Skyrmions with Attractive Interactions in an Ultrathin Magnetic Film

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We determined the parameters of a classical spin Hamiltonian describing an Fe monolayer on Pd(111) surface with a $Pt_{1-x}Ir_x$ alloy overlayer from *ab initio* calculations. While the ground state of the system is ferromagnetic for $x = 0.00$, it becomes a spin spiral state as Ir is intermixed into the overlayer. Although the Dzyaloshinsky-Moriya interaction is present in the system, we will demonstrate that the frustrated isotropic exchange interactions play a prominent role in creating the spin spiral state, and these frustrated couplings lead to an attractive interaction between Skyrmions at short distances. Using spin dynamics simulations, we show that under these conditions the individual Skyrmions form clusters, and that these clusters remain stable at finite temperature.

The magnetic Skyrmion corresponds to a configuration where the directions of the spin magnetic moments at different lattice sites span the whole sphere [1,2], in contrast to collinear ferromagnetic or antiferromagnetic systems and spin spiral states. Several years after the theoretical prediction [3,4], a lattice of magnetic Skyrmions was first identified in the chiral magnet MnSi [5]. Since this discovery, Skyrmions have been detected experimentally in several other bulk systems; examples include FeGe [6,7], FeCoSi [8,9], Cu_2OSeO_3 [10], GaV_4S_8 [11], and Co-Zn-Mn alloys [12].

In agreement with the original theoretical description [4,13], the appearance of Skyrmions in the above systems was attributed to the Dzyaloshinsky-Moriya interaction [14,15] present in noncentrosymmetric magnets. This chiral interaction competes with the ferromagnetic exchange and easy-axis anisotropy, and may lead to a planar spin spiral ground state in the system [16,17], which can in turn transform into a Skyrmion lattice at finite external magnetic field.

Because frustrated isotropic exchange interactions may also stabilize a spin spiral phase, Skyrmions could also be present in such systems at a finite external magnetic field, even if the Dzyaloshinsky-Moriya interaction is absent because of symmetry reasons. It was shown in Ref. [18] for a model Hamiltonian with competing ferromagnetic and antiferromagnetic interactions on a triangular lattice that, at least at finite temperature, this is indeed the case. It was demonstrated later [19–21] that the presence of an easy-axis on-site anisotropy extends the stability range of the Skyrmion lattice to zero temperature. If only isotropic exchange interactions are present, Bloch-type and Néel-type Skyrmions with different helicities, as well as Skyrmions and anti-Skyrmions with opposite topological charges [19], are energetically degenerate. Furthermore, the magnetization profile of Skyrmions with frustrated exchange interactions is different from that of Skyrmions stabilized by the Dzyaloshinsky-Moriya interaction. This leads to an interaction potential between Skyrmions with several local energy minima, while the interaction between Dzyaloshinsky-Moriya Skyrmions is repulsive at all distances at low temperatures [22].

Magnetic Skyrmions have also been explored in ultrathin film systems such as a PdFe bilayer [23] or an Fe triple layer [24] on an Ir(111) surface, and Pt-Co-Ir multilayers [25]. Because bulk inversion symmetry is broken at the surface, the Dzyaloshinsky-Moriya interaction is present in such systems; consequently, the theoretical descriptions [26–29] so far have been based on the conventional model [3,4]. On the other hand, several recent publications [26,30,31] have identified the frustrated isotropic exchange interactions as the driving mechanism behind the creation of spin spiral ground states in specific ultrathin films.

Because their size is in the nanometer regime and they can be manipulated by relatively weak spin-polarized currents [32,33], Skyrmions are promising candidates for future applications in data storage and logic devices [34–36]. At finite temperature, isolated Skyrmions propagate diffusively on the field-polarized background [37], and their uncontrolled motion leads to a loss of information in memory devices. It has been demonstrated in simulations

[32,38] and experiments [39] that it is possible to control this diffusive motion by lattice defects.

In this Letter, we have performed *ab initio* calculations on a $(Pt_{1-x}Ir_x)$Fe bilayer system on Pd(111) surface to determine the coupling coefficients in a classical Hamiltonian. We will demonstrate using Landau-Lifshitz-Gilbert [40] spin dynamics simulations that individual Skyrmions may be stabilized in the collinear field-polarized state of the system under experimentally realizable external magnetic fields. The Dzyaloshinsky-Moriya interaction is responsible for determining the helicity of Skyrmions, while the frustrated exchange interactions modify their shape and lead to an oscillating Skyrmion-Skyrmion interaction potential. Our simulations provide evidence that the short-range attractive interaction pins the Skyrmions next to each other, and that the Skyrmion clusters formed are resistant against diffusion processes at finite temperature.

The classical Hamiltonian describing the magnetic moments in the Fe layer reads

$$H = \frac{1}{2}\sum_{i\neq j}S_i\mathcal{J}_{ij}S_j + \sum_i S_i\mathcal{K}S_i - \sum_i MS_iB, \quad (1)$$

where the unit vectors S_i represent the spins and B denotes the external magnetic field. The exchange coupling tensors $\mathcal{J}_{ij}$ and the on-site anisotropy tensors $\mathcal{K}$, as well as the magnetic moment M in Eq. (1), have been determined by combining the screened Korringa-Kohn-Rostoker method [41,42] with the relativistic torque method [43]. For the details of the calculations see the Supplemental Material [44]. The isotropic exchange interactions $J_{ij} = \frac{1}{3}\mathrm{Tr}\mathcal{J}_{ij}$ represent scalar Heisenberg couplings between the spins; the antisymmetric parts of the coupling tensors $D_{ij}^\alpha = \frac{1}{2}\varepsilon^{\alpha\beta\gamma}\mathcal{J}_{ij}^{\beta\gamma}$ can be identified with the Dzyaloshinsky-Moriya vectors [27]. In the sign convention of Eq. (1), $J_{ij} < 0$ describes ferromagnetic coupling between the spins, while $J_{ij} > 0$ is antiferromagnetic.

The J_{ij} isotropic exchange interactions are depicted in Fig. 1(a). By partially replacing Pt with Ir in the nonmagnetic overlayer, the magnitude of the nearest-neighbor ferromagnetic exchange interaction decreases, though this does not considerably influence the antiferromagnetic interactions with the second and third neighbors. This means that decreasing the average number of valence electrons in the overlayer drives the system from the ferromagnetic towards the spin spiral state, in agreement with the results of Ref. [31] for a similar layered system.

In order to determine the ground state of the system, we have calculated the energies $E_{SS}(q)$ of harmonic spin spirals with wave vector q, and compared them to the energy E_{FM} of the ferromagnetic state along the easy out-of-plane direction. The results are summarized in Fig. 1(b). For the calculations we have chosen right-handed cycloidal spin spirals,
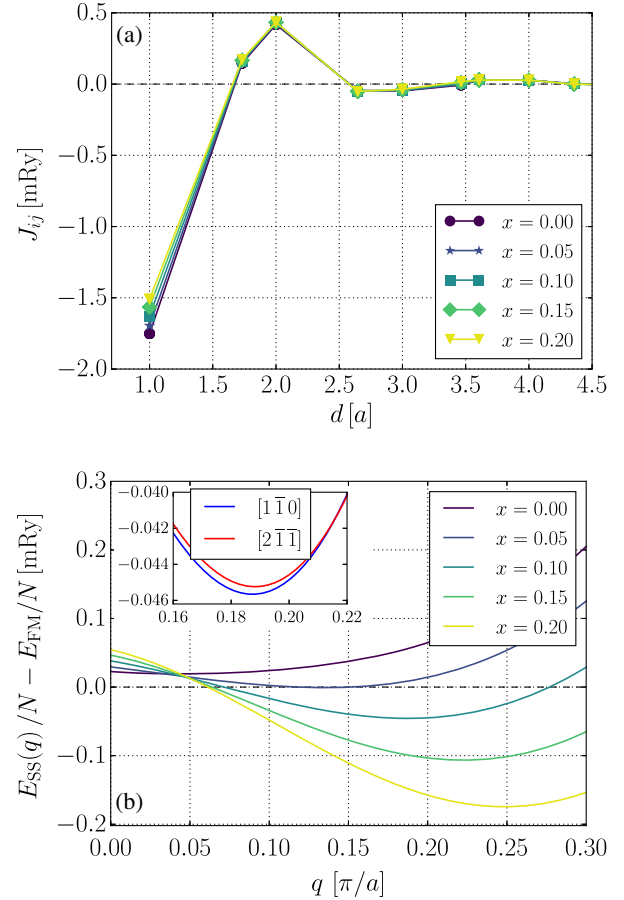

FIG. 1.   (a) Calculated isotropic exchange interactions J_{ij} in a $(Pt_{1-x}Ir_x)$Fe bilayer on Pd(111) as a function of the distance d between the Fe atoms, given in lattice constants of the triangular lattice a. (b) Energy per spin in the spin spiral state $E_{SS}(q)/N$ as a function of the wave vector along the $[1\bar{1}0]$ direction, relative to the ferromagnetic state E_{FM}/N. Inset shows a slight anisotropy favoring the $[1\bar{1}0]$ axis over the $[2\bar{1}\bar{1}]$ direction for $x = 0.10$.

$$S_i = \frac{q}{|q|}\sin(qR_i) + n\cos(qR_i), \quad (2)$$

where n is the outwards-pointing normal vector of the bilayer. Because the frustrated isotropic exchange interactions do not influence the rotational plane of the spiral, the energetically preferred right-handed cycloidal sense was determined by the Dzyaloshinsky-Moriya interactions, in agreement with the C_{3v} symmetry of the surface [4]. Because of the anisotropy of the system, $E_{SS}(q) - E_{FM}$ does not converge to zero for harmonic spin spirals as $q \to 0$, but it approximates well the energy of the anharmonic spin spirals at finite wave vectors [30].

Although the ground state of the system is out-of-plane ferromagnetic for a pure Pt overlayer $x = 0.00$, the minimum of the spin spiral dispersion relation is below E_{FM} at the higher Ir concentrations displayed in Fig. 1(b). While the in-plane components of the Dzyaloshinsky-Moriya vectors prefer the creation of spin spiral states, they weaken

with increasing Ir concentration; for numerical values see the Supplemental Material [44]. This means that in the system considered here the increasing frustration of the isotropic exchange interactions, shown in Fig. 1(a), is responsible for the creation of the spin spiral. For small wave vectors, the anisotropy of the lattice only has a weak effect on the directional dependence of $E_{SS}(\boldsymbol{q})$ [21]. However, we note that spirals with wave vectors along $[1\bar{1}0]$ (the nearest neighbors in real space) are slightly preferred over ones with wave vectors along $[2\bar{1}\bar{1}]$ (the next-nearest neighbors), as shown in the inset of Fig. 1(b).

By applying an external magnetic field perpendicularly to the surface, the system will eventually transform into a collinear field-polarized state, possibly going through a Skyrmion lattice phase for intermediate field values. We have observed localized noncollinear magnetic field configurations in the collinear phase by performing spin dynamics simulations. Because of the Dzyaloshinsky-Moriya interaction, Skyrmions with topological charge $Q = -1$ are energetically the most favorable [1], if the magnetization of the collinear state is pointing outwards from the surface. We calculated the interaction energy between two such Skyrmions from numerical simulations, as illustrated in Fig. 2. During the simulations, we fixed the spin at the center of the Skyrmions to be antiparallel to the magnetization of the collinear state, and found the energy minimum with this constraint by the numerical solution of the Landau-Lifshitz-Gilbert equation. For $x \geq 0.05$, we found that the interaction energy oscillates while decaying. However, only the first local minimum is well visible in Fig. 2(a) due to the exponential decay [19,20]. The presence of the local minima is clearly a consequence of the frustrated isotropic exchange interactions, since the Dzyaloshinsky-Moriya interaction prefers Skyrmions that repulse each other at all distances; we found this to be the case for a pure Pt overlayer ($x = 0.00$), where the frustration of the interactions is the weakest. Note that the minimum is deeper for Skyrmions separated along the $[2\bar{1}\bar{1}]$ direction compared to the $[1\bar{1}0]$ direction; this means that the preferred direction of nearest-neighbor bonds between Skyrmions is perpendicular to the wave vector minimizing the spin spiral energies in Fig. 1(b).

Figure 2(b) demonstrates that the oscillation in the interaction energy is accompanied by an oscillation of the spin components in real space [19,20,61]. In the local minimum of the interaction potential, the Skyrmions form a bond with the same sign of the in-plane spin component in the overlapping regime. On the other hand, the shape of Skyrmions created by the Dzyaloshinsky-Moriya interaction can be well approximated by two domain walls located next to each other [62], where the only sign change in the in-plane spin component is at the center of the Skyrmion. The frustrated exchange interactions create further local extrema of the in-plane spin components where the rotational sense of the spins switches from right-handed to
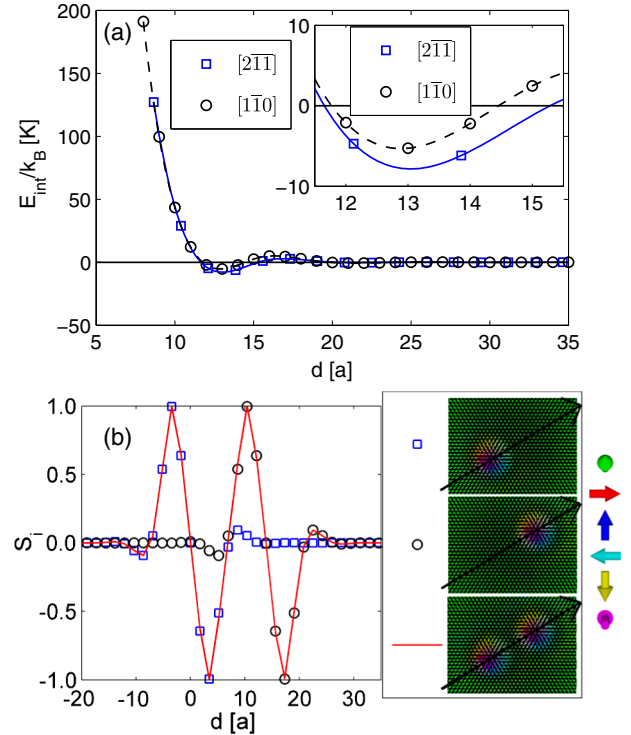

FIG. 2. Attractive interaction between Skyrmions for $x = 0.10$ and $B = 4.22$ T. The field-polarized state represents the ground state for $B > 4.18$ T [44]. (a) Interaction energy between two Skyrmions along the $[2\bar{1}\bar{1}]$ and $[1\bar{1}0]$ directions. Squares and circles denote data points; the interpolated lines are guides to the eye. Inset shows a close-up of the first minimum. (b) Real-space spin configuration of two isolated Skyrmions (squares, circles) and two interacting Skyrmions (line), with the centers of the Skyrmions at the same lattice points in the two cases. S_i is the projection of the vector $\boldsymbol{S}_i$ on the $[2\bar{1}\bar{1}]$ line connecting the centers, shown by arrows in the legend. The color bar describes the orientation of the spins in the legend and in Fig. 3.

left-handed (helicity reversal [19]), which is energetically unfavorable from the standpoint of the Dzyaloshinsky-Moriya interactions. This means that the antiferromagnetic isotropic exchange interactions with the second and third neighbors are competing not only with the ferromagnetic nearest-neighbor interaction, but also with the Dzyaloshinsky-Moriya interactions, in order to form bonds between the Skyrmions.

Because the oscillating interaction potential determines an energetically favorable bond length between the Skyrmions, it is possible to arrange them into arbitrarily shaped clusters at zero temperature. One example is displayed in Fig. 3(a). As shown in Fig. 3(b), the initial configuration is mostly conserved during simulations performed at $T = 4.7$ K, indicating thermal stability. For comparison, Fig. 3(c) demonstrates how the information encoded in the original state is lost due to the diffusive motion and repulsive interaction between Skyrmions with the system parameters $x = 0.00$, $B = 0.00$ T.
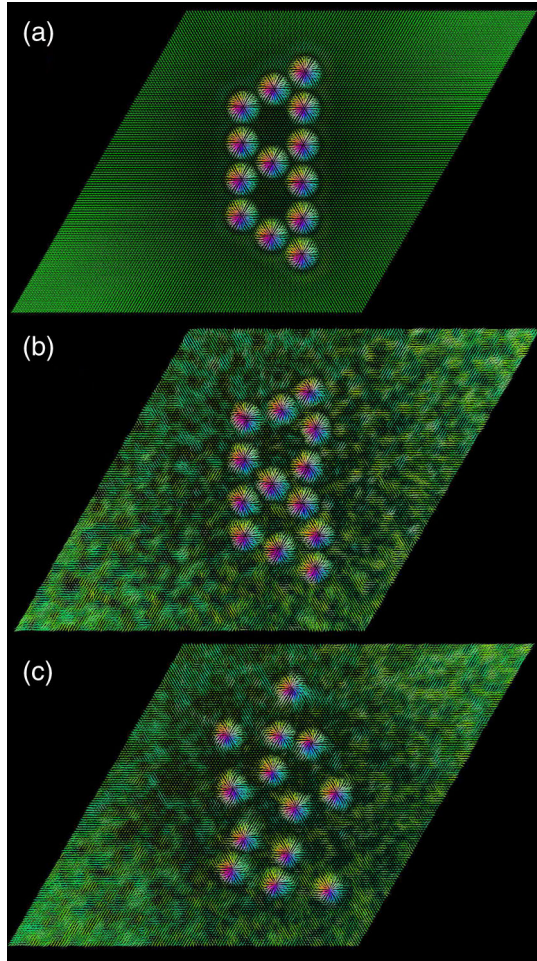
FIG. 3. (a) Ordered initial configuration of Skyrmions at $T = 0$ K, for $x = 0.10$ and $B = 4.22$ T. (b),(c) Final configuration after a thermalization of $t = 605$ ps at $T = 4.7$ K, for different coupling parameters: (b) attractive Skyrmions at $x = 0.10$ and $B = 4.22$ T, (c) repulsive Skyrmions at $x = 0.00$ and $B = 0.00$ T. The lattice size is $N = 128 \times 128$ atoms.

The attractive interaction between Skyrmions at finite temperature can also be characterized by calculating the pair correlation function $f(r)$, normalized as

$$\int_0^\infty f(r) 2\pi r \, dr = 1. \qquad (3)$$

Figure 4 displays the pair correlation function after thermalization. We considered an initial configuration of 31 Skyrmions in random arrangement on an $N = 128 \times 128$ lattice, the same size as in Fig. 3; approximately 80 Skyrmions would fit into the same lattice size in a close-packed configuration with the applied simulation parameters. Figure 4(a) shows that the distribution is basically uniform in space outside the strongly repulsive core for repulsive Skyrmions ($x = 0.00$, $B = 0.00$ T), indicating that the diffusive motion is dominating in this

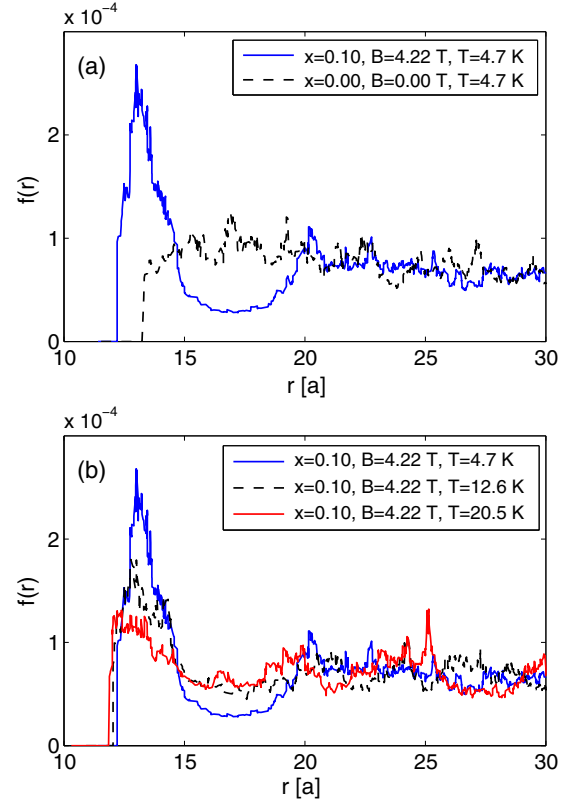

FIG. 4. Pair correlation function $f(r)$ of Skyrmions after a thermalization of $t = 484$ ps. (a) Difference between repulsive ($x = 0.00$, $B = 0.00$ T) and attractive ($x = 0.10$, $B = 4.22$ T) Skyrmions at $T = 4.7$ K. (b) Temperature dependence of $f(r)$ for $x = 0.10$ and $B = 4.22$ T. The system contained 31 Skyrmions on an $N = 128 \times 128$ atomic lattice.

case. On the other hand, one can clearly identify a preferred nearest-neighbor distance for attractive Skyrmions ($x = 0.10$, $B = 4.22$ T) around $r \approx 13a$, coinciding with the potential energy minimum in Fig. 2(a). This favors the formation of clusters, similar to the artificially created one in Fig. 3(b). The normalization of $f(r)$ is ensured by a decreased number of Skyrmions in the ring between $15a$ and $20a$. It is shown in Fig. 4(b) that when the temperature becomes slightly higher ($T \approx 20$ K) than the energy barrier protecting the local minimum in Fig. 2(a) ($|E_{\mathrm{int}}/k_B| \approx 8$ K), the peak in the distribution function disappears, and the clusters are destroyed by thermal fluctuations.

To summarize, we have demonstrated that isolated magnetic Skyrmions may be stabilized in a $(\mathrm{Pt}_{1-x}\mathrm{Ir}_x)$Fe bilayer on Pd(111). The frustrated isotropic exchange interactions create an oscillating Skyrmion-Skyrmion interaction potential, at the expense of the Dzyaloshinsky-Moriya interactions which prefer repulsion at all distances. Because of the attractive interaction, the Skyrmions may be arranged into clusters. The bonds between the Skyrmions stabilize their relative positions at finite temperature, which may be important for future applications in memory devices.

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