitions in magnetic systems, the mechanism and rate of a thermally induced transition is characterized by the first order saddle point on the energy surface, \( s^\dagger \), representing the highest point along the MEP \[11, 12\]. There, the Hessian matrix, \( H \), has only one negative eigenvalue. The over-the-barrier description of the transition mechanism and method for estimating the rate is accurate at high enough temperature. As the temperature is lowered, the quantum mechanical tunneling eventually becomes the dominant transition mechanism. The challenge is to estimate this onset temperature for tunneling, \( T_c \).

The onset temperature can be found using instanton theory \[32, 34\] by finding the highest temperature at which a periodic solution of the equation of motion in imaginary time exists in the infinitesimal vicinity of the first-order saddle point on the energy surface. From the period of an instanton, \( \beta \), the corresponding temperature can be found as \( T = \hbar / k_B \beta \). The Euclidean (imaginary-time) action for a system with \( N \) spins that have direction \( s_i \), \( i = 1, \ldots, N \), and magnitude \( \mu \) is \[35\]

\[
Q[s, \partial_s s, \tau] = i\mu \sum_{k=1}^{N} \int_{-\beta/2}^{\beta/2} d\tau A_k \partial_s s_k + \int_{-\beta/2}^{\beta/2} d\tau H(s),
\]

where \( s = \{s_1, s_2, \ldots, s_N\} \), \( s_k(\tau) \) is a closed trajectory, \( s_k(-\beta/2) = s_k(\beta/2) \), and \( H(s) \) is the Hamiltonian. The first term in the equation is the Berry phase and \( A_k \) is referred to as Berry connection \[36\]. It is related to the area of a sphere for each spin bounded by the trajectory and can be expressed as \[35\]

\[
A_k \partial_s s_k = 2 \arctan \left( \frac{\partial_s s \cdot (s \times s_0)}{2 + 2s \cdot s_0 + \partial_s s \cdot s_0} \right)
\]

To find the instanton in the vicinity of \( s^\dagger \), the action in Eq. (1) is expanded up to second order

\[
Q[s] = Q^\dagger + \delta Q + \frac{1}{2} \delta^2 Q,
\]

where \( Q^\dagger = \beta E^\dagger \) is the energy at \( s^\dagger \) and \( s = s^\dagger + \delta s \). In addition to the boundary conditions, \( \delta s_k(-\beta/2) = \delta s_k(\beta/2) = 0 \), a normalization constraint needs to be added for all the spins, \( \delta s_k \cdot s_k = 0 \). At the stationary point \( \delta S = 0 \). The second order variation of the action in the vicinity of the saddle can be written as

\[
\frac{1}{2} \delta^2 Q = i\mu \sum_{k=1}^{N} \int_{-\beta/2}^{\beta/2} d\tau \left[ \frac{\partial_s \delta s_k \times s^0}{1 + s^0 \cdot s_k} + \frac{\partial_s \delta s_k \times s^0}{(1 + s^0 \cdot s_k)^2} \right] + \frac{\partial_s \delta s_k \times s_k^\dagger}{(1 + s^0 \cdot s_k^\dagger)^2} \right] \delta s_k + \int_{-\beta/2}^{\beta/2} d\tau \left[ \delta s \cdot \nabla^2 H \cdot \delta s \right],
\]

where \( s^0 \) is an arbitrary reference direction and \( \nabla^2 H \) is the Hessian matrix evaluated at \( s^\dagger \) and the tilde specifies a restriction to the tangent space \[9\]. The linearized Landau-Lifshitz equation can be obtained by setting \( \delta^2 Q = 0 \) and taking the cross product with \( s^\dagger \) from the left

\[
\partial_s \delta s_k = \frac{i}{\mu} \left[ s_k^\dagger \times \nabla^2_H \cdot \delta s_k \right], \quad \forall k \in [1, N].
\]

This can be written in a matrix form as

\[
\frac{\partial}{\partial \tau} \delta s = \frac{i}{\mu} M \delta s.
\]

The matrix \( M \) has \( N \) pairs of complex conjugate eigenvalues, \( \lambda_k \). One of the pairs, chosen here to correspond to \( k=1 \), is real valued but the other \( N-1 \) pairs are pure imaginary \( \lambda_k = \pm i\eta_k \). A discussion of the properties of
the matrix M can be found in Ref. [38].

The general solution for Eq. (6) has the following form

$$\delta s(\tau) = \sum_{k=1}^{N} c_k \mathbf{u}_k e^{\omega_k \tau} + c_k^* \mathbf{u}_k^* e^{-\omega_k \tau}, \quad \omega_k = i \lambda_k / \mu, \quad (7)$$

where the $\mathbf{u}_k$ are the eigenvectors and $\lambda_k$ the eigenvalues of $M$, and $c_k$ the expansion coefficients. The pair of real eigenvalues $\lambda_1$ corresponds to the periodic solution with eigenfrequency $\omega_1 = i \lambda_1 / \mu$ and this is the instanton. The period of motion along this trajectory relates to the frequency as $1 / \beta = |\omega_1| / 2 \pi$, which gives the onset temperature for tunneling as

$$T_c = \frac{\hbar |\omega_1|}{2 \pi k_B}. \quad (8)$$

Equation (8) has the same form as for particle systems [34], but there is a significant difference between two. In the case of particle rearrangements $\omega_1 = \sqrt{-\lambda_1}$, where $\lambda_1$ is the negative eigenvalue of the Hessian matrix at the first-order saddle point on the energy surface, whereas for magnetic systems, governed by the Landau-Lifshitz equation of motion, $\omega_1$ depends on the determinant of the Hessian matrix, i.e. all the eigenvalues. For a single spin described by spherical polar coordinates $\theta$ and $\phi$ [20]

$$T_c = \frac{\sqrt{E_{\theta\theta}^{\text{II}} E_{\phi\phi}^{\text{II}}} - \left(E_{\theta\phi}^{\text{II}}\right)^2}{2 \pi k_B |\sin \theta|}, \quad (9)$$

where $E_{\theta\theta}^{\text{II}}$, $E_{\phi\phi}^{\text{II}}$, and $E_{\theta\phi}^{\text{II}}$ are second derivatives of the energy function. For a multi-spin system, it is better to work with Cartesian coordinates, as has been done in the present case because of the problems that can arise if any of the spins points towards either of the two poles.

Once the onset temperature for tunneling has been found, the transition rate at that temperature can be evaluated from HTST [11].

In the calculations presented here, the energy of the system is described by an extended Heisenberg model

$$\mathcal{H} = \sum_{(ij)} [\mathbf{D}_{ij} \cdot (\mathbf{s}_i \times \mathbf{s}_j) - J \mathbf{s}_i \cdot \mathbf{s}_j] - \sum_{i=1}^{N} \left[\mu \mathbf{B} \cdot \mathbf{s}_i + K s_i^2 \right], \quad (10)$$

where $\mathbf{D}_{ij}$ the Dzyaloshinskii-Moriya vector lying in the plane of the lattice parallel to the vector pointing between two nearest neighbor sites $i$ and $j$, thereby supporting Bloch type Skyrmions, $J$ is the exchange coupling parameter, $K$ the out of plane anisotropy constant, and $\mathbf{B}$ the uniform external magnetic field applied perpendicular to the lattice plane. The sum $(ij)$ includes distinct nearest neighbor pairs. The system consists of 2500 spins on a triangular lattice with lattice spacing $a = 1$ and periodic boundary conditions. This model can, in particular, represent well the PdFe/Ir(111) system [8].

Fig. 1 shows the calculated onset temperature, $T_c$, for the tunneling of the Skyrmion to the ferromagnetic state as a function of scaled Dzyaloshinskii-Moriya parameter, $D / J$, and scaled anisotropy parameter, $K / J$, when the applied magnetic field is $B = 0.73 B_D$, where $B_D = D^2 / (\mu J)$ is the critical field [10]. The value of $T_c$ varies as a function of these parameters over the range of 1 to 4 K. The value calculated for the PdFe/Ir(111) system [13] is marked with a white star.

The change in annihilation mechanism from over-the-barrier to tunneling could be observed by measuring the lifetime of Skyrmions as a function of temperature, manifested by a shift from temperature dependent lifetime above $T_c$ to temperature independent lifetime below $T_c$. In order for this to be practical, the Skyrmion lifetime at $T_c$ should be on laboratory time scale, on the order of seconds or minutes. It is therefore important to also estimate the lifetime of the Skyrmion at $T_c$ and this can be done using HTST. Fig. 2 shows the lifetime as a function of the scaled parameters as well as the size of the Skyrmion. The lifetime changes remarkably strongly over the chosen small range of parameter values. Since the lifetime is also a strong function of temperature, the window for observing the crossover to tunneling is limited to a narrow range in parameter values. But, this range does exist and it is most likely possible to find materials where such measurements could be performed.
The stability of Skyrmions is related to their size \[11\], as can be seen from Fig. 2. The smaller the Skyrmion, the shorter the lifetime and this helps bring the lifetime at the onset temperature to laboratory time scale. By increasing the applied magnetic field, the size of the Skyrmion can be reduced and lifetime thereby shortened, as illustrated in Fig. 3 where results for the PdFe/Ir(111) system are shown. While the crossover temperature at \( B = 0.73BD \) is relatively high for PdFe/Ir(111) system, the lifetime at \( T_c \) is much too long for annihilation to be observed at that temperature. However, if the applied field is increased to \( B = 6 \text{T} \), the lifetime is brought down to minutes and the crossover temperature is still not too low, about 1.5 K. Fig. 3 shows how the the energy barrier for annihilation is reduced as the field is increased. We, therefore, predict that tunneling of Skyrmions could be observed in this system on laboratory time scale by applying a strong enough magnetic field. Other materials may of course be better suited for the observation of Skyrmion tunneling, but the PdFe/Ir(111) system is at least one possible candidate.

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