

Spin-polarized scanning tunneling microscopy characteristics of skyrmionic spin structures exhibiting various topologies

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The correct identification of topological magnetic objects in experiments is an important issue. In the present paper we report on the characterization of metastable skyrmionic spin structures with various topological charges ($Q = -3, -2, -1, 0, +1, +2$) in the $(\text{Pt}_{1-x}\text{Ir}_x)\text{Fe}/\text{Pd}(111)$ ultrathin magnetic film by performing spin-polarized scanning tunneling microscopy (SP-STM) calculations. We find that an out-of-plane magnetized tip already results in distinguished SP-STM contrasts for the different skyrmionic structures corresponding to their symmetries. Our paper also establishes an understanding of the relationship between in-plane SP-STM contrasts and skyrmionic topologies through an investigation of the variation of the in-plane angle between the spins along the perimeter of the structures, which can be characterized by the local vorticity or linear density of the winding number. For spin structures exhibiting a uniform sign of the local vorticity throughout the whole skyrmionic area, we demonstrate that (i) $|Q|$ can be determined from a single SP-STM image taken by any in-plane magnetized tip and (ii) an in-plane tip magnetization rotation provides the sign of Q independently of the sign of the effective spin polarization in the tunnel junction. We also discuss cases where the local vorticity is changing sign. Finally, by increasing the Ir content of the PtIr overlayer, we find an appearing secondary outer ring in-plane SP-STM contrast that is indicative of attractive skyrmions or antiskyrmions.

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I. INTRODUCTION

Magnetic skyrmions correspond to specific spin configurations in magnetic materials characterized by a finite topological charge [1]. Skyrmions may order into a hexagonal lattice and represent a stable thermodynamic phase [2], the presence of which has been experimentally observed in a wide array of materials in bulk or thin-film form [3–11]. Skyrmions may also appear as localized metastable states in the collinear phase of magnets [12]. This property turns them into ideal candidates as bits of information in future technological applications [13,14]. The experimental observation of such isolated skyrmions is primarily connected to ultra-thin-film systems [15–18]. The understanding of skyrmion formation in these materials is also supported by computational efforts ranging from *ab initio* calculations determining the interaction parameters [19–23] to studies focusing on finite-temperature effects regarding the skyrmion stability [24–27].

As real-space spin structures, isolated skyrmions can conveniently be imaged by using spin-polarized scanning tunneling microscopy (SP-STM) [28,29]. The controlled manipulation (creation and annihilation) of isolated skyrmions using an SP-STM tip has been demonstrated in Refs. [16,17], which may be the key for writing and deleting information in magnetic media in future applications.

Most experimental observations of skyrmions in ultrathin films so far are connected to systems with strong Dzyaloshinsky-Moriya interactions [30,31], where all skyrmions possess the same topological charge [12]. In comparison, it was recently demonstrated that frustrated Heisenberg exchange interactions may lead to the stabilization of localized spin configurations with different topological charges [32–34]. Investigating the SP-STM images of such structures is also worthwhile to consider. Based on *ab initio* calculations performed for a Pd/Fe bilayer on the Ir(111) surface, Dupé *et al.* [35] reported SP-STM contrast characteristics for a set of metastable skyrmionic structures, where they identified circular contrasts using an out-of-plane magnetized tip and two types of contrasts employing an in-plane magnetized tip: (i) a two-lobes contrast for spin structures with topological charge $|Q| = 1$ and (ii) a four-lobes contrast for a higher-order antiskyrmion with $|Q| = 2$. In Ref. [36], other types of metastable skyrmionic structures were also investigated in the $(\text{Pt}_{1-x}\text{Ir}_x)\text{Fe}/\text{Pd}(111)$ ultrathin magnetic film, and it was discussed how their shapes become distorted due to the interplay between the Dzyaloshinsky-Moriya and the frustrated Heisenberg exchange interactions. These observations enable the generalization of the findings of Dupé *et al.* [35] for the SP-STM contrasts of topologically distinct skyrmionic structures.

By employing SP-STM calculations on the metastable skyrmionic spin structures with various topological charges ($Q = -3, -2, -1, 0, +1, +2$) taken from Ref. [36], our paper

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establishes a connection between SP-STM contrasts and skyrmionic topologies, most importantly through an investigation of the variation of the in-plane angle between the spins along the perimeter of the structures, which can be characterized by the local vorticity or linear density of the winding number. Our findings are expected to be applicable to the topological characterization of skyrmionic spin structures based on experimentally measured SP-STM images using a series of in-plane tip magnetization orientations. In ideal cases characterized by a uniform sign of the local vorticity throughout the whole skyrmionic area, we propose that the magnitude and the sign of Q of the topological object can be determined. A measured series of in-plane SP-STM contrasts are also expected to help in identifying spin structures if the local vorticity is not uniform. Such a scenario is expected in anisotropic environments of skyrmion formation, e.g., in reconstructed or confined film geometries [17,37], where arbitrary skyrmionic shapes with complex domain-wall structures can be found.

Besides stabilizing skyrmionic structures with different topological charges, frustrated Heisenberg exchange interactions also modify the shape of isolated skyrmions with $Q = -1$. The characteristic feature is an oscillation of the spin around the direction of the background magnetization [33], which shows up as a sequence of sign changes in the in-plane spin component [38]. This oscillation indicates a short-range attractive interaction between skyrmions or antiskyrmions. In

our SP-STM calculations, the sign changes of the in-plane spin component appear as a secondary outer ring in the contrast. This contrast feature can be tuned by the Ir content of the PtIr overlayer, which affects the exchange interactions in the Fe layer [38].

The paper is organized as follows. In the Theory section the employed SP-STM calculation method and tunneling parameters are described, and the topological charge and the vorticity of skyrmionic structures in magnetic films are defined, providing also a theoretical connection between SP-STM contrasts and the local vorticity. After that, calculated SP-STM images are presented and discussed, and conclusions are drawn on the relationship between the SP-STM contrast and the skyrmionic topology as well as on the SP-STM detection of attractive skyrmionic objects.

II. THEORY

A. SP-STM

For the calculation of the SP-STM images of the skyrmionic spin structures, the three-dimensional (3D) Wentzel-Kramers-Brillouin (WKB) approximation [39] of electron tunneling has been employed, where the tunneling current at the tip apex position \mathbf{R}_{tip} and bias voltage V is calculated as the superposition of one-dimensional WKB contributions from the sample surface atoms (sum over a) as [40]

$$I(\mathbf{R}_{\text{tip}}, V) = \epsilon^2 \frac{e^2}{h} \sum_a \int_0^V dU \exp \left[-\sqrt{\frac{8m}{\hbar^2} \left(\frac{\Phi_S + \Phi_T + eV}{2} - eU \right)} |\mathbf{R}_{\text{tip}} - \mathbf{R}_a| \right] \times n_S^a(E_F^S + eU) n_T(E_F^T + eU - eV) [1 + P_S^a(E_F^S + eU) P_T(E_F^T + eU - eV) \cos \phi_a]. \quad (1)$$

Here, the exponential factor describes the tunneling transmission, where all electron states are assumed as exponentially decaying spherical states [41–43] with an effective rectangular potential barrier in the vacuum between the sample and the tip. The electronic structures enter the model by considering $n_{S(T)}$ the atom-projected charge density of states and $P_{S(T)}$ the spin polarization of the sample surface (S) and the tip apex (T). ϕ_a is the angle of the localized magnetic moment of surface atom a with respect to the tip magnetization direction, e is the elementary charge, \hbar is the (reduced) Planck constant, m is the electron's mass, and $\Phi_{S(T)}$ and $E_F^{S(T)}$ denote the electron work function and the Fermi energy of the sample surface (tip), respectively. The $\epsilon^2 e^2/h$ factor ensures the correct dimension of the current. The value of ϵ has to be determined by comparing the calculated results of the charge current with experiments, or with calculations using standard methods, e.g., the Bardeen approach [44]. In our calculations $\epsilon = 1$ eV has been chosen [45] that gives comparable current values with those obtained by the Bardeen method implemented in the BSKAN code [46,47]. Note that the choice of ϵ has no qualitative influence on the reported SP-STM contrasts and conclusions.

In the present paper, SP-STM images correspond to constant-current surfaces calculated at the bias voltage

$V = 0$ V, where Eq. (1) takes the form [43]

$$I(\mathbf{R}_{\text{tip}}) \propto \sum_a \exp \left[-\sqrt{8m\Phi/\hbar^2} |\mathbf{R}_{\text{tip}} - \mathbf{R}_a| \right] \times [1 + P_S P_T \cos \phi_a], \quad (2)$$

assuming $P_S^a = P_S$ for all surface atoms and $\Phi = \Phi_S = \Phi_T$. Motivated by a recent work [35], we choose the effective spin polarization of $P_{\text{eff}} = P_S P_T = \pm 0.4$ and consider the effect of its sign on the SP-STM contrasts. Electron work functions of $\Phi_S = \Phi_T = 5$ eV were taken. With the selected parameters, the current value $I = 10^{-4}$ nA of the constant-current surfaces corresponds to about 6-Å minimal tip-sample distance and corrugation values between 30 and 40 pm. Note that smaller corrugation values found in an experiment [29] are either due to a different P_{eff} magnitude or to a larger tip-sample separation in the experiment, for a theoretical explanation of the latter effect see, e.g., Ref. [48]. P_{eff} also plays a crucial role in spin-polarized scanning tunneling spectroscopy [49].

B. Skyrmionic topology

For the characterization of the observed isolated skyrmionic objects, we rely on the topological charge Q , which expresses how many times the spin vectors span the whole unit sphere.

Q is defined as

$$Q = \frac{1}{4\pi} \int \mathbf{S} \cdot (\partial_x \mathbf{S} \times \partial_y \mathbf{S}) dx dy, \quad (3)$$

where $\mathbf{S} = [\sin \Theta \cos \Phi, \sin \Theta \sin \Phi, \cos \Theta]$ is the unit vector of local magnetization and the integral has to be performed over the area of the localized spin structure in the surface (xy) plane. The integral can be transformed to a form considering surface polar coordinates (r, φ) [1,25]:

$$Q = \frac{1}{4\pi} \int_0^\infty \int_0^{2\pi} (\partial_r \Theta \partial_\varphi \Phi - \partial_\varphi \Theta \partial_r \Phi) \sin \Theta d\varphi dr. \quad (4)$$

It was demonstrated in Ref. [36] that due to the presence of the Dzyaloshinsky-Moriya interaction in the system not all of the observed skyrmionic objects possess a circular shape. The integral over φ in Eq. (4) may be performed along the contour lines $r(\varphi)$ of Θ , defined as being perpendicular to the gradient at all points:

$$\frac{dr(\varphi)}{d\varphi} = -\frac{\partial_\varphi \Theta}{\partial_r \Theta}. \quad (5)$$

Here we only consider single-domain skyrmionic structures, where $\partial_r \Theta$ remains nonzero in the whole considered configuration, and all the contours still only wind once around the origin. By introducing the local vorticity along the contour lines,

$$\mathcal{M}(r, \varphi) = \frac{d\Phi(r, \varphi)}{d\varphi} = \partial_\varphi \Phi + \partial_r \Phi \frac{dr(\varphi)}{d\varphi}, \quad (6)$$

and substituting into Eq. (4) one arrives at the expression

$$\begin{aligned} Q &= \frac{1}{2} \int_0^\infty \left[\frac{1}{2\pi} \int_0^{2\pi} \mathcal{M}(r, \varphi) d\varphi \right] \sin \Theta \partial_r \Theta dr \\ &= -\frac{1}{2} [\cos \Theta(r)]_0^\infty m, \end{aligned} \quad (7)$$

where the vorticity is defined as

$$m = \frac{1}{2\pi} \int_0^{2\pi} \mathcal{M}(r, \varphi) d\varphi. \quad (8)$$

The integral on the right-hand side of Eq. (8) gives an integer, expressing how many times and in which direction the in-plane component of the spins rotates around the circle. Since there are no topological defects in the system, the value of m does not depend on the choice of the contour line, and is actually the same when performing the integral along an arbitrary closed curve in the surface plane enclosing the center of the localized spin configuration.

Finally, the relationship between the local vorticity \mathcal{M} and the topological charge density \mathcal{Q} is

$$\mathcal{Q}(r, \varphi) = \frac{1}{4\pi r} \partial_r \Theta \sin \Theta \mathcal{M}(r, \varphi). \quad (9)$$

Since $\partial_r \Theta$ is negative for the skyrmionic spin structures considered in this paper, i.e., $\Theta(r=0) = \pi$ and $\Theta(r=\infty) = 0$, the signs of the topological charge density and the local vorticity are the opposite,

$$\text{sign} \mathcal{Q}(r, \varphi) = -\text{sign} \mathcal{M}(r, \varphi), \quad (10)$$

and, correspondingly, the relation between the topological charge and the vorticity is $Q = -m$ [36].

C. Relation between SP-STM and skyrmionic topology

In SP-STM with an in-plane magnetized tip of the orientation $\mathbf{e}_{\text{tip}} = (\cos \varphi_{\text{tip}}, \sin \varphi_{\text{tip}}, 0)$, the tunneling current is proportional to [cf. Eq. (2)]

$$I(\mathbf{R}_{\text{tip}}) \propto \mathbf{S} \cdot \mathbf{e}_{\text{tip}} = \sin \Theta \cos(\Phi - \varphi_{\text{tip}}). \quad (11)$$

Suppose that we select a single point on the constant-current surface (SP-STM image) with a current value of $I(\mathbf{R}_{\text{tip}})$, and by infinitesimally rotating the tip magnetization by $d\varphi_{\text{tip}}$ we follow the trajectory of the point on the constant-current surface along the $r(\varphi)$ contour of Θ . This procedure can mathematically be expressed as

$$\begin{aligned} dI(\mathbf{R}_{\text{tip}}) &\propto \mathcal{M}(r, \varphi) d\varphi - d\varphi_{\text{tip}} = 0, \\ \frac{d\varphi}{d\varphi_{\text{tip}}} &= \mathcal{M}^{-1}(r, \varphi). \end{aligned} \quad (12)$$

Equation (12) means that the angular velocity $d\varphi/d\varphi_{\text{tip}}$ of contrast features in constant-current images obtained by in-plane tip magnetization rotation equals to the inverse of the local vorticity. This enables the direct extraction of information in SP-STM experiments on the local vorticity $\mathcal{M}(r, \varphi)$ or on the topological charge density $\mathcal{Q}(r, \varphi)$ using the proportionality relation in Eq. (9).

III. RESULTS AND DISCUSSION

Figure 1 shows a set of metastable skyrmionic spin configurations obtained in a $(\text{Pt}_{0.95}\text{Ir}_{0.05})\text{Fe}/\text{Pd}(111)$ ultrathin magnetic film, where attractive skyrmions have recently been reported [38]. Formation and stability of the metastable structures are extensively discussed in Ref. [36]. Note that the configurations shown in Figs. 1(a)–1(c) (first row) have also been reported by Dupé *et al.* [35]. In the following, we focus on the comparison of our obtained SP-STM contrasts with the work of Dupé *et al.*, and also generalize some of their findings based on our extended set of results on higher-order skyrmions. The

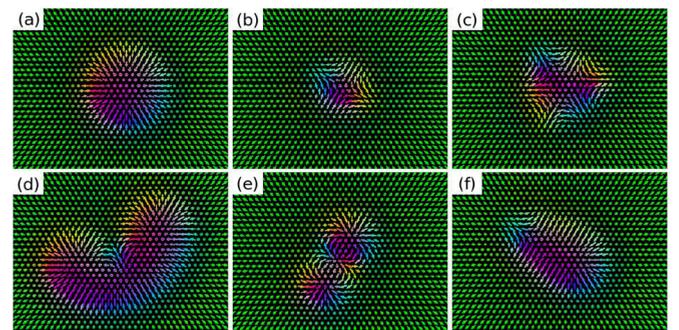


FIG. 1. Metastable localized skyrmionic spin configurations with different topological charges in the $(\text{Pt}_{0.95}\text{Ir}_{0.05})\text{Fe}/\text{Pd}(111)$ ultrathin magnetic film [36]: (a) skyrmion with $Q = -1$, (b) antiskyrmion with $Q = 1$, (c) antiskyrmion with $Q = 2$, (d) skyrmion with $Q = -2$, (e) skyrmion with $Q = -3$, and (f) chimeric skyrmion with $Q = 0$. The value of the external field is $B = 0.23$ T (a–d, f) and $B = 2.35$ T (e); the ground state is field polarized for $B > 0.21$ T.

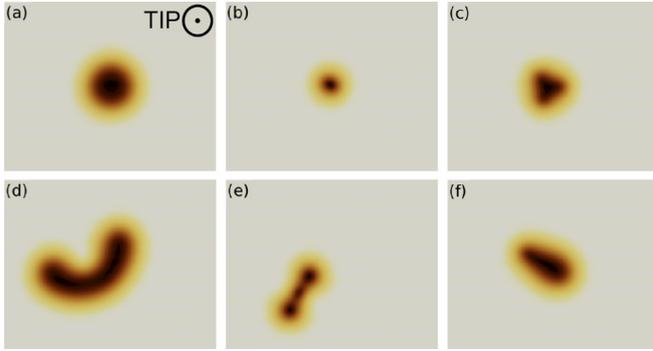


FIG. 2. Calculated SP-STM images of the set of skyrmionic spin configurations shown in Fig. 1 using an out-of-plane magnetized tip [pointing to the $+z$ [111] direction as illustrated in (a)] with $P_{\text{eff}} = +0.4$. The color scale and image areas are the same for all skyrmionic structures.

characterization of the skyrmionic structures is performed by SP-STM calculations considering different fixed tip magnetization orientations, which are sensitive to the in-line orientation of the local magnetization of the complex surface spin structures, causing the magnetic contrast depending on the sign of P_{eff} . We note that the setting of an arbitrary tip magnetization orientation through a 3D vector field is possible in SP-STM experiments according to Ref. [50].

SP-STM images using an out-of-plane magnetized tip (pointing to the $+z$ [111] direction) are shown in Fig. 2. In case of a positive P_{eff} the ferromagnetic background provides a bright contrast, and the skyrmionic spin structures are imaged as dark regions exhibiting different shapes. The latter finding is in striking difference with Ref. [35]. Strictly speaking, we find a circular contrast for the skyrmion with $Q = -1$ [Fig. 2(a)] only. This circular contrast is distorted for the antiskyrmions: $Q = 1$ shows slightly elongated contrast along a specific axis [Fig. 2(b)], and $Q = 2$ shows a rounded triangular contrast [Fig. 2(c)]. The higher-order skyrmions with $Q = -2$ and -3 and the chimera skyrmion with $Q = 0$ show noncircular contrasts with an out-of-plane magnetized tip [Figs. 2(d)–2(f)]. These observed SP-STM contrasts are in correspondence with the symmetry (cylindrical for $Q = -1$ and $C_{|1+Q|}$ for $Q \neq -1$), alignment, and distortion of the real-space spin structures, discussed in Ref. [36]. Note that the SP-STM contrasts reported in Fig. 2 are reversed using an out-of-plane magnetized tip pointing to the $-z$ direction keeping the sign of P_{eff} , or keeping the tip magnetization direction in $+z$ and reversing the sign of P_{eff} .

We observe a wider variety of SP-STM contrasts when the tip magnetization is changed from out of plane to in plane. SP-STM images using an in-plane magnetized tip (pointing to the $+x$ [110] direction) are shown in Fig. 3. The two-lobes contrast in Figs. 3(a) and 3(b) has been experimentally observed [29] and the four-lobes contrast in Fig. 3(c) has been calculated [35] earlier. The other spin structures provide more complicated contrast patterns using an in-plane magnetized tip [Figs. 3(d)–3(f)]. In general, we find the following trend: the number of bright and dark spots is equal for each case shown in Figs. 3(a)–3(e) with an alternating order along the perimeter of the skyrmionic structures, and the number of one type of spots

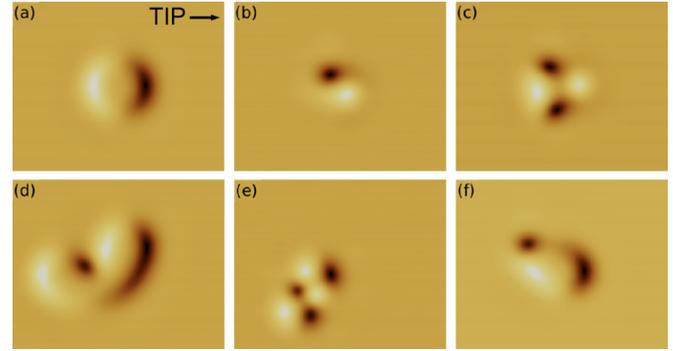


FIG. 3. Same as Fig. 2 using an in-plane magnetized tip [pointing to the $+x$ [110] direction as illustrated in (a)] with $P_{\text{eff}} = +0.4$.

(bright or dark) equals the absolute value of the topological charge, $|Q|$. This can be understood from the definition of the vorticity in Eq. (8), which counts how many times the spins are rotated in the plane when passing around the perimeter of the skyrmionic structure once. Given the fixed tip magnetization direction, this selects the number $|Q|$ of bright and dark contrast regions corresponding to parallel and antiparallel alignments of the surface spin structure with respect to the magnetization direction of the tip (for $P_{\text{eff}} > 0$). To provide a deeper insight, Fig. 4 shows calculated topological charge densities of all considered skyrmionic spin structures. Using the negative proportionality of the local vorticity and the topological charge density in Eq. (10), we find uniform sign of the local vorticities in each of Figs. 4(a)–4(e). This also means that the in-plane rotation of the spins along the perimeter of each skyrmionic structure in Figs. 1(a)–1(e) is, respectively, in the same direction.

A special case is presented in Figs. 1–4(f): the chimera skyrmion with $Q = 0$. In Fig. 3(f) one bright and two dark spots are observed because the in-plane rotation of the spins along the perimeter is changing direction [see the in-plane spin components in Fig. 1(f)] providing local vorticities of opposite sign at the skyrmionic (lower right) and antiskyrmionic (upper left) parts of the chimera skyrmion [see Fig. 4(f)], resulting in total in zero Q . We return to this case after studying the

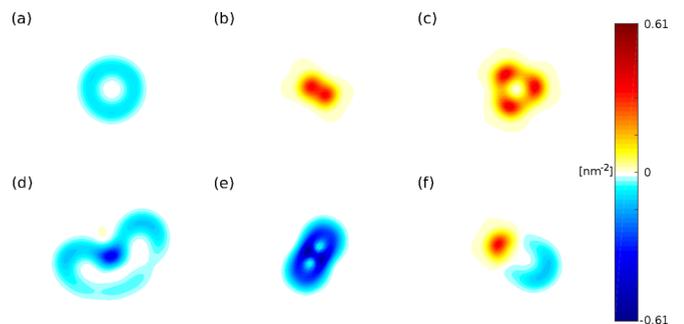


FIG. 4. Topological charge densities (\mathcal{Q}) of the set of skyrmionic spin configurations shown in Fig. 1. Red and blue colors, respectively, denote positive and negative signs. Note that the sign of the local vorticity (\mathcal{V}) is the opposite of the sign of the topological charge density in the given spin structures; see Eq. (10).

effect of the tip magnetization rotation on the in-plane SP-STM contrasts of the various skyrmionic structures.

Figures 5(a) and 5(b) show SP-STM images of the skyrmion in Fig. 1(a) with $Q = -1$ and of the antiskyrmion in Fig. 1(b) with $Q = 1$, respectively, by rotating the tip magnetization direction (black arrows) in the surface plane indicated by gray circular arrows. The contrast rotation is illustrated by red circular arrows for the two cases. Upon rotating the tip magnetization direction in steps of $\Delta\varphi_{\text{tip}} = 45^\circ$, we find that the contrast maximum and minimum always rotate by $\Delta\varphi = 45^\circ$ for the skyrmion [Fig. 5(a)]. Hence the two-lobes contrast rotates codirectionally in phase with the tip magnetization rotation for the skyrmion, in agreement with Ref. [35]. According to Eq. (12), this corresponds to a constant value of $\mathcal{M} = 1$ along the path of the contrast rotation. For the antiskyrmion in Fig. 5(b), by rotating the tip magnetization direction from $\varphi_{\text{tip}} = 0$ to 360° in steps of $\Delta\varphi_{\text{tip}} = 45^\circ$ (i.e., starting from the center right subfigure and following the anticlockwise rotation of the gray circular arrow), the contrast maximum and minimum rotate in order by $\Delta\varphi = -50, -60, -40, -30, -50, -60, -40$, and -30° . This corresponds to an antidirectional two-lobes contrast rotation, in agreement with Ref. [35]. According to Eq. (12), \mathcal{M} varies between $-1.50 (= 45^\circ / -30^\circ)$ and $-0.75 (= 45^\circ / -60^\circ)$ along the contrast rotation path, while an antiphase rotation would correspond to a constant local vorticity of $\mathcal{M} = -1$. The same findings hold when reversing the sign of P_{eff} (not shown) since in that case the individual image contrasts are inverted, and this does not affect the rotation direction of the SP-STM contrasts upon in-plane tip magnetization rotation. This means that the SP-STM contrast rotation rule first identified by Dupé *et al.* [35] is insensitive to the sign of $P_{\text{eff}} = P_S P_T$ and, thus, to the sign of the surface and tip spin polarizations, P_S and P_T , respectively.

Let us now consider cases when $|Q| \neq 1$. Figure 6 shows SP-STM images of the antiskyrmion in Fig. 1(c) with $Q = 2$, of the skyrmion in Fig. 1(d) with $Q = -2$, and of the skyrmion in Fig. 1(e) with $Q = -3$, respectively, by rotating the tip magnetization direction (black arrows) in the surface plane indicated by gray circular arrows. We find that the number of bright and dark spots ($|Q|$ each) and their alternating order along the perimeter are preserved during the tip magnetization rotation, and this suggests that $|Q|$ can ideally be determined from a single SP-STM measurement with a magnetic tip of any in-plane direction. Note that the size and shape of the different contrast regions can be drastically different along the perimeter; the most pronounced case is $Q = -2$. The contrast rotation is illustrated by red circular arrows for all three cases in Fig. 6. Again, we find codirectional rotation of the contrast with the tip magnetization rotation for the skyrmions ($Q < 0$) and antidirectional rotation for the antiskyrmions ($Q > 0$). This, together with the insensitivity of the contrast rotations to the sign of P_{eff} , should enable the determination of the sign of the vorticity m and the topological charge Q based on a series of SP-STM measurements performed with rotated in-plane sensitive magnetic tips: a codirectional rotation of the contrast denotes $m > 0$ (here $Q < 0$), i.e., skyrmion, and an antidirectional rotation of the contrast denotes $m < 0$ (here $Q > 0$), i.e., antiskyrmion. Moreover, note that the contrast rotations in Fig. 6 are neither in phase ($\mathcal{M} = 1$ along

the path) nor in antiphase ($\mathcal{M} = -1$ along the path) with respect to the tip magnetization rotations due to $|Q| \neq 1$. An approximately constant $\mathcal{M} = -2$ and 3 along the path is, respectively, obtained for $Q = 2$ [Fig. 4(c)] and $Q = -3$ [Fig. 4(e)] only. We also find that the local angular velocity of the contrast rotation is indeed inversely proportional to the local vorticity \mathcal{M} ; see Eq. (12). Again, the most pronounced case is $Q = -2$, where a faster and slower contrast rotation is, respectively, obtained for smaller and larger absolute values of the local vorticity, i.e., here \mathcal{M} is not constant along the path; compare the contrast features in the middle row of Fig. 6 with Fig. 4(d). Importantly, if we assign unique identification labels of the bright (B1-B $|Q|$) and dark (D1-D $|Q|$) spots, a 360-deg in-plane rotation of the tip magnetization does not bring back the same spot to its original position, although the STM image looks the same. To achieve this, a $|Q| \times 360$ -deg tip magnetization rotation is needed. This can be clearly seen in the series of SP-STM images for each considered skyrmionic structure in Fig. 6: any uniquely identified spot arrives back to its original position at a 2×360 - and 3×360 -deg tip magnetization rotation for $Q = \pm 2$ and -3 , respectively. These findings suggest that $|Q|$ can be determined from a series of SP-STM measurements with in-plane rotated magnetic tips, where the position of one particular contrast region is monitored. It is important to note that the above suggested procedures to obtain the sign and magnitude of Q in SP-STM experiments can only work in case of a uniform sign of the local vorticity throughout the whole skyrmionic area.

A special case is the chimera skyrmion in Fig. 1(f) with $Q = 0$. SP-STM images of this spin structure are shown in Fig. 7 by rotating the tip magnetization direction (black arrows) in the surface plane indicated by a gray circular arrow. Here, we find opposite local contrast rotations illustrated by red circular arrows for the skyrmionic (lower right) and antiskyrmionic (upper left) parts of the chimera skyrmion, corresponding to different signs of the local vorticity [see Fig. 4(f)], the only case among the skyrmionic spin structures in Fig. 4. These local contrast rotations behave as described in Fig. 6, i.e., the skyrmionic part shows codirectional and the antiskyrmionic part shows antidirectional rotation of the contrast with respect to the rotation of the in-plane tip magnetization direction, and the local angular velocity of the contrast rotation is inversely proportional to the local vorticity; see Eq. (12). From this it uniquely follows that the number of bright and dark spots is not anymore preserved upon the rotation of tip magnetization. In most of the images of Fig. 7 there are either two bright spots and one dark spot or the opposite composition: two dark and one bright. The existence of images with an equal number of bright and dark contrast regions, e.g., one bright and one dark in the lower left and upper right images in Fig. 7, makes the identification of the chimera skyrmion impossible from a single SP-STM measurement using a tip with an arbitrary in-plane magnetization orientation. Nevertheless, as shown above, the in-plane rotation of the tip magnetization is quite helpful in the identification of the local vorticity for the $Q = 0$ chimera skyrmion and expectedly for other skyrmionic structures with arbitrary shapes and complex domain-wall structures by employing Eq. (12).

We have to stress that the shape of the magnetic skyrmions formed in anisotropic environments, e.g., in reconstructed

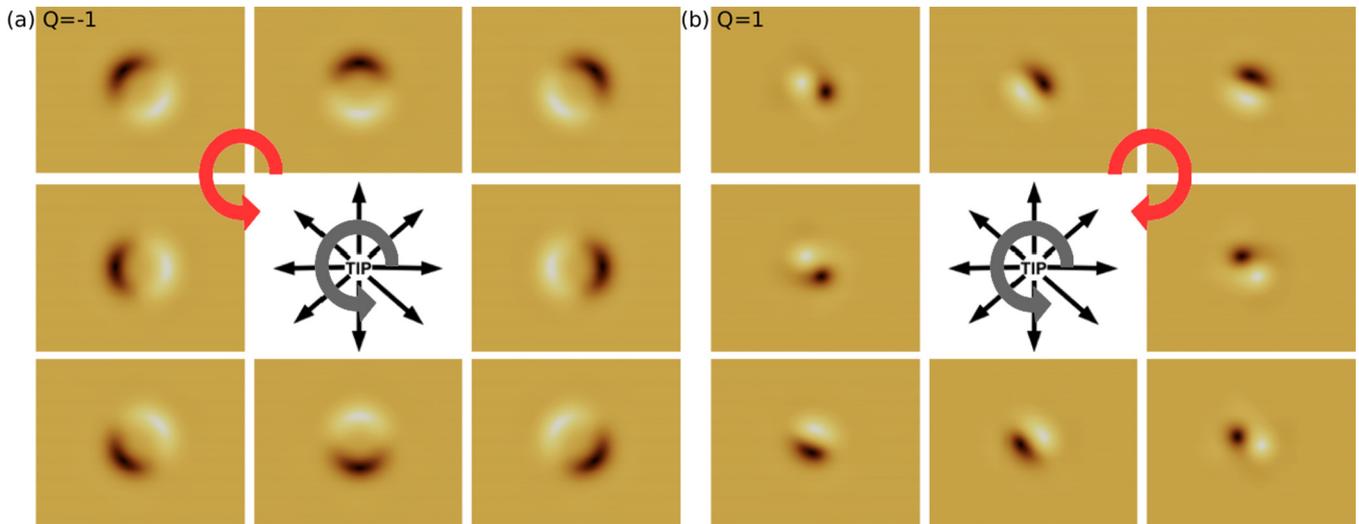


FIG. 5. Calculated SP-STM images by rotating the tip magnetization direction in the surface plane (denoted by gray circular arrows) using $P_{\text{eff}} = +0.4$ for (a) the skyrmion with $Q = -1$ and (b) the antiskyrmion with $Q = 1$. The opposite directions of contrast rotation are illustrated by red circular arrows in the two cases.

films [17,37], considerably affects the local vorticity and, consequently, the observed in-plane SP-STM contrasts. Such an example is an axially nonsymmetric spin structure derived from experimental SP-STM contrasts in Ref. [17], which shows both equal and unequal numbers of bright and dark contrast regions imaged with in-plane magnetized tips. This already suggests the presence of a nonuniform sign of the local vorticity. A deeper theoretical analysis puts forward that the observed spin structure is a magnetic skyrmion with $|Q| = 1$ exhibiting a noncircular shape [37]. It is interesting to note the similarity of the out-of-plane SP-STM contrast of this reported skyrmion and our skyrmion with $Q = -2$ [see Fig. 2(d)]. The comparison of the rotated in-plane contrasts in the middle row of Fig. 6 and those of Ref. [17], however, excludes the $Q = -2$ skyrmion in the experiment. This is in agreement with our above discussion since the proposed spin structure in Ref. [37] shows changing directions of the in-plane rotation

of the spins along the perimeter, and this clearly results in a nonuniform sign of the local vorticity present in the system. The reason for the distorted shape of the skyrmions and their nontrivial local vorticity in the experiment is the modified exchange interactions due to the reconstructed geometry of the magnetic film. A similar effect has also been observed in another complex magnetic (spin spiral) state in a confined geometry [51].

Finally, we present a series of SP-STM images in Fig. 8, where the evolution of the shape of the skyrmion with $Q = -1$ and of the antiskyrmion with $Q = 1$ is shown as the function of the Ir concentration of the PtIr alloy overlayer. The exchange parameters of Fe in the $(\text{Pt}_{1-x}\text{Ir}_x)\text{Fe}/\text{Pd}(111)$ ultrathin films have been taken from Ref. [38]. The different external B field values at different Ir concentrations are given in the caption of Fig. 8. These values were selected to ensure similar diameters of the skyrmionic structures, and because the field-polarized

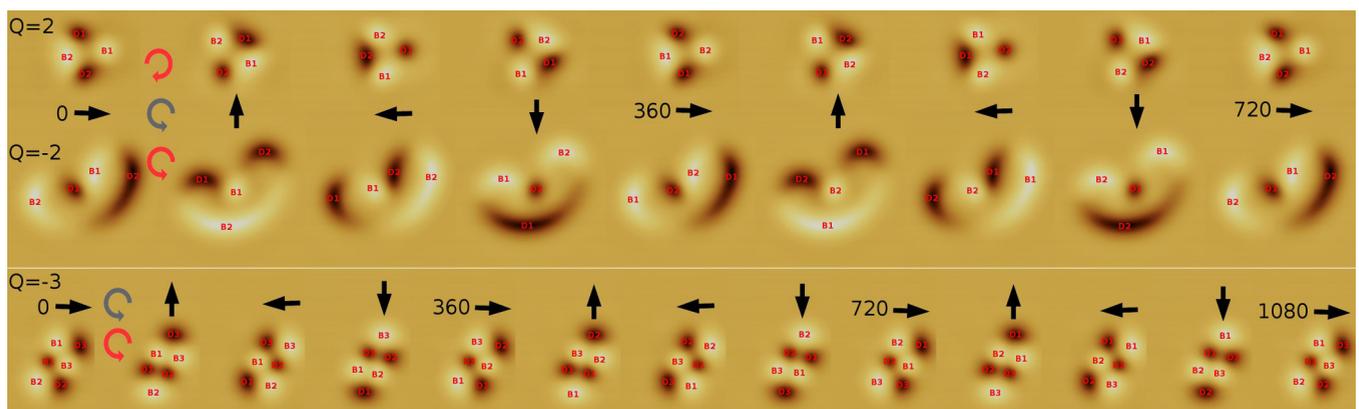


FIG. 6. Calculated SP-STM images by rotating the tip magnetization direction in the surface plane (denoted by gray circular arrows) using $P_{\text{eff}} = +0.4$ for the $Q = 2, -2$, and -3 skyrmionic structures. The contrast rotation is illustrated by red circular arrows in each case. The individual contrast regions are labeled as B1-B $|Q|$ for bright and D1-D $|Q|$ for dark, and their positions upon the tip magnetization rotation are shown. Note that the same image is obtained at a 360-deg tip magnetization rotation with misplaced individual contrast regions, and a $|Q| \times 360$ -deg tip magnetization rotation is needed to obtain the same positions of B1-B $|Q|$ and D1-D $|Q|$ contrast regions.

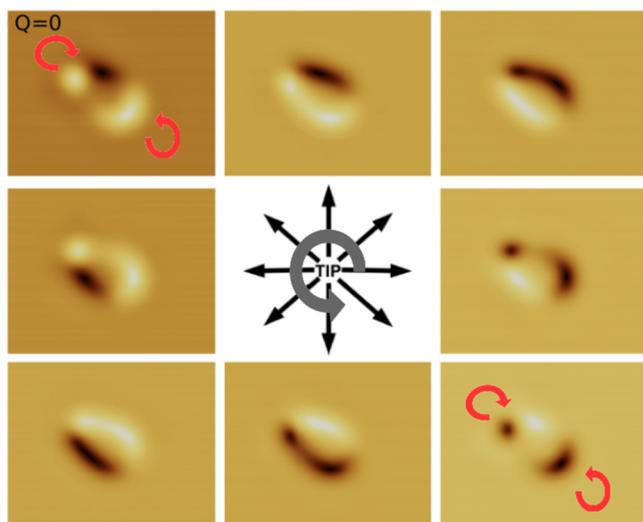


FIG. 7. Calculated SP-STM images by rotating the tip magnetization direction in the surface plane (denoted by a gray circular arrow) using $P_{\text{eff}} = +0.4$ for the chimera skyrmion with $Q = 0$. The opposite directions of contrast rotation of the skyrmionic (lower right) and antiskyrmionic (upper left) parts of the chimera skyrmion are illustrated by red circular arrows.

state where the objects are metastable is reached at higher field values for higher Ir concentrations [36]. Note that the magnitude and direction of the B field can drastically modify the size and shape of skyrmions [29]. First of all, we find in Fig. 8 that the primary two-lobes magnetic contrast of the two investigated skyrmionic structures does not change by tuning the composition of the alloy overlayer in the studied range (0–20% Ir in PtIr). However, for both skyrmionic structures the appearance of an outer ring contrast is evident by increasing the Ir content of the overlayer: At $x = 0.00$ Ir concentration no ring contrast is present, where repulsive skyrmions are observed [38]; at $x = 0.10$ a ring contrast appears, which is even more pronounced at $x = 0.20$ Ir concentration. The outer

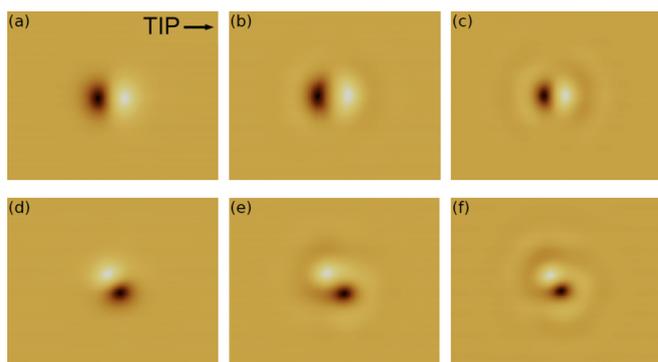


FIG. 8. Simulated SP-STM images using an in-plane magnetized tip (pointing to the $+x$ $[1\bar{1}0]$ direction) with $P_{\text{eff}} = +0.4$ for the skyrmion with $Q = -1$ (a–c) and for the antiskyrmion with $Q = 1$ (d–f) at different Ir concentrations in the $(\text{Pt}_{1-x}\text{Ir}_x)\text{Fe}/\text{Pd}(111)$ ultrathin magnetic film (external B field values are given in parentheses): $x = 0.00$ (0.00 T) (a, d), $x = 0.10$ (4.22 T) (b, e), and $x = 0.20$ (17.84 T) (c, f).

ring contrast is a signature of oscillating in-plane components of the spins, which leads to short-range attractive interactions between skyrmionic structures [33,38]. This is physically governed by the frustration of Heisenberg exchange interactions [36,38] upon changing the composition of the alloy overlayer. In agreement with Ref. [33], our results highlight that not only skyrmions but also antiskyrmions can show an attractive behavior that could be useful for technological exploitations. We propose that attractive skyrmionic structures can be identified in SP-STM images taken with in-plane magnetized tips by the presence of an outer ring contrast that, as we find, has a much smaller corrugation than the primary magnetic contrast. It is expected that the corrugation of the outer ring contrast is directly related to the attractive potential of the skyrmionic structures. It would be worthwhile to study this effect both experimentally and theoretically in the future.

IV. CONCLUSIONS

We have investigated metastable skyrmionic spin structures with various topological charges ($Q = -3, -2, -1, 0, +1, +2$, where the vorticity is $m = -Q$) in the $(\text{Pt}_{1-x}\text{Ir}_x)\text{Fe}/\text{Pd}(111)$ ultrathin magnetic film by means of SP-STM calculations. Based on the calculated SP-STM images, we conclude that an out-of-plane magnetized tip already results in distinguished SP-STM contrasts for the different skyrmionic structures corresponding to their symmetries. For systems exhibiting a uniform sign of the local vorticity, we have demonstrated that the magnitude of the topological charge can be determined from the image contrast of a single SP-STM measurement using an in-plane magnetized tip: the number of bright and dark contrast regions alternating along the perimeter of the skyrmionic spin structure equals $|Q|$ each. For such systems we have also found that an in-plane tip magnetization rotation provides the sign of Q independently of the sign of the effective spin polarization in the tunnel junction: codirectional rotation of the contrast with respect to the tip magnetization rotation indicates skyrmions ($m > 0$, here $Q < 0$) and antidiagonal contrast rotation antiskyrmions ($m < 0$, here $Q > 0$). We also showed that an in-plane rotation of the tip magnetization by $|Q| \times 360$ deg is needed to bring back the same bright or dark contrast region to its original position in the SP-STM image, proving that the local angular velocity of the contrast rotation is inversely proportional to the local vorticity. This finding could also be useful to determine $|Q|$ in case of spin structures with a uniform sign of the local vorticity.

A special case has been identified, the chimera skyrmion with $Q = 0$, where the number of bright and dark contrast regions is not preserved upon in-plane tip magnetization rotation but its skyrmionic and antiskyrmionic parts characterized by local vorticities of opposite sign obey the contrast rotation described above. We propose that a series of experimentally measured in-plane SP-STM contrasts can help the topological identification of skyrmionic objects: an unequal number of bright and dark contrast regions in a single measurement or a nonpreserved number of bright and dark contrast regions upon in-plane tip magnetization rotation indicates a nonuniform sign of the local vorticity for the considered structure. If neither of these apply then a uniform sign of the local vorticity

can be stated and the sign and magnitude of Q and m can be determined as described above. We also propose that theoretical calculations of geometrically modified exchange interactions will explain the formation of distorted real-space magnetic textures and their local vorticity and SP-STM contrasts in the future. Moreover, using an in-plane magnetized tip, we found that an outer ring contrast appears, indicative of both attractive skyrmions and antiskyrmions due to the tuning of exchange interactions, in our case upon increasing the Ir content of the PtIr overlayer.

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