Discrete helicoidal states in chiral magnetic thin films

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Magnetometry and magnetoresistance measurements in MnSi thin films and rigorous analytical solutions of the micromagnetic equations show that the field-induced unwinding of confined helicoids occurs via discrete steps. A comparison between the magnetometry data and theoretical results shows that finite size effects confine the wavelength and lead to a quantization of the number of turns in the helicoid. We demonstrate a prototypical spintronic device where the magnetic field can push or pull individual turns into a magnetic spring that can be read by electrical means.

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The epitaxial induced strain in thin film cubic helimagnets and Dzyaloshinskii-Moriya interactions induce complex magnetic textures over a wide range of temperatures. Particularly, chiral skyrmions, usually metastable in bulk cubic helimagnets, are predicted to be thermodynamically stable in nanolayers of these materials.

Recently, these specific solitonic states over extended regions of the magnetic phase diagram have been observed in thin films and mechanically thinned nanolayers. These states are freely created and driven with an ultra-low current density, and are considered as promising objects for novel types of magnetic data storages. The induced anisotropy also extends the region of the magnetic phase diagram occupied by distorted helical states (helicoids), which have been proposed for a number of spintronic applications.

Bulk MnSi belongs to the cubic $P2_13$ ($T^4$) space group of magnetic crystals that lack inversion symmetry, which through the spin-orbit interaction produces the DM interaction responsible for the long wavelength helical order. Owing to a small cubic anisotropy, the helical order does not unwind in a magnetic field since modest fields cause a reorientation of the helix along the field direction. In bulk crystals, a field-induced ferromagnetic state is created by a second-order transition via a conical phase, as observed in MnSi, FeGe, and Cu$_2$OSeO$_3$. However, in lower symmetry crystals, the DM interaction can be uniaxial (e.g. Cr$_{1/3}$NbS$_2$). In this case, a field that is applied transverse to the helical propagation vector does not reorient the helix, but rather it creates helicoidal distortions. While bulk MnSi belongs to the former group, the strain induced by the Si substrate in epitaxial MnSi thin films lowers the symmetry to a trigonal $R3$ ($C_3^3$) space group and creates a uniaxial anisotropy with the hard-axis along the film normal. This anisotropy helps to stabilize the helicoidal state with an in-plane magnetization and the propagation vector perpendicular to the film surface.

A transverse field unwinds the helices and transforms them via first-order transitions into either elliptic skyrmion or elliptic cone phases.

This Letter investigates the role of finite size in the confinement of helicoids in MnSi thin films. Unlike bulk crystals, we observe discrete jumps in the magnetization at specific fields and thicknesses that correspond to the annihilation of individual turns of the helicoid. These jumps result from the truncation of the helicoids by the film interfaces, which stabilizes a quantized number of turns in the helicoids. The sudden changes in magnetic structure during the unwinding of the helicoids in a magnetic field produce anomalies in the magnetoresistance (MR) that enable an electronic reading of the number of turns in the helicoidal structure. With the ability to trap individual helicoidal turns, we demonstrate that the films exhibit the essential functionalities of a spintronic device based on chiral states.

The effects of finite size were explored both theoretically and experimentally. We performed MR and superconducting quantum interference device (SQUID) magnetometry measurements on MnSi thin films with thicknesses $9 \text{ nm} < d < 40 \text{ nm}$, which were grown by molecular beam epitaxy on Si(111) substrates (See Ref. [2]). MR was collected from four-probe resistance measurements on lithographically patterned Hall bars and cleaved rectangular strips using Au-wire leads attached to the films with In-solder. Both MR and SQUID measurements were conducted at 5 K with the field oriented along the MnSi[110] direction of the film and were compared to analytical solutions to a Dzyaloshinskii model that was extended to the case of thin films.

Chiral modulations in low-anisotropy helimagnets are well described theoretically by the standard Dzyaloshinskii model. We define $\mathbf{m} = \mathbf{M}/M_s$ as a unit vector in the direction of the magnetization $\mathbf{M}$ ($M_s = |\mathbf{M}|$), and $\mathbf{H}$ as the field applied along the $x$-axis. For a helicoid propagating along the $z$-axis, $\mathbf{m} = (\sin \theta, \cos \theta, 0)$ and the
energy density in the Dzyaloshinskii model is reduced to
\[ w(\theta) = A \left( \frac{d\theta}{dz} \right)^2 - D \frac{d\theta}{dz} - H M_s \cos \theta, \tag{1} \]
in terms of the exchange stiffness energy with constant, \( A \), the Dzyaloshinskii-Moriya coupling with constant, \( D \), and the Zeeman energy. Model (1) introduces two fundamental parameters: the zero field helical wavelength, \( L_D = 4 \pi A / D \) and the saturation field in the zero anisotropy limit, \( H_D = D^2 / (2 A M_s) \). In the case of MnSi films, \( L_D = 13.9 \text{ nm} \), and the average \( H_D \) is 0.77 T with a standard deviation of 0.05 T for the range of sample thicknesses presented in this Letter. These parameters are tabulated for a number of other cubic helimagnets in Refs. [1, 20].

Minimization of functional \( w(\theta) \) leads to the well known differential equation for a non-linear pendulum \[ \ddot{\theta} + \frac{g}{\ell} \sin \theta = 0. \]
Analytical solutions for helicoids in bulk helimagnets continuously transform from a single-harmonic helix with period \( L(0) = L_D \) at zero field into a set of isolated kinks at critical field \( H_{k1} / H_D = \pi^2 / 16 = 0.618 \), which accurately describes the observations in Ref. [25].

The unique magnetic properties of magnetic nanostructures are generally a result of reduced dimensionality and surface/interface induced magnetic anisotropy [21, 22]. For confined helicoids, these two factors can be readily taken into account by minimization of functional (1) in a layer of finite thickness with boundary conditions including surface/interface magnetic anisotropy. Our previous work did not find any significant influence of interface anisotropy on the magnetic properties of the thin films in the range of thicknesses presented here [2]. However, the breaking of translational symmetry in finite helicoids due to the presence of interfaces substantially influences their properties. For an in-plane magnetic field, confined helicoids in nanolayers evolve into the field induced ferromagnetic phase via a number of discrete jumps between states that have a quantized number of helicoidal turns. We calculate these states by minimization of functional (1) in a layer of finite thickness \( d \) with free boundary conditions. We have derived analytical solutions for confined helicoids (see Supplement) and present their main features in Fig. 1.

Figure 1(a) shows the three quantized states that are supported in a film with thickness \( d = 2.14 L_D \) with either zero, one, or two turns of the helix, depending on the applied field. The details of the evolution of the magnetic structure of this film as a function of applied field is presented in Fig. 1(b). The figure shows that the wavelength varies weakly with applied field up to \( H_{k1} = 0.28 H_D \). Above \( H_{k1} \), one turn of the helicoid is pushed out, and the second turn is pushed out above \( H_{k2} = 0.60 H_D \). For \( H > H_{k2} \), the system is in a twisted ferromagnetic state, with a ferromagnetic state in the center of the film, and chiral modulations at each interface. Our theory predicts that this state will persist up to high field. Alternatively,

![FIG. 1. (color on-line) (a) Calculated spin configurations for a helimagnet thin film of thickness \( d = 2.14 L_D \) with applied in-plane fields \( H = 0.25 H_D \) (left), \( 0.55 H_D \) (middle), \( 0.61 H_D \) (right). (b) Component of \( M \) along \( \mathbf{H} \) as a function of field and depth in the film of thickness \( d = 2.14 L_D \). (c) The average energy density for the linear approximation given by Eq. (2) for \( d = 1.32 L_D \) (left), and \( d = 2.14 L_D \) (right) in applied fields \( H = 0.0, 0.2, 0.4, \) and \( 0.6 \) \( H_D \)](image-url)
at \( k_{-1} < k_0 \). At higher fields, additional local minima form. These wells arise due to \( k \)-dependent oscillations in the Zeeman energy from the uncompensated moments in the finite helicoids.

One of the features of helicoidal films that distinguishes it from bulk behaviour is its evolution in small field. The slope of the helicoid wavelength in the low field limit, \( \eta_0 = (H_D/L_D)(dL/dH)_{H=0} \) can be readily derived as a function of the film thickness by an expansion of the potential \( \Phi \) in Eq. 2 for small values of \( H \):

\[
\eta_0(\nu) = \frac{1}{4} \left( \frac{\sin \pi \nu}{\pi \nu} - \cos \pi \nu \right) \sin (\sin \pi \nu).
\]

In this equation, the confinement ratio, \( \nu = d/L_D \), determines the sign of \( \eta_0 \). Surprisingly, \( \eta_0 \) can take on negative values that correspond to the helicoid tightening its pitch with increasing field as the system evolves in the \( k_{+1} \) potential well. This tightening of \( L \) is observed for the \( d = 2.14L_D \) film in Fig. 2(b). For positive \( \eta_0 \), the pitch relaxes with applied field because the helicoid resides in the \( k_{-1} \) potential well, as illustrated by the calculation for \( d = 1.83L_D \) in Fig. 2(c). The thickness dependence of the oscillations in \( \eta_0 \) are presented in Fig. 2(c). After an initial evolution in either the \( k_{+1} \) or \( k_{-1} \) wells, rigorous solutions for Eq. show that the helicoids transition via a series of first-order transitions toward the lowest \( k \)-state that corresponds to the twisted ferromagnetic state shown on the right in Fig. 2(a).

![Figure 2](image.png)

**FIG. 2.** Magnetoresistance measured at \( T = 5 \) K for film thicknesses 2.14\( L_D \) (a) and 1.83\( L_D \) (b) in a field \( H || \text{MnSi}[110] \). Open circles are the decreasing field data, closed squares are increasing field. Marked fields \( H_{h1} \) and \( H_{h2} \) are the transition fields taken from magnetometry measurements from the increasing branch data. (c) The slope of the magnetoresistance (blue squares) compared to \( \eta_0 \) (black line) from Eq. 3. The red line shows \( \eta_0 \) with 0.9 nm rms thickness variations.

In the following we present experimental evidence that illustrates that the Dzyaloshinskii model well describes the evolution of the helicoids in MnSi thin films. The oscillations in \( \eta_0(\nu) \) have important implications for the MR measurements in Fig. 2 where \( MR = (\rho(H) - \rho(0))/\rho(0) \). For the \( d = 2.14L_D \) film with \( \eta_0 < 0 \) in Fig. 2(a), there is an anomalous positive magnetoresistance in low fields, in contrast to the \( \eta_0 > 0 \) films that show a more conventional MR < 0, as is seen for the \( d = 1.83L_D \) film in Fig. 2(b). Since the MR is expected to increase with decreasing \( L \), we compare the thickness dependence of \(-dMR/dH\) measured at \( H = 0 \) to \( \eta_0 \) in Fig. 2(c), and we find that the oscillations in the slope of MR follow the predicted oscillations in the slope of the wavelength. To account for inhomogeneities in thickness, we convolve \( \eta_0 \) (thin black line) with a gaussian with a width that is specified by typical rms roughnesses of 0.4 nm and 0.8 nm for the top and bottom interfaces, as determined by x-ray reflectometry. The result is shown by the thick red line.

As the film approaches a critical field that signals the forcing out of a turn in the helicoid, labeled \( H_{h1} \) in Fig. 2(a), the MR becomes negative for all thicknesses because the wavelength increases monotonically with thickness above \( H_{h1} \) according to the model. For thicknesses \( L_D < d < 2L_D \) we see a single drop in the MR at \( H_{h1} \) (Fig. 2(b)) whereas at thicknesses \( 2L_D < d < 2.5L_D \) two first-order transitions occur as illustrated by the two well defined hysteretic drops in the MR of Fig. 2(a). The origin of these drops is more clearly explained by the SQUID data.

The peaks in the field dependence of the static susceptibility \( dM/dH \) (Figs 3(b)) confirm that there are either one or two first-order magnetic transitions below the saturation field \( H_{sat} \). The transition fields obtained from the peaks in \( dM/dH \) are normalized by \( H_D \), which is estimated from the field \( H_{sat} \) indicated by a minimum in \( d^2M/dH^2 \). The analysis used to relate \( H_D \) to the saturation field is described in Ref. 3. In Fig. 3(a), we plot the \( H_{h1} \) and \( H_{h2} \) obtained from the peaks in \( dM/dH \) in Fig. 3(b), and compare them to the theoretical threshold fields where a single turn of the helicoid is annihilated (red lines). The predicted transitions occur at a field that is about 0.05\( H_D \) lower than the experimental values. This may be due to interfacial anisotropies and/or the softening of the exchange and Dzyaloshinskii-Moriya interactions at the interfaces that are not accounted for in the model. This may also be a result of a systematic error in measurement of \( H_{sat} \) used to calculate \( H_D \), since the minimum in \( d^2M/dH^2 \) is weak and broad at \( T = 5 \) K. The estimate of \( H_D \) is further complicated by the twisting of the magnetization at the interfaces shown in the twisted ferromagnetic state in Fig. 3(a), whereas the procedure we use to estimate \( H_D \) assumes a conical state at high field. Further work will be needed to address the effect this has on the estimation of \( H_D \). However, what is important to note is that the model does predict the observed thickness dependence of \( H_{h1} \).
For film thicknesses close to $2L_D$, a second transition field is also present. Our previous work shows that at $T \gtrsim 15$ K, a low transition field $H_\beta$ coincides with the nucleation of skyrmions in the film. However, for the measurements at $T = 5$ K presented in this Letter, a comparison with the model calculations indicates a different origin. For the film thicknesses that are close to $2L_D$, a second peak in $dM/dH$ appears at $H_{h2}$. According to the model, this coincides with the thickness where an additional turn of the helicoid is able to fit between the film interfaces, and the field $H_{h2}$ corresponds to the field where this additional turn is forced out. The values for $H_{h2}$ follow the same trend in thickness as $H_{h1}$, which supports the claim that these two fields correspond to the same phenomena. At $d = 2.85L_D$, only one transition field is observed. Although a second transition should be present, the transition fields $H_{h2}$ is close enough to $H_{h1}$ that it may not be possible to resolve due to the width of the peaks in $dM/dH$.

The calculated magnetization is also in good agreement with SQUID measurements. To make a fitting parameter free comparison, we extract the thickness dependence of the net magnetization $M$ from SQUID measured hysteresis curves for a collection of samples, and normalize the data to the saturation magnetization $M_s$ obtained by extrapolation of the high-field $M$ above $\mu_0H = 2$ T to zero field. At $H = 0.05H_D$, the $M/M_s$ oscillates as a function of thickness due to an oscillation in the number of uncompensated spins, similar to what we observed in remanence \( H = 0 \). The data is 30% lower than the calculation due to insufficient alignment of the magnetic domains by the small field. When the field is increased to $H = 0.25H_D$, Fig. 4(b) shows the oscillations are nearly washed out, in agreement with the calculation shown by the thin black line. Above this field, we observe a qualitative change in behaviour. In Fig. 4(d), $M$ increases linearly above approximately $d/L_D = 1.3$ since larger thicknesses enables the helicoid to accommodate a more non-linear structure with a larger fraction of the magnetization along the field direction, as shown in Fig. 4(c). Figure 4(c) also shows that there is a drop in the magnetization in Fig. 4(d) when an additional turn in the helicoid is nucleated. This agreement between the data and the model calculations in Fig. 4(b) - (d) is achieved without any fitting parameters. The transitions in Fig. 4(c) and (d) are not as sharp as the model shown by the black lines due to heterogeneities in the samples. Variations in thickness can be simulated by convolving the calculated $M(d)$ with a gaussian, as was done in Fig. 4. The mentioned difficulty in determining $H_D$ is also expected to contribute to the uncertainties.

In conclusion, we have shown that a confining geometry in helimagnetic films with a strong easy-plane anisotropy stabilizes a quantized number of turns in the helicoid. The magnetization processes in the helicoidal state display the required functionalities of a device based on chirally twisted states, namely, (i) the states are discrete and reproducible, (ii) switching between these states is in principle possible due to the metastability demonstrated by the observed hysteresis, and (iii) the state of the helicoid can be read by electronic means. Furthermore, in higher magnetic fields, the thicker films are able to confine solitonic kinks that open the possibility to use cubic helimagnetic materials to explore some of the predicted effects in a helicoid soliton lattice \cite{16, 17}. We thank F. N. Rybakov for fruitful discussions and M. Johnson for technical assistance. TLM and MNW ac-

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FIG. 3.  (color on-line)  (a) Critical fields extracted for increasing (filled triangles) and decreasing (open triangles) magnetic fields with $\mathbf{H}|\text{MnSi}[1\bar{1}0]$ at $T = 5$ K. The solid red lines in (a) are the calculated threshold fields for the removal of a turn in the helicoid. (b) DC susceptibility measurements in an increasing magnetic field for thicknesses $L_D < d < 2.85L_D$, offset vertically for clarity. Red dotted lines track the observed transition fields.

FIG. 4. (color on-line) Fitting-parameter-free comparison of SQUID magnetization measurements at $T = 5$ K in increasing fields (open squares) and decreasing fields (filled squares) for $H/H_D$ values of (a) 0.05, (b) 0.25, (c) 0.35, and (d) 0.45. The values for $M$ are extracted from $M - H$ loops. The average value for $H_D$ is 0.77 T, and $\mathbf{H}|\text{MnSi}[1\bar{1}0]|\hat{x}$. (a)-(d) Calculations of $M(d)$ with (thick red line) and without (thin black line) 0.9 nm rms thickness variations. (e) Calculated depth profiles of $M_x$ as a function of thickness.
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SUPPLEMENTAL MATERIAL

Here we derive analytical solutions for helicoids described by the functional

\[ w(\theta) = A \left( \frac{d\theta}{dz} \right)^2 - D \frac{d\theta}{dz} - HM_s \cos \theta, \tag{4} \]

(Eq. 1 in the paper) for a layer of thickness \( d \) and free boundary conditions. The Euler equation for functional \( F \) has the first integral

\[ \frac{1}{k_0^2} \left( \frac{d\theta}{dz} \right)^2 + h \cos \theta = c, \tag{5} \]

where \( c \) is an integration constant, and \( h \) is the reduced field \( H/H_D \) and \( k_0 = D/(2A) = 2\pi/L_D \) is the propagation vector in zero field \( H \). The film interfaces break the translational symmetry of the helicoids, and for free boundary conditions at finite magnetic fields, Eq. (4) has solutions with either \( \theta(0) = 0 \) or \( \pi \). The solution to the Euler equations is given by,

\[ k_0 z = \int_0^{\theta} \frac{dt}{\sqrt{c - 2h \cos t}} \tag{6} \]

For \( h > 0 \), Eq. (6) describes magnetization profiles with \( \theta_0 = \theta(0) = 0 \), and for \( h < 0 \), profiles with \( \theta_0 = \pi \). Equation (6) can be expressed in terms of an incomplete elliptic integral of the first kind, and then inverted in order to find an expression for \( \theta \) in terms of the Jacobi amplitude function,

\[ \theta(z) = 2 \text{ am} \left( k_0 z \sqrt{h}/\kappa; i\kappa \right) + \theta_0 \tag{7} \]

where \( \kappa \) is the modulus of the elliptic function \( E \):

\[ \kappa = \sqrt{\frac{4h}{c - 2h}}. \tag{8} \]

The equilibrium helicoid configuration \( \theta(z) \) is a function of the two control parameters \( d \) and \( h \), and is obtained by finding \( \kappa \) that minimizes the energy density \( F \) averaged over the layer thickness,

\[ \bar{w}(\kappa) = \frac{1}{d} \int_{-d/2}^{d/2} \bar{w} \left( 2 \text{ am} \left( k_0 z \sqrt{h}/\kappa; i\kappa \right) \right) dz. \tag{9} \]

The solution to Eq. (9) together with Eq. (4) permits us to calculate the depth profile \( M_e(z) = M_s \cos(\theta(z)) \) in the inset of Fig. 5 for a \( d = 1.92L_D \) film in a field \( H = 0.365H_D \). The saturation magnetization \( M_s = 0.416\mu_B/Mn \) is obtained from SQUID magnetometry measurements. The calculated \( M_e(z) \) together with the nuclear scattering length density profile obtained from fits to the x-ray reflectometry data in Ref. [3] enable us to compute the polarized neutron reflectometry (PNR) scattering cross-sections. We compare the calculate PNR spectra to those of Ref. [3] in Fig. 5. The calculated spectra are in good agreement with the measurements with a small improvement in \( \chi^2 \) compared to the fit obtained in Ref. [3]. The remaining small deviations between the calculation and data could be a result of the nucleation of other phases such as skyrmion like vortices at grain boundaries, as seen in Ref. [4].

The modulus obtained from the minimization of Eq. (10) is related to the wavelength, \( L \), which is derived from the condition \( \theta(z = L/2) = \pi/2 \) [1]:

\[ \frac{L(h)}{L_D} = \frac{\kappa}{\pi \sqrt{h} K(\kappa)}. \tag{10} \]

The difference between the solution for a bulk crystal and that of a thin film is illustrated in Fig. 6, which shows
the anomalous tightening of the helicoid below $H_{h2}$ and the discontinuities in the wavelength at fields $H_{h1}$ and $H_{h2}$.

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