Erratum: Symmetry breaking in spin spirals and skyrmions by in-plane and canted magnetic fields (2016 New J. Phys. 18 075007)

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Due to an error in the production process, references [34, 35] were mistakenly swapped. The correct references are as follows.

[34] Hanneken C et al 2015 Nature Nanotechnol. 10 1039
[35] OOMMF code (http://math.nist.gov/oommf)
Symmetry breaking in spin spirals and skyrmions by in-plane and canted magnetic fields

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Abstract

The influence of in-plane and canted magnetic fields on spin spirals and skyrmions in atomic bilayer islands of palladium and iron on an Ir(111) substrate is investigated by scanning tunneling microscopy at low temperatures. It is shown that the spin spiral propagation direction is determined by the island’s border which can be explained by equilibrium state calculations on a triangular lattice. We find a different response of spin spirals to in-plane magnetic fields for a propagation direction parallel to the applied field as compared to perpendicular, which originates from their cycloidal nature. As a result, the spin spiral propagation direction may be reoriented by in-plane fields. Furthermore, it is demonstrated that also skyrmions are distorted in canted fields which allows to determine the sense of magnetization rotation as enforced by the interfacial Dzyaloshinskii–Moriya interaction.

1. Introduction

Ultrathin magnetic films on heavy metal substrates can exhibit complex spin textures due to the competition of Heisenberg exchange interaction, magnetocrystalline anisotropy, and Dzyaloshinskii–Moriya interaction (DMI) [1]. A significant contribution of DMI is related to a strong spin–orbit coupling at the interface of the magnetic layer and the heavy metal substrate combined with the broken inversion symmetry at interfaces [2]. The resulting spin states are spin spirals and skyrmions with a unique rotational sense enforced by the DMI [1, 3–5]. In general, spin spirals are periodic magnetic textures of helical, cycloidal, or conical nature, whereas magnetic skyrmions are particle-like, topologically distinct states which are either of cycloidal or helical nature. While they were theoretically predicted in the nineties [6], the first experimental evidences for the existence of skyrmions were reported only a few years ago [4, 5, 7–9]. Since then, skyrmions have attracted significant interest due to their potential applications in future spintronic devices [10–13].

Among the non-centrosymmetric bulk crystals that exhibit spin spirals and magnetic skyrmions are MnSi [7, 14–17], Fe₃Co₃Si [8, 18], FeGe [9], and Cu₂OSeO₃ [19]. These types of materials typically show helical order as magnetic ground state and the application of magnetic fields leads to spin spiral propagation reorientations [20–23], conical phases, and skyrmions. Recently, it was found that thin films of MnSi exhibit a skyrmion phase in a wider range of magnetic fields and temperatures compared to bulk crystals [15, 24]. In ultrathin magnetic films of one or few atomic layers the DMI can also be induced purely from the interface [2–5]. Skyrmions in such ultrathin magnetic films are, in contrast to bulk non-centrosymmetric crystals, confined by the effectivley two-dimensional structure. As a consequence, it is interesting to study the response of such interface-driven skyrmions to in-plane and canted magnetic fields, in contrast to skyrmions in bulk crystals which can always orientate along the externally applied field. In thin films of non-centrosymmetric materials magnetic anisotropy can pin magnetic textures to a particular direction relative to the crystal, allowing the distortion of spin spirals and skyrmions by canted magnetic fields [25]. Lin et al simulated within a classical spin model the distortion of skyrmions due to canted fields which leads to an asymmetric shape of skyrmions [26]. A corresponding effect has been observed for chiral domain walls, leading to an asymmetric domain wall movement in canted fields [27].
previously rotational-symmetric skyrmions. Furthermore, we prove that the skyrmions of this system are cycloidal and have the theoretically predicted propagation direction of spin spirals which favors a perpendicular alignment to the magnetic in-plane field at low temperatures due to the competition of Heisenberg exchange and DMI fields on the static and dynamic properties of magnetic skyrmions.

Here, we investigate the influence of in-plane and canted magnetic fields on the spin structure of atomic bilayer islands of palladium and iron on Ir(111) by using scanning tunneling microscopy at low temperatures (see the supplement for the experimental setup). We show that the propagation direction of the spin spiral is determined by the coupling to the islands’ boundaries and thus by their shape. The application of in-plane fields along the spin spiral’s propagation direction distorts the spin spiral, thereby revealing its cycloidal nature. The symmetry breaking by the in-plane fields allows, depending on the island’s shape, a reorientation of the propagation direction of spin spirals which favors a perpendicular alignment to the magnetic in-plane field. Furthermore, we prove that the skyrmions of this system are cycloidal and have the theoretically predicted clockwise sense of magnetization rotation by applying canted fields causing a symmetry breaking for the previously rotational-symmetric skyrmions.

2. Spin spirals in Pd–Fe bilayer islands and their response to out-of-plane magnetic fields

Bilayer islands of Pd–Fe on Ir(111) were prepared similar to previous studies. They show a spin spiral ground state in zero external field at low temperatures due to the competition of Heisenberg exchange and DMI fields, as can be revealed by high-resolution spin-polarized scanning tunneling microscopy (SP-STM), see figure 1. While Fe grows in fcc stacking at the lower part of the Ir(111) atomic step edges, the Pd grows as either hcp or fcc stacked islands on top of the atomic Fe layer or at the lower step edge of the extended Fe film. Here, we focus on fcc stacked Pd islands on top of fcc grown Fe layers. The orientation of the hexagonal atomic lattice of the Pd–Fe bilayer, which is pseudomorphically grown on the Ir(111) substrate, can be derived from the straight edges of the Pd islands on the Fe monolayer, see figure 1(a). Since SP-STM is sensitive to the projection of the local sample magnetization onto the quantization axis given by the SP-STM tip’s magnetization direction, the observed stripes on top of the topographically flat island correspond to the wavefronts of the spin spiral (figures 1(b) and (c)). These stripes are not strictly parallel across the island but exhibit bends and even branches. A comparison of the orientation of the hexagonal atomic lattice and the propagation direction of the spin spiral for the area marked in figure 1(a) indicates that in the interior of the island preferentially aligns with the high symmetry direction. A closer inspection of the spin spiral’s propagation vectors at the island edge (see white arrows in figure 1) reveals that the spin spiral prefers to propagate along the island’s border.

In order to obtain deeper insight into the mechanisms of the arrangement of the spin-spirals with respect to the island’s borders, we performed calculations of the equilibrium energy of a spin-spiral state on a triangular lattice. For simplicity, we take an effective nearest neighbor exchange interaction and DMI of strengths $J$ and $D$ per atom into consideration with a $D/J$ ratio close to the experimental system. We find that within an extended magnetic film a reorientation of the high symmetry direction to the [101] direction reduces the energy by only $1.44 \times 10^{-5}$ J for each atom. In contrast, we can show that the energy of an atom at the rim along a close-packed row of a magnetic film depends much stronger on the direction of the propagation vector of the local spin-spiral configuration. The orientation of parallel to the rim reduces the energy by.
9.2 $\times 10^{-2}$ J for each atom at the rim compared to an alignment of $k_{SS}$ perpendicular to the rim. In both cases, the internal energy of the spin-spiral state is minimized by an alignment of $k_{SS}$ with a crystallographic direction in such a way that all bonds contribute to the reduction of both the exchange energy and the DM energy. The larger contribution of the rim is related to its symmetry breaking. In this simple analytical model we have not included the predicted edge tilt at the border [33], but we have checked that it would also favor a $k_{SS}$ parallel to the island edge. A more detailed explanation including the calculations can be found in the supplement. As an example, the values can be approximated for the island shown in figure 1. We determined the number of atoms at the border by dividing the perimeter by the nearest-neighbor distance (2.715 Å) of the pseudomorphic film. In the same way, the absolute number of atoms in the island was estimated by dividing the island’s area by the area of a hexagonal unit cell. Finally, we estimate that within the magnetic island of figure 1(a) reorientation of $k_{SS}$ from [112] to [101] can reduce the energy by 3.2 meV, whereas a change of $k_{SS}$ at the island border from perpendicular to parallel to the rim would lead to a reduction of 350 meV. Thus, we find that the influence of the border on the direction of $k_{SS}$ is about 100 times larger than that of the inner part for this particular island. This means that the details of the spin spiral in an island, such as bends and branches, are governed by the borders.

Besides SP-STM measurements, which are directly sensitive to the projection of the local sample’s magnetization onto the quantization axis given by the magnetization direction of the SP-STM tip, the recently discovered non-collinear magnetoresistance (NCMR) effect [34] can be exploited to investigate changes in the non-collinearity of the sample magnetization even with a non-magnetic STM tip. In general, NCMR leads to a change in the measured differential tunneling conductance, $dI/dV$, as a result of variations of the local non-collinearity of the sample’s spin texture. Thus, lateral changes of the non-collinearity can be investigated by spatially resolved NCMR mapping. When studying the response of a magnetic system to an external field the measurement of NCMR with non-magnetic probe tips avoids ambiguities in data interpretation, compared to SP-STM studies with ferromagnetic probe tips where a reorientation of the tip’s magnetization direction in an external magnetic field might occur.

Figure 2 shows NCMR images of a Pd–Fe bilayer island as a function of an externally applied magnetic field, as measured with a non-magnetic STM tip. In the Pd–Fe bilayer system, the NCMR contrast can be observed in a bias voltage interval of 600–800 mV; at other bias voltages this contrast vanishes. It roughly scales with the nearest-neighbor-angle $\theta_{nn}$ and the $dI/dV$ signal decreases with increasing $\theta_{nn}$ [34]. A homogeneous spin spiral is characterized by a constant $\theta_{nn}$ and thus would display a constant $dI/dV$ signal, as sketched in figure 2(a). The Pd–Fe bilayer system is known to exhibit an out-of-plane easy axis [28–30], and thus the spin spiral is inhomogeneous which means that $\theta_{nn}$ oscillates between in-plane and out-of-plane parts of the spin spiral as
illustrated in figure 2(a). Because of this inhomogeneity the characteristic stripe pattern of the spin spiral can be observed by NCMR imaging, see figure 2(b). Furthermore, for this island we find that the direction of $k_{\text{SS}}$ is governed by the border. In figures 2(c) and (d) the response to an out-of-plane magnetic field is demonstrated. In the mixed phase at $B = 1$ T spin spirals and skyrmions coexist [5], see figure 2(c). The part of the spin spiral with magnetic moments aligned parallel to the external field increases in width, leading to a local decrease of $q_{\text{nm}}$ and thus an increased $dI/dV$ signal due to NCMR. The area antiparallel to $B$ shrinks, implying larger $q_{\text{nm}}$ and thus decreased $dI/dV$ signal. The axially symmetric skyrmions in the Pd–Fe bilayer appear in NCMR images as rings at small magnetic fields, because the region of largest non-collinearity is close to the area with in-plane magnetization. With increasing field the skyrmions shrink yielding smaller rings and eventually dots [34]. For higher fields, at about $B = 2$ T, there are only single, pinned skyrmions left, see figure 2(d).

3. Spin spirals in in-plane magnetic fields

Figure 3(a) shows a Pd–Fe bilayer island in zero field while figures 3(b) and (c) show an island with an in-plane field applied which is either collinear or perpendicular to $k_{\text{SS}}$, respectively. While in the latter case no change in the appearance of the spin spiral is observed, a clear difference in the measured $dI/dV$ signal is revealed for figure 3(b), where $k_{\text{SS}}$ is roughly collinear to $B$. The line profiles depicted in figure 3(e) reveal an alternating depth of the minima in the measured $dI/dV$ signal in case of $k_{\text{SS}}$ parallel to $B$, in contrast to a sine-like $dI/dV$ signal.
signal for the zero-field case. As the change of the measured $dI/dV$ signal due to NCMR depends on $\theta_{\text{nn}}$, this finding demonstrates that the local non-collinearity of the spin spiral’s in-plane parts changes depending on their alignment relative to the applied in-plane field. We again conclude that sample areas where $\theta_{\text{nn}}$ decreases are parallel to $\mathbf{B}$, whereas an increasing $\theta_{\text{nn}}$ is observed for areas with magnetic moments antiparallel to $\mathbf{B}$. This behavior is in agreement with the proposed cycloidal nature of the spin spiral. For a helical spin spiral, the in-plane parts of the local magnetization are orientated perpendicular to $\mathbf{k}_{\text{SS}}$. Therefore, such a spin spiral would exhibit the same change of contrast as the cycloidal spin spiral shows in figure 3(b), however, for a perpendicular orientation of the in-plane field to $\mathbf{k}_{\text{SS}}$. Using similar arguments, we can also understand the NCMR image for an in-plane field perpendicular to $\mathbf{k}_{\text{SS}}$, see figure 3(c). Here, we expect that the cycloidal spin spiral changes into a transversal-conical phase in order to arrange with the external field. In this case, the $\theta_{\text{nn}}$ are expected to decrease with increasing in-plane field. Micromagnetic simulations for a spin spiral in zero field and in a field collinear to $\mathbf{k}_{\text{SS}}$ using OOMMF [35] and parameters obtained from experiments on hcp-stacked Pd–Fe bilayer islands [30] corroborate our explanation for the NCMR contrast changes as can be seen by the periodic change of $\theta_{\text{nn}}$ between both cases, shown in figure 3(d). The simulation has also been conducted for the case of an in-plane field perpendicular to $\mathbf{k}_{\text{SS}}$ which reveals a significantly smaller change in $\theta_{\text{nn}}$ compared to the collinear field orientation. For this field orientation the strongest change in $\theta_{\text{nn}}$ occurs at the in-plane parts of the spin spiral since there the external field does not have to compete with the out-of-plane easy axis anisotropy. The simulation shows a tilting of the magnetic moments at the center of the simulated island towards the field direction of roughly $11^\circ$ and $8^\circ$ for the in-plane and the out-of-plane part of the spin spiral, respectively. Thus we expect the difference in $dI/dV$ between in-plane and out-of-plane parts to decrease due to an in-plane field perpendicular to $\mathbf{k}_{\text{SS}}$ which is, however, difficult to observe experimentally.

Figure 4. Spectroscopic $dI/dV$ maps of Pd–Fe bilayer islands obtained with a non-magnetic STM tip. While for the measurement in (a) the magnetic field was applied after cool-down of the sample, in (b)–(d) the sample was field-cooled (from 30 to 4.7 K) in in-plane fields as indicated. The maps of the highlighted areas are high-resolution scans revealing changes in the propagation direction of the spin spirals.
Figure 4 shows several Pd–Fe bilayer islands at 4.7 K and in an in-plane field as indicated. At this temperature the application of in-plane fields only leads to the above mentioned distortions of the spin spiral. After warming up the sample to 30 K and cooling it down again, while the field is applied, the $k_{SS}$ of some of the islands reorientate to become perpendicular to the field (see figure 4(b)). This demonstrates that the application of in-plane fields leaves the system in a metastable state as it lacks the energy to overcome the barriers to lower energy states. In contrast, figures 4(c) and (d) show several islands that were subsequently field-cooled in in-plane fields perpendicular to each other. On the highlighted island the largest part of the spin spiral switches its $k_{SS}$ to become perpendicular to the field. The other islands also show varying degrees of change in the $k_{SS}$ of the spin spirals. Thus a reorientation of the spin spiral’s propagation direction upon application of an in-plane field is possible, but the magnetic field competes with the strong coupling of $k_{SS}$ to the island’s rim. These results show that a spin spiral propagation perpendicular to a magnetic in-plane field is energetically preferred.

4. Skyrmions in canted magnetic fields

Application of out-of-plane magnetic fields leads to a phase transition from the spin-spiral state to the skyrmionic state, as shown in figure 2. For the out-of-plane fields used here the in-plane part of the skyrmion shows the highest non-collinearity and thus the lowest $dI/dV$ signal (see figure 2(c)). Figures 5(a) and (b) show numerous skyrmions in canted fields. The STM measurement parameters that are necessary to observe the NCMR contrast can lead to annihilation and creation of skyrmions on the time-scale of the STM scan, thereby causing sudden jumps, i.e. line noise, in the STM images. While in figure 5(a) the skyrmions have a tendency to exhibit a lower $dI/dV$ signal on their left side, the skyrmions in the inverted in-plane field show likewise a tendency of a reduced $dI/dV$ signal on their right side (see figure 5(b)). A particularly stable skyrmion is marked and examined by high-resolution maps (figures 5(c) and (d)) together with the corresponding line profiles along the direction of the applied in-plane field (figure 5(f)). The observed change in $dI/dV$ is similar to the one for the spin spirals: again, the $dI/dV$ signal changes according to the orientation of the spin structure relative to the...
direction of the applied magnetic field, and we find the strongest change in the in-plane regions. Since in NCMR contrast images the $dI/dV$ signal depends roughly on $\theta_{nm}$ we conclude that $\theta_{nm}$ in the in-plane parts of the spin structure increases on one side of the skyrmion and decreases on the other side. Thus the previously rotational symmetric skyrmion [5, 30] experiences a symmetry breaking and is transformed into a non-circular skyrmion with mirror plane symmetry along the direction of the in-plane field component. However, the exact shape of the skyrmion also strongly depends on the vicinity to other skyrmions or defects. Especially, the measured $dI/dV$ signal in the area between skyrmions, which exhibits a small $\theta_{nm}$ depends on the distance between the skyrmions or skyrmions and defects in the Pd–Fe bilayer. This explains the different $dI/dV$ signals at the rims of the skyrmions that can be observed in the line-profiles of figure 5(f). The response to the canted magnetic field confirms that the observed skyrmions are cycloidal. For a helical skyrmion the asymmetry in the in-plane parts would show up on the axis perpendicular to the applied field direction. The data obtained also allows a determination of the sense of rotation as enforced by the DMI. The observed asymmetry directly reveals in which direction the in-plane parts of the skyrmion, that are collinear to the applied field, are pointed and—in combination with the orientation of the applied field’s out-of-plane component—we can derive the perpendicular orientation of the magnetic moments in the center of the skyrmion and its surrounding (compare figure 5(e)). The spatial distribution of the magnetic moments is found to exhibit a clockwise rotation from left to right (as illustrated by black arrows in figure 5(I)), and thereby our experimental results confirm the theoretical predictions by Dupé et al [28] and Simon et al [29].

5. Conclusion

We have shown that the spin spirals in Pd–Fe bilayer islands prefer to propagate along the islands’ borders. A calculation corroborated the experimental results showing that the coupling of the spin spiral to the border is by two orders of magnitude stronger than the coupling to a particular symmetry direction of the hexagonal atomic lattice. In-plane magnetic fields change the nearest-neighbor-angles between magnetic moments of the spin spiral’s in-plane parts if applied collinear to the spin spiral’s propagation direction, thereby providing an experimental proof that the spin spiral is cycloidal. In contrast, for a perpendicular orientation of the in-plane field relative to the spin spiral’s propagation direction, our simulations suggest a distortion towards a transversal-conical spin spiral. Field-cooled samples in differently orientated in-plane fields lead, depending on the island’s shape, to a reorientation of the spin spiral propagation direction. On the other hand, the application of magnetic fields at 4.7 K only results in distortions of the spin spiral. A canted field induces an asymmetry in the skyrmion’s shape along the field direction, thereby breaking its rotational symmetry. For skyrmions the induced asymmetry not only reveals their cycloidal nature, but additionally allows the determination of the sense of magnetization rotation.

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