Mechanism and activation energy of magnetic skyrmion annihilation obtained from minimum energy path calculations

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The mechanism and activation energy for the annihilation of a magnetic skyrmion is studied by finding the minimum energy path for the transition in a system described by a Heisenberg-type Hamiltonian extended to include dipole-dipole, Dzyaloshinskii-Moriya, and anisotropy interactions so as to represent a Co monolayer on a Pt(111) surface. The annihilation mechanism involves isotropic shrinking of the skyrmion and slow increase of the energy until the transition state is reached after which the energy drops abruptly as the ferromagnetic final state forms. The maximum energy along the minimum energy path, which gives an estimate of the activation energy within the harmonic approximation of transition state theory, is found to be in excellent agreement with direct Langevin dynamics simulations at relatively high temperature carried out by Rohart *et al.* [Phys. Rev. B **93**, 214412 (2016)]. The dipole-dipole interaction, the computationally most demanding term in the Hamiltonian, is found to be important but its effect on the stability of the skyrmion and shape of the transition path can be mimicked accurately by reducing the anisotropy constant in the Hamiltonian.

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I. INTRODUCTION

Magnetic skyrmions are localized, noncollinear spin configurations demonstrating soliton-like behavior and may be promising candidates for future data storage due to their small size and topological stability [1]. They have been identified both in bulk magnetic materials and in thin magnetic films. For a review, see Ref. [2]. While most skyrmions have been observed and studied at low temperature, multilayer materials have also been made where skyrmions are stable at room temperature [3]. The stability of skyrmions against thermal fluctuations is an essential prerequisite for their use in memory devices. Originally, skyrmions were introduced in the context of elementary particles as configurations of continuous fields with topological charge. In a continuum limit, topological protection makes them stable against arbitrarily large fluctuations. However, in discrete systems where magnetic moments are localized on atoms, the topological protection is not strict. Instead, a skyrmion state is separated from the topologically simple (e.g., ferromagnetic) state by a finite energy barrier which determines its stability and lifetime. A methodology for accurately determining the mechanism and rate of annihilation of such noncollinear magnetic states is needed, for example, in the exploitation of these kinds of states in devices.

The assessment of the stability of magnetic states with respect to thermal fluctuations is an important problem also in a more general context [4]. The preparation of a magnetic system in a particular state can be destroyed by thermally activated transitions to other available states. Thermal activation also needs to be taken into account when assessing the stability of a system with respect to external perturbations such as a magnetic field, contributing, for example, to the temperature dependence of hysteresis loops [5–7].

Recent theoretical studies of the thermodynamic stability of skyrmions have been based on Monte Carlo simulations using a generalized Heisenberg Hamiltonian extended to include anisotropy as well as Dzyaloshinskii-Moriya interaction to stabilize the skyrmion state [8,9]. A system consisting of Fe and Pd overlayers on an Ir(111) substrate has been a particular focus and a phase diagram has been constructed [9]. Parameters characterizing the free energy surface have also been extracted from such simulations [8].

Transitions from a magnetic state which is stable on the time scale of seconds are rare events on the time scale of oscillations of the magnetic moments. Direct dynamical simulations of spin transitions in such systems are, therefore, not practical. By raising the temperature, thermally activated transitions can be made fast enough to be observable in simulations of spin dynamics, but the increased importance of entropic effects at high temperature can lead to a crossover to a different transition mechanism. The separation of time scales between the oscillations of magnetic moments and the rate of activated transitions, however, makes it possible to apply rare-event theories such as transition state theory (TST) [10] or Kramers theory [11]. Within the harmonic approximation to TST (HTST) [12] and within Kramers theory, the activation energy of a transition, the primary quantity determining thermal stability, is given by the energy difference between the local minimum of the energy surface corresponding to the initial state and the highest first-order saddle point (SP) located on a path connecting the initial- and final-state minima. In adaptations of these rate theories to magnetic systems [5,13–16], the magnitude of the magnetic vectors is either assumed to be independent of orientation or it is treated as a fast variable obtained from self-consistent calculations for fixed values of the slow orientational variables [17]. The energy surface of a system of N magnetic moments is then a function of $2N$ degrees of freedom, defining the orientation of the magnetic moments.

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The assessment of the thermal stability of a magnetic state essentially becomes a problem of identifying the mechanism and estimating the activation energy for the annihilation of the state. This can be accomplished by finding the minimum energy path (MEP) for the transition. For a magnetic system with several degrees of freedom, locating an MEP can be challenging. The difficulty arises from the need to minimize the energy with respect to all but one degree of freedom, the direction along the path in a multidimensional space. It is not known *a priori* which degree of freedom should be treated differently. The special degree of freedom along the MEP is often referred to as the *reaction coordinate*. The geodesic nudged elastic band (GNEB) method can be used for such calculations [18]. It is similar to a well-known method for atomic systems, the nudged elastic band (NEB) method [19,20], except that an additional projection is included due to the curved manifold of systems of magnetic vectors and a geodesic measure of distances [18].

Previously, an MEP for the annihilation of a magnetic skyrmion has been found by Bessarab and coworkers [18] using a Heisenberg-type Hamiltonian where the skyrmion is stabilized by the inclusion of the Dzyaloshinskii-Moriya interaction. The calculation made use of the GNEB method and the MEP turned out to involve gradual isotropic shrinking of the skyrmion followed by an abrupt decrease of the energy past the transition state as the ferromagnetic state forms. More recently, calculations have been presented based on a Heisenberg-type Hamiltonian where the skyrmion is also stabilized by the inclusion of the dipole-dipole interaction [21]. In addition to a path involving isotropic shrinking of the skyrmion as found by Bessarab *et al.*, a path with lower activation energy was also reported where a rotation of the central spins occurs at the same time as the diameter of the skyrmion becomes smaller. The second path was reported to have lower activation energy and thereby represents the dominant annihilation mechanism.

Since these are complex systems involving many degrees of freedom, it is quite possible that more than one MEP can exist connecting the skyrmion and ferromagnetic states. In principle, some kind of global optimization of MEPs can be used for such systems [22]. Most importantly, however, one needs to ensure that the minimization procedure used truly gives an MEP by verifying, in particular, that the maximum energy along the path corresponds to a first-order saddle point on the energy surface [18,23]. This can be accomplished by making sure the gradient vanishes at this point and the matrix of second derivatives (the Hessian matrix) has one and only one negative eigenvalue.

We present here results on the annihilation mechanism and activation energy of a magnetic skyrmion in the Co/Pt(111) model system and address in particular whether the skyrmion can annihilate by more than one mechanism. An MEP for the transition is found with a maximum energy that is in excellent agreement with the activation energy deduced from direct Langevin simulations for a relatively high temperature [21]. This demonstrates the applicability of MEP calculations and harmonic transition state theory when estimating the rate and identifying the mechanism of thermally driven magnetic transitions. The MEP has a significantly lower maximum energy than the two previously reported paths. Various different configurations of the initial path all led to the same MEP. We also address the importance of the dipole-dipole interaction and the possibility of including its effect simply by reducing the anisotropy constant in the Hamiltonian, as suggested by Rohart and coworkers [21]. The article is organized as follows: The model and methodology are described in the following section, then the results of the calculations are presented, and the article concludes with a discussion and summary.

II. MODEL AND METHODOLOGY

The model used here is the same as that presented by Rohart and coworkers [21]. The Co layer is described by a set of classical spins, $\mathbf{S}_i$, one on each atom, on a hexagonal lattice commensurate with the Pt(111) substrate where the site-to-site distance is $a = 2.51$ Å. The Hamiltonian is given by

$$E = \sum_{\langle i,j \rangle} [-J\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j + \mathbf{d}_{ij} \cdot (\hat{\mathbf{s}}_i \times \hat{\mathbf{s}}_j)] - \sum_i K(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{z}})^2 - \frac{\mu_0}{8\pi} \sum_{i,j \neq i} \frac{3(\mathbf{S}_i \cdot \hat{\mathbf{u}}_{ij})(\mathbf{S}_j \cdot \hat{\mathbf{u}}_{ij}) - \mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - \mu_0 \sum_i \mathbf{S}_i \cdot \mathbf{H}, \tag{1}$$

where $\hat{\mathbf{s}}_i = \mathbf{S}_i / ||\mathbf{S}_i||$. The first two terms are the Heisenberg and Dzyaloshinskii-Moriya interaction (DMI) with constants J and $\mathbf{d}$, respectively. The summation is over near-neighbor pairs $\langle i, j \rangle$. For a thin film, $\mathbf{d}_{ij} = d(\hat{\mathbf{u}}_{ij} \times \hat{\mathbf{z}})$, with $\hat{\mathbf{u}}_{ij}$ being the unit vector between sites i and j and $\hat{\mathbf{z}}$ being normal to the plane. The third term is the uniaxial anisotropy, characterized by constant K. The fourth term is the dipole-dipole (DD) interaction where r_{ij} is the distance between sites i and j (the summation is in the present case limited to 168 nearest neighbors). The last term is the Zeeman energy in field H, which is applied along $\hat{\mathbf{z}}$. The values of the various constants and the justification for the chosen values for representing a Co monolayer on Pt(111) have been given by Rohart *et al.* [21]. The values of the parameters are $J = 29$ meV/bond, $K = 0.4$ meV/atom, $d = -1.5$ meV/bond and $||\mathbf{S}_i|| = 2.1\ \mu_B$.

The ground state of the system has ferromagnetic (FM) ordering but a skyrmion with radial symmetry in a hedgehog configuration (in-plane magnetization pointing along the radial direction) represents a metastable state 498 meV higher in energy than the FM state. If the DD interaction is skipped, the energy barrier between the skyrmion state and the FM state nearly vanishes. It is therefore essential to include the DD interaction, but this represents the most computationally intensive part of the simulation.

The calculation of the MEP for the transition from the skyrmion state to the FM state involves the generation of intermediate replicas of the system (referred to as *images*) so as to produce a discrete representation of the transition path. A total of 21 images were used to represent the path, with the two endpoint images fixed at the energy minima corresponding to the skyrmion and FM state. The 19 intermediate images are then relaxed in a collective way. Such an optimization procedure, GNEB, for magnetic systems has been described in detail by Bessarab *et al.* [18] and is based on a widely used method, NEB, for finding MEPs for atomic rearrangements [19,20]. At each iteration of the minimization, the tangent to

the path needs to be updated using a stable tangent estimate [24]. It is important to have the right estimate of the tangent in order to identify for which degrees of freedom the energy should be minimized and for which degree of freedom not (i.e., the reaction coordinate). If the estimate of the tangent is not correct, the minimization will most likely not result in the placement of the images on the MEP. The highest image is pushed up in energy along the path using the climbing image algorithm [25] and thereby ends up being placed at a first-order saddle point on the energy surface. This is verified by first of all checking whether the gradient of the energy is close enough to zero at this point and second by evaluating a few of the lowest eigenvalues of the Hessian matrix using the iterative Lanczos algorithm to check whether one and only one of the eigenvalues is negative. Such tests are important to verify that an accurate estimate of the activation energy can be obtained from the optimized path [18].

The calculation needs to be started with some initial placement of the images, and if several MEPs are present between the given initial and final states of the transition, then the optimization will most likely lead to the MEP that is directly accessible from the initial path without any increase in energy. In that sense, the converged MEP is the one closest to the initial placement of the images. Several initial placements of the images were tried here in order to test whether more than one MEP is present for the annihilation of the skyrmion in this system.

III. RESULTS

An illustration of the skyrmion and the MEP obtained is shown in Fig. 1. Various initial placements of the images all led to the same converged MEP. The highest-energy climbing image is indeed found to be a first-order saddle point, the norm of the gradient vector for all 36 246 spins in the system being smaller than 10^{-4} meV/rad and the value of the lowest two eigenvalues being -1.18 and 0.002 meV/rad^2. The mechanism involves isotropic shrinking of the skyrmion and slow increase in energy until the maximum is reached after which the energy drops abruptly towards the ferromagnetic final state. The activation energy, which is the difference between the maximum energy along the MEP minus the initial-state energy, is found to be 37 meV. The orientation of the spins as a function of the distance from the center of the skyrmion at a few of the images, including the maximum energy image, is shown in Fig. 1(c).

When a magnetic field of 0.25 T is turned on [as specified by Eq. (1)], the path remains qualitatively similar but the activation energy drops to 21 meV. The skyrmion radius becomes smaller as illustrated in Fig. 1(d). The contribution of the various terms in the Hamiltonian to the total energy along the MEP is shown in Fig. 2(a) in the case of zero field and in Fig. 2(c) for a field of 0.25 T.

The DD interaction is important in this system. If it is not included, the MEP at this resolution (number of images) shows only a decrease in energy, even though the skyrmion is metastable in the sense that direct minimization of the skyrmion state does not result in a slide down to the FM state. Since the evaluation of the DD interaction is the most computationally intensive part of the calculation, it is interesting to see whether it can be represented in a simpler and more efficient way. Rohart *et al.* have suggested reducing the anisotropy constant so as to effectively mimic the inclusion of the DD interaction [21]. It has been demonstrated analytically in a continuum description that such a replacement is indeed equivalent for rotationally symmetric spin configurations [26]. As can be seen from Figs. 2(a) and 2(c), the variation of the DD interaction energy closely mimics that of the anisotropy energy. A path found after turning off the DD interaction and reducing the value of the anisotropy constant K from 0.4 to 0.293 meV is shown in Fig. 1(b). The energy of the skyrmion state above the FM state is the same with this effective Hamiltonian and the shape of the MEP and the activation energy turn out to be similar to results obtained with the full Hamiltonian. The only difference is a slightly longer total displacement along the path, i.e., total rotation of the spins in the system. The variation of the energy along the MEP found for the effective Hamiltonian where explicit DD interaction is not included is shown in Fig. 2(b) for the case of zero field and in Fig. 2(d) for a field of 0.25 T. The value of the effective anisotropy constant obtained here is slightly different from the value 0.276 meV reported in Ref. [21], likely because of the different way the DD interaction is evaluated.

It is important to relax all the spins collectively. When each spin is relaxed separately, i.e., the tangent to the path defined for each spin separately rather than collectively, additional constraints are introduced and the path optimization will not, in general, lead to convergence to the MEP. The result of such a calculation is also shown in Fig. 1(b). The apparent activation energy obtained from the maximum energy along this path is 97 meV, more than twice as high as the activation energy obtained from the MEP and an analysis of the Hessian at this image shows that it does not correspond to a first-order saddle point on the energy surface.

IV. DISCUSSION

The results of the MEP calculations presented here show that the activation energy for the annihilation of the skyrmion in this system is not higher than 37 meV in zero field and 21 meV when a destabilizing external magnetic field of 0.25 T is applied. While it is not possible to exclude that other MEPs exist between the skyrmion and FM in this system, our attempts to generate other MEPs by starting with different initial placement of the images all led to the same MEP after optimization of the path had been carried. It is in any case clear that the low-temperature activation energy cannot be higher than what the MEP found here indicates. An alternative path would only be relevant if it offers a mechanism with lower activation energy. Rohart and coworkers carried out Langevin dynamics simulations of the skyrmion annihilation [21]. The calculations were carried out with a magnetic field of 0.25 T and relatively high temperature, around 80 K, to make the annihilation transition fast enough to occur on the short time scale represented by the simulations. The temperature dependence of the rate extracted from the simulations was found to follow the Arrhenius law (see Fig. 2 in Ref. [21]) and an activation energy of 26 ± 4 meV was obtained from the slope. This is in excellent agreement with the maximum energy along the MEP we obtain in a field of 0.25 T, 21 meV.

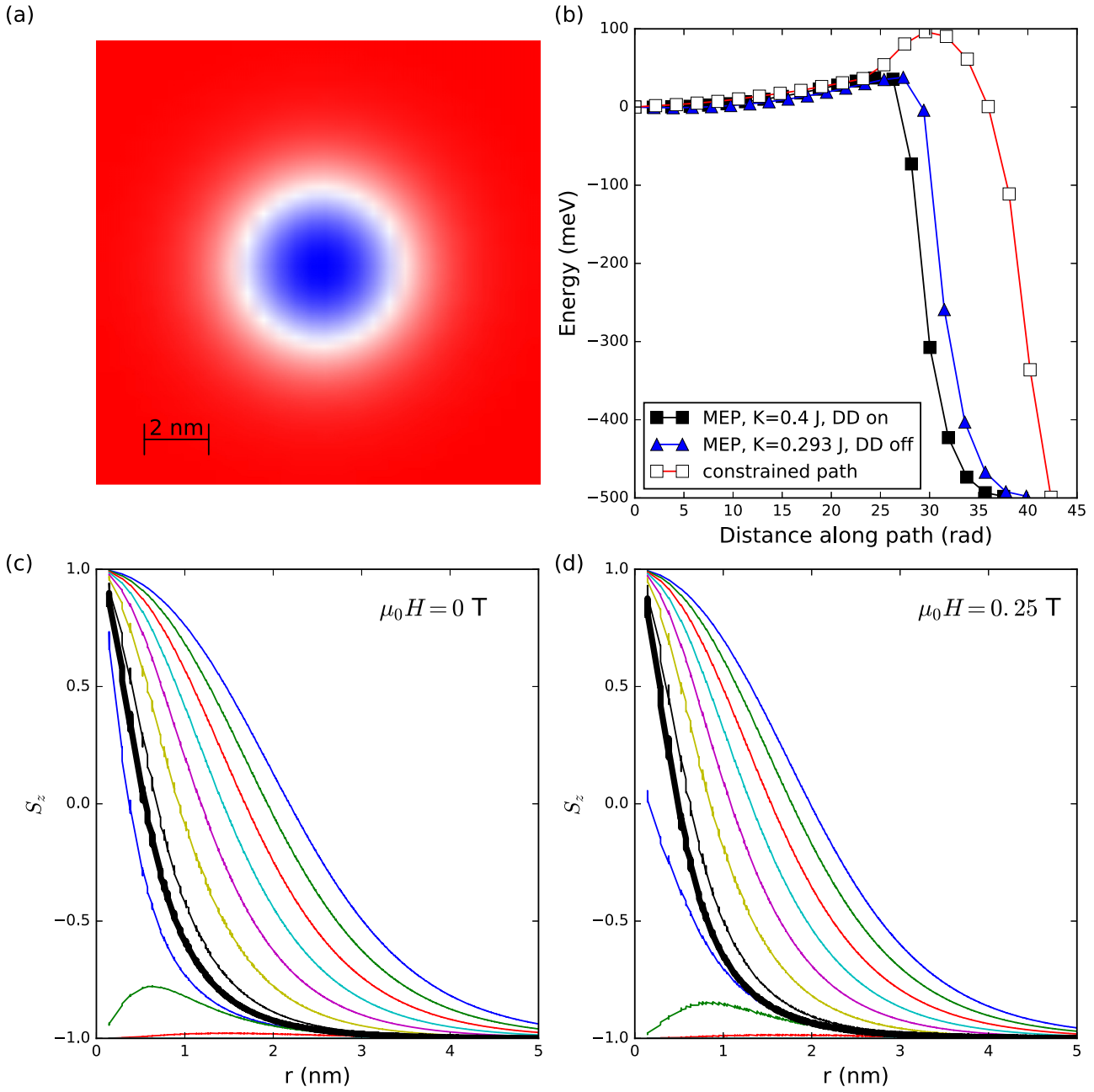

FIG. 1. (a) Skyrmion configuration in zero field. The color code represents the perpendicular spin component from blue (upward) to red (downward) through white (in plane). (b) Energy along minimum energy paths obtained with full optimization [18] using the Hamiltonian, Eq. (1), and anisotropy constant $K = 0.4$ meV (filled squares), and where the dipole-dipole interaction is not included but anisotropy constant reduced to $K = 0.293$ meV (filled triangles). The activation energy for annihilation is 37 meV in the former and 38 meV in the latter. This shows how the effect of the dipole-dipole interaction can effectively be included by reducing the anisotropy constant. A path obtained by incomplete optimization where each spin is relaxed separately instead of collective relaxation is also shown (open squares). From this path the activation energy would be overestimated as 97 meV. This illustrates how important it is to carry out full optimization when finding minimum energy paths. (c), (d) Perpendicular spin component as a function of the distance from the center of the skyrmion for several images along the minimum energy path for annihilation, (c) in zero magnetic field and (d) in a field of 0.25 T. The curve corresponding to the highest energy image is thicker.

The maximum energy along the MEP gives an estimate of the activation energy within the harmonic approximation of transition state theory [14]. At the high temperature where the skyrmion is unstable enough for short-time-scale Langevin simulations to reveal annihilation events, significant deviations can be expected from the harmonic approximation, so the agreement between our MEP calculations and the Langevin simulations of Rohart *et al.* is remarkably good. The Hamiltonian used here is practically the same as that of Rohart *et al.* except that in our calculations the DD interaction was evaluated by a sum over 168 nearest neighbors, while they made use of a fast Fourier transform [27].

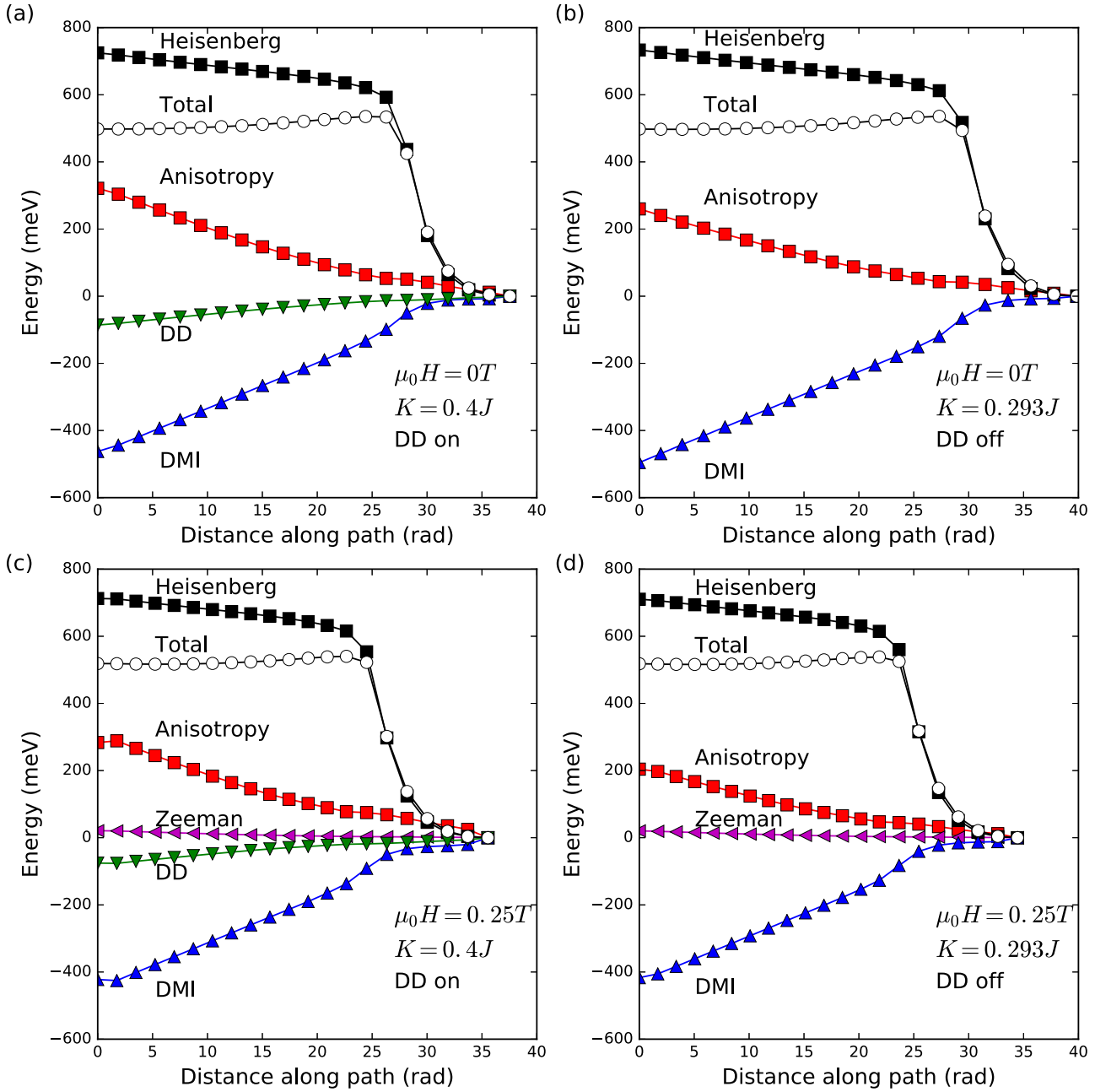

FIG. 2. The various components of the energy along the minimum energy paths: Dzyaloshinskii-Moriya interaction (DMI), dipole-dipole (DD) interaction, anisotropy, Heisenberg, and total energy. Without magnetic field: (a) including DD and full anisotropy, $K = 0.4$ meV; (b) excluding DD and smaller anisotropy constant, $K = 0.293$ meV. With a magnetic field of 0.25 T: (c) including DD and full anisotropy, $K = 0.4$ meV; (d) excluding DD and smaller anisotropy constant, $K = 0.293$ meV, as in panel (b). This shows that the reduction in the anisotropy constant can mimic closely the effect of the dipole-dipole interaction, both at zero field and at a field of 0.25 T.

Rohart *et al.* [21] also reported results of path calculations and presented two paths indicating two different transition mechanisms for the annihilation of the skyrmion. The one that resembles more closely the MEP we have found using the GNEB method has a significantly higher maximum energy, 90 meV, in zero field. The shape of the path as well as the height of the maximum turns out to be similar to the constrained path shown in Fig. 1(b). In the GNEB method, each image of the system along the path is relaxed within a hyperplane that has normal parallel to the tangent to the path. There is a single hyperplane for each image, so only one constraint appears in the minimization. In the algorithm presented in the supporting information of Ref. [21], each atom is treated independently and the tangent to the path defined separately for each spin. The number of constraints is then equal to number of spins, rather than just one constraint as in the GNEB method. The additional constraints introduced in the optimization algorithm in Ref. [21] prevent the path from converging to an MEP.

The second path reported in Ref. [21] which involves rotation of the spins near the center in addition to the isotropic shrinking has a maximum of 64 meV, which is also significantly larger than the maximum energy along the MEP

we obtained, 37 meV. When we placed the initial configuration of the images in such a way as to mimic this path, the GNEB optimization resulted in the same MEP as we obtained before. We, therefore, do not see evidence of a second MEP for the annihilation of the skyrmion in this system.

The pre-exponential factor in the Arrhenius expression for the rate can in principle also be evaluated from harmonic transition state theory [14]. Such calculations have, for example, been carried out for Fe islands of various shape and size on a W(110) substrate [28] and in analysis of hysteresis loops of a spring magnet [7]. This involves evaluating all the eigenvalues of the Hessian matrix at both the first-order saddle point and at the initial-state minimum. The Hessian matrix for this system is, however, so large, $(7 \times 10^4)^2$, that such a calculation is not straightforward (even though a few of the lowest eigenvalues can readily be evaluated using the Lanczos algorithm). Furthermore, the skyrmion can quite easily move from one site to another, so there are translational modes corresponding to nearly zero eigenvalues in the initial state which are not properly represented in the harmonic approximation to transition state theory. These zero modes need to be treated separately. At the first-order saddle point, zero modes are not present. As a result, the pre-exponential factor can be expected to have a small value. Essentially, this reflects a significantly larger entropy in the initial skyrmion state as compared with the transition state. The value extracted from the Langevin dynamics simulations of Rohart *et al.* [21], $10^9\ s^{-1}$, is indeed relatively small, for example, compared with prefactors for remagnetization transitions in Fe islands [28]. The evaluation of the pre-exponential factor from harmonic transition state theory for this system remains a challenge that requires further development of methodology. The implementation of full transition state theory and subsequent dynamical corrections, the so-called Wigner-Keck-Eyring (WKE) two-step procedure [29], would, furthermore, give estimates that are more accurate than the harmonic approximation but are computationally much more intensive.

In summary, the results presented here demonstrate that the activation energy for skyrmion annihilation can be estimated accurately by finding the MEP of the transition. In the presence of a destabilizing magnetic field of 0.25 T, the estimate from the MEP using harmonic transition state theory gives 21 meV while the direct Langevin simulations of Rohart *et al.* at relatively high temperature [21] gave 26 ± 4 meV. This demonstrates that calculations of MEPs can be useful in the design of systems and materials where an assessment of the stability of skyrmions is needed. The calculation of the MEP must, however, be carried out with full optimization [18] without additional constraints. While it is important in the present system to include the dipole-dipole interaction, a computationally intensive calculation, it is possible to effectively include its effect by reducing the anisotropy constant. This provides considerable savings in the computational effort.

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