

# Diversity of states in a chiral magnet nanocylinder

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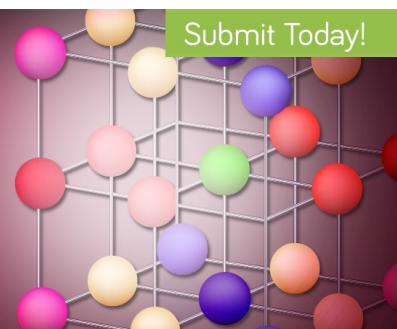
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## ABSTRACT

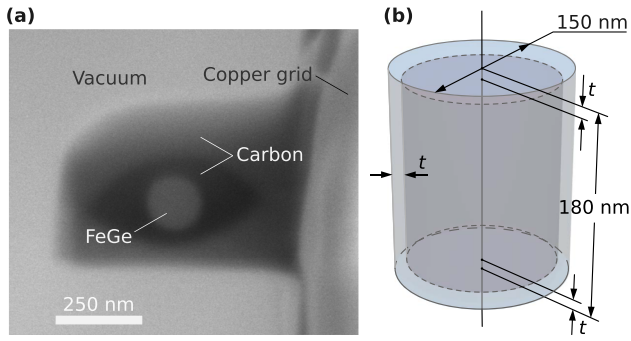
The diversity of three-dimensional magnetic states in a FeGe nanocylinder is studied using micromagnetic simulations and off-axis electron holography in the transmission electron microscope. In particular, we report the observation of a dipole string—a spin texture composed of two coupled Bloch points—which becomes stable under geometrical confinement. Quantitative agreement is obtained between experimental and theoretical phase shift images by taking into account the presence of a damaged layer on the surface of the nanocylinder. The theoretical model is based on the assumption that the damaged surface layer, which results from focused ion beam milling during sample preparation, has similar magnetic properties to those of an amorphous FeGe alloy. The results highlight the importance of considering the magnetic properties of the surface layers of such nanoscale samples, which influence their magnetic states.

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## I. INTRODUCTION

Cubic chiral magnets with bulk Dzyaloshinskii–Moriya interaction (DMI)<sup>1,2</sup> are known to host a wide range of noncollinear magnetic textures, such as helical spirals,<sup>3,4</sup> skyrmions,<sup>5–7</sup> anti-skyrmions,<sup>8</sup> skyrmion bags,<sup>9–11</sup> braids,<sup>12</sup> chiral bobbles,<sup>13,14</sup> and heliknotons.<sup>15</sup> The most commonly used method to visualize such magnetic states is transmission electron microscopy (TEM), which is able to provide nanometer-sized spatial resolution in real space. While conventional Lorentz TEM has been widely used for observations of magnetic states in extended plates,<sup>6,7</sup> it is less suitable for

studies of geometrically confined nanoscale samples, such as disks<sup>16</sup> and stripes,<sup>17</sup> in part because the *out-of-focus* imaging condition results in the presence of Fresnel fringes at the sample edges, which can dominate the magnetic contrast of primary interest. Contrast arising at the sample edges can be subtracted more easily by using the *in-focus* technique of off-axis electron holography, which provides direct access to all spatial frequencies of the electron optical phase shift.<sup>18,19</sup> Here, we use the off-axis electron holography technique to study a FeGe nanocylinder with a diameter of 150 nm and a thickness of 180 nm that was prepared using a focused ion beam (FIB) milling, as shown in Fig. 1(a). In contrast to prior work on



**FIG. 1.** (a) Scanning electron micrograph of a FeGe nanocylinder cut from a bulk crystal using FIB milling (see the supplemental video in Ref. 16). The image was recorded using a short exposure time to avoid damage to the sample by the electron beam. C coating was used to minimize damage to the sample during FIB preparation. (b) Schematic illustration of the sample geometry.  $t$  is the thickness of the damaged layer formed due to FIB sample preparation.

a FeGe nanodisk<sup>16</sup> that had a diameter of 160 nm and a thickness of 90 nm, different magnetic states are observed in this cylindrical sample.

Following a well-established approach based on micromagnetic simulations, we calculate theoretical phase shift images corresponding to the lowest energy states and compare them to experimental phase images recorded using off-axis electron holography. Our results show that magnetic states in nanoscale samples cut from bulk crystals using FIB milling can be strongly affected by the presence of a damaged surface layer, as schematically shown in Fig. 1(b). We assume that (i) the damaged layer has a disordered or amorphous structure, (ii) the DMI is negligibly small due to the absence of crystal order in the damaged layer, and (iii) the net magnetization and Heisenberg exchange coupling are nearly the same as those in the inner, undamaged region of B20-type crystals. By varying the thickness of the damaged layer in our micromagnetic model, we obtain quantitative agreement between theoretical phase shift images and experimental measurements. One of the experimentally observed states is identified as an exotic magnetic state composed of two Bloch points of opposite topological indices, which is known in the literature as a toron,<sup>20,21</sup> monopole–antimonopole pair,<sup>22</sup> and dipole string.<sup>23–25</sup>

## II. MICROMAGNETIC CALCULATIONS

For theoretical analysis of magnetic states in a sample of confined geometry, we use a standard model for isotropic chiral magnets that includes Heisenberg exchange, DMI, and demagnetizing fields. The energy density functional can be written in the form<sup>12,26</sup>

$$\mathcal{E} = \int_{V_m} d\mathbf{r} \mathcal{A} \sum_{i=x,y,z} |\nabla m_i|^2 + \mathcal{D} \mathbf{m} \cdot (\nabla \times \mathbf{m}) - M_s \mathbf{m} \cdot \mathbf{B} + \frac{1}{2\mu_0} \int_{\mathbb{R}^3} d\mathbf{r} \sum_{i=x,y,z} |\nabla A_{d,i}|^2, \quad (1)$$

where  $\mathbf{m}(\mathbf{r}) = \mathbf{M}(\mathbf{r})/M_s$  is the magnetization unit vector field,  $M_s$  is the saturation magnetization,  $\mu_0$  is the vacuum permeability,  $\mathbf{A}_d(\mathbf{r})$

is the component of magnetic vector potential induced by the magnetization, and  $\mathcal{A}$  and  $\mathcal{D}$  are the Heisenberg exchange constant and DMI constant, respectively. In general, it is assumed that the magnetic field  $\mathbf{B}(\mathbf{r})$  is the sum of the external magnetic field  $\mathbf{B}_{\text{ext}}$  and the demagnetizing field produced by the sample itself, i.e.,  $\mathbf{B} = \mathbf{B}_{\text{ext}} + \nabla \times \mathbf{A}_d$ .

Here, we only consider the case of zero external magnetic field,  $B_{\text{ext}} = 0$ . The last two terms in Eq. (1), therefore, represent the demagnetizing field energy. We used the following material parameters for FeGe:<sup>8,12,14</sup>  $\mathcal{A} = 4.75 \text{ pJ m}^{-1}$ ,  $\mathcal{D} = 0.853 \text{ mJ m}^{-2}$ , and  $M_s = 384 \text{ kA m}^{-1}$ . The sample is assumed to have a cylindrical shape with a diameter of 150 nm and a height of 180 nm, as shown schematically in Fig. 1(b).

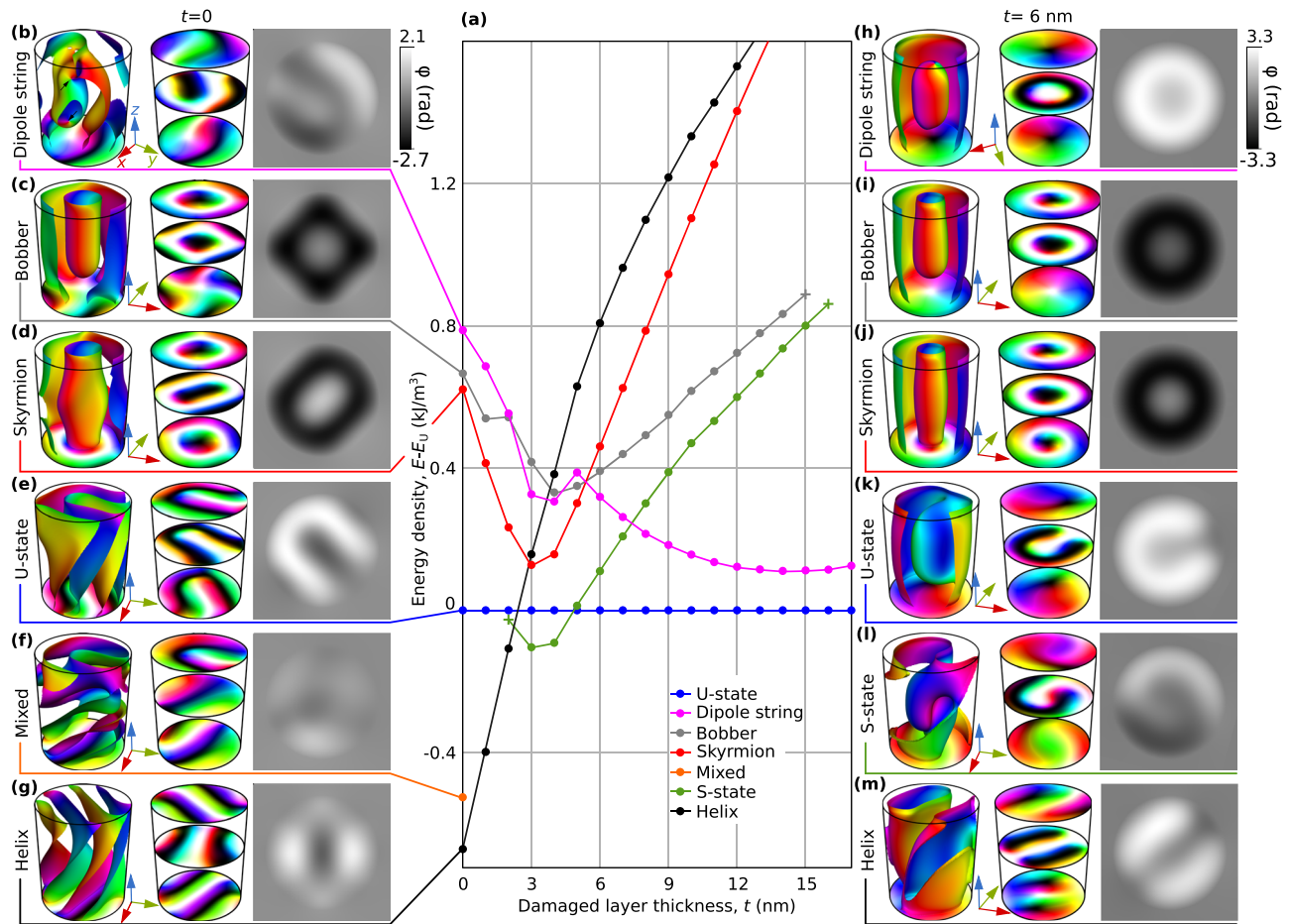
The damaged surface layer of thickness  $t$  is assumed to have the same material parameters as above, but the DMI is set to zero, which can be justified as follows: Because of the damage produced by Ga ions during FIB preparation, the ordered crystal structure that is present in the sub-surface layers of the sample is assumed to be broken. The properties of such a disordered state are expected to be similar to those of a well studied amorphous FeGe alloy. In particular, it is known that, at nearly equal concentrations of Fe and Ge, the Curie temperature of amorphous FeGe<sup>27,28</sup> is close to that of the corresponding crystalline B20 structure.<sup>29</sup> At the same time, amorphous FeGe does not exhibit non-collinear magnetic order, at least, at temperatures below  $T = 130 \text{ K}$ .<sup>30</sup> It is, therefore, reasonable to expect that the exchange stiffness in amorphous FeGe is close to that in a B20-type crystal, but that the contribution of the DMI is negligibly small. The saturation magnetization of amorphous FeGe at  $T = 100 \text{ K}$ <sup>28,30</sup> is also reported to be nearly identical to that of B20-type FeGe.

The mesh size in our simulations was fixed to be 1 nm in all spatial directions. This choice allows the impact of the damaged layer to be studied by varying its thickness in steps of 1 nm. Stable solutions of the functional (1) were found by numerical energy minimization using the Excalibur code.<sup>31</sup> In order to ensure reproducibility of the results, we also performed a check using the publicly available Mumax<sup>32</sup> code. The corresponding Mumax script is provided in the [supplementary material](#).

In order to identify the lowest energy states in the FeGe nanocylinder, we performed a large number of energy minimization cycles using different initial states. Representative results of these calculations are shown in Fig. 2 for the states with the lowest energies. The magnetic texture of each state in Figs. 2(b)–2(m) is shown in the form of both an isosurface  $m_z = 0$  and a cross section of the simulated domain in the top, middle, and bottom planes. The color code reflects the direction of the magnetization according to the standard color scheme used in Mumax, where white and black denote  $m_z = 1$  and  $m_z = -1$ , respectively, while red–green–blue correspond to the angle between the  $m_{xy}$  projection of the magnetization and the  $x$  axis. The third image in each row is the electron optical phase shift  $\varphi$  calculated with the incident electron beam antiparallel to the  $z$  axis, according to the expression<sup>33</sup>

$$\varphi(x, y) = \frac{2\pi e}{h} \int_{-\infty}^{+\infty} dz \mathbf{A}_d(\mathbf{r}) \cdot \hat{\mathbf{e}}_z, \quad (2)$$

where  $e$  is a positive elementary charge and  $h$  is Planck's constant. The results shown in (b)–(g) correspond to the case of no damaged



**FIG. 2.** Results of micromagnetic simulations for the FeGe nanocylinder for different thicknesses of the damaged surface layer. (a) Energy densities of representative stable magnetic configurations plotted as a function of damaged layer thickness with respect to the energy of the U-state shown in (e). Crosses indicate the critically damaged layer thickness above (or below) which the state becomes unstable. The left and center images in (b)–(m) show the magnetization vector field as isosurfaces  $m_z = 0$  and cross sections of the simulated cylindrical domain in three equidistant planes, respectively. The black arrows in (b) mark the positions of Bloch points. The right images in (b)–(m) show the calculated electron phase shift  $\phi$  when the electron beam is parallel to the main axis of the cylinder. The gray scale bar in (b) applies to all of the images in (b)–(g). The gray scale bar in (h) applies to all of the images in (h)–(m).

layer (i.e.,  $t = 0$ ), while the results shown in (h)–(m) correspond to a damaged layer of thickness  $t = 6$  nm.

Figure 2(a) summarizes the energies of the different states calculated as a function of damaged layer thickness. The energies are provided as differences with respect to the energy of the U-state, which represents the ground state of the system in the case of a thick damaged layer. The diagram shows that the energies depend strongly on the thickness of the damaged layer. States that contain Bloch points, the dipole string depicted in (b) and (h), or the chiral bobber depicted in (c) and (i) never appear to be the lowest energy states, as expected since such singularities typically lead to an increase in Heisenberg exchange, which is the leading energy term. In contrast, for  $t \geq 8$  nm, the dipole string becomes a metastable state, whose energy is slightly higher than that of the ground state and much lower than the energies of other states (e.g., a skyrmion). The energy of the dipole string reaches its minimum at  $t \approx 14$  nm

and then increases with  $t$ . Remarkably, in contrast to the situation for a 90-nm-thick FeGe nanodisk,<sup>16</sup> the skyrmion is never the ground state of the system, either in the presence or in the absence of a damaged layer. For  $t \geq 6$  nm, the energy of a skyrmion is even higher than that of a chiral bobber. The U-state in (e) and (k) can be considered as the helical state depicted in (g) and (m) but is slightly distorted by additional modulations across the thickness. The mixed state in (f) corresponds to a mixture of a helix and a U-state and has an energy in between these two states. In the range  $0 < t < 3$  nm, a few states correspond to mixtures of a U-state and a helix. However, they are not shown in the figure, as their energies are always higher than the helix energy.

The magnetic configuration corresponding to the S-state shown in Fig. 2(l) can, in principle, also be thought of as a distorted helix. A distinctive feature of this state is the characteristic contrast of the phase shift, which resembles a Yin-Yang symbol.



In Sec. III, we show that such contrast is observed experimentally using off-axis electron holography. Another remarkable feature of the S-state is its finite range of existence. It remains stable only in the  $3 \text{ nm} \leq t \leq 16 \text{ nm}$  range of damaged layer thickness. For  $t < 3 \text{ nm}$ , it converges to a helix or one of the mixed states, while for  $t > 16 \text{ nm}$ , it converges to a U-state.

Since we consider only the case of  $B_{\text{ext}} = 0$ , each of the states shown in Fig. 2 has an energetically equivalent state with the opposite net magnetization. For instance, the magnetization in the core of the dipole string depicted in (h) is along the positive direction of the  $z$  axis, but the dipole string with the opposite magnetization in the center has the same energy. Since the DMI conserves the chirality of the magnetization, the phase shift images of such configurations are distinct only by an inversion of the contrast.

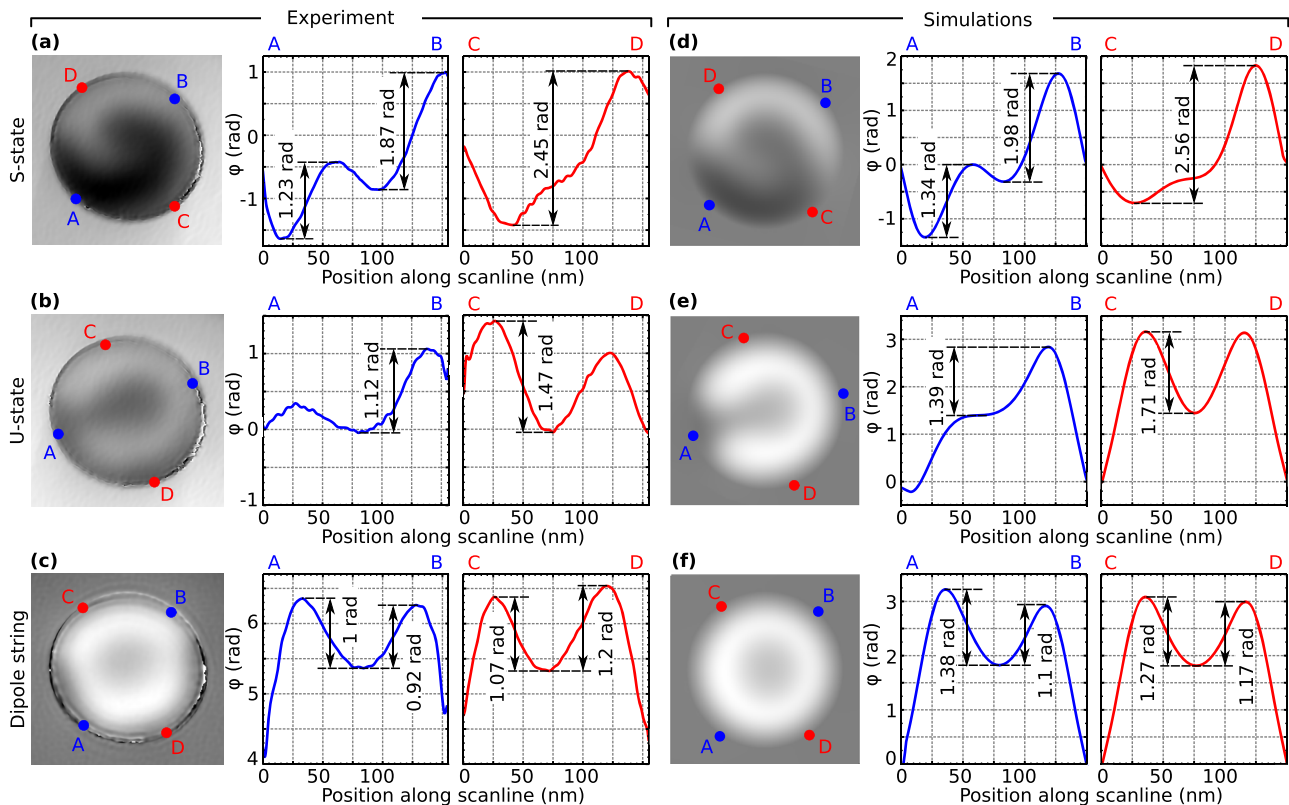
In Sec. III, we compare experimental measurements with the micromagnetic simulations.

### III. EXPERIMENTAL OBSERVATIONS

Magnetic states were recorded using off-axis electron holography in an FEI Titan G<sup>2</sup> 60–300 TEM operated at 300 kV. The microscope was operated in Lorentz mode with the objective lens switched off to provide magnetic-field-free conditions at the

position of the specimen. A liquid N<sub>2</sub> specimen holder (Gatan model 636) was used to control the sample temperature to be  $\sim 95 \text{ K}$ . Off-axis electron holograms were recorded using an electrostatic biprism positioned in the conventional selected-area aperture plane of the microscope on a  $4\text{k} \times 4\text{k}$  Gatan K2 IS direct electron counting detector. To improve the signal-to-noise ratio, 30 object holograms and 30 vacuum reference holograms, with a 4 s exposure time for each hologram, were recorded for each magnetic state. The holograms were analyzed using a standard fast Fourier transform algorithm. In order to remove the mean inner potential (MIP) contribution to the total phase shift, the phase images were recorded both at low temperature and at room temperature. They were then aligned and subtracted from each other on the assumption that the MIP contribution was the same and that there were no significant changes in specimen charging or dynamical diffraction.

Examples of experimentally measured phase images are shown in Figs. 3(a)–3(c), alongside phase line profiles extracted along segments AB and CD passing through the minimum and maximum values of  $\varphi$ . Different states were observed on performing multiple cycles of demagnetization of the sample. Two of these states, which are shown in Figs. 3(a) and 3(b), appeared with nearly identical frequency, while the state shown in Fig. 3(c) was observed more seldom. We infer that the magnetic states in (a) and (b) have the low-



**FIG. 3.** Quantitative comparisons between experimental and theoretical phase shift images for the FeGe nanocylinder. The left column in (a)–(c) shows experimental images. The left column in (d)–(f) shows corresponding theoretical images calculated for a damaged layer thickness  $t = 6 \text{ nm}$ . The latter images are identical to those shown in Figs. 2(i), 2(k), and 2(h), respectively. Corresponding phase shift profiles along the lines AB and CD are shown in the center and right columns.

est energies, whereas that in (c) is metastable. While the energies of the states in (a) and (b) are thought to be comparable, it is difficult to identify which of them represents the ground state.

The theoretical images for the state calculated without a damaged layer shown in Figs. 2(b)–2(g) do not fit the experimental images well (see also the [supplementary material](#), Fig. S1). There are some similarities between Fig. 2(b) and the experimental image shown in Fig. 3(a). However, the dipole string without a damaged layer ( $t = 0$ ) represents a high energy state and thus should not appear experimentally more often than lower energy states. Moreover, a quantitative comparison of the phase shift signal in Fig. 2(b) does not match the experimental images (see the [supplementary material](#), Fig. S1, for details). In contrast, the micromagnetic simulation calculations for a damaged layer thickness of 6 nm show good agreement with the experimental results. In particular, the experimental phase image and corresponding line profiles in (a) match the calculations for the S-state in (d). It should be noted that only phase shift differences across each image, rather than the absolute value of the phase shift, are relevant for such comparisons. The experimental results in (b), in turn, perfectly fit the U-state shown in (e). Finally, the theoretical image for the dipole string in (f) provides the best fit to the experimental image shown in (c). Although the theoretical images of the chiral bobber and skyrmion shown in Figs. 2(h) and 2(i) also provide contrast similar to that in Fig. 3(c), only the dipole string fits the experimental data on a quantitative level (see the [supplementary material](#), Fig. S2). Moreover, according to the micromagnetic simulations, the energies of the skyrmion and chiral bobber for  $t = 6$  nm are higher than that of dipole string. Therefore, it is reasonable to expect that the dipole string occurs with a higher probability than the skyrmion or chiral bobber.

In our experimental measurements, we also imaged a state whose contrast and absolute values of phase shift were close to those expected for a chiral bobber (see the [supplementary material](#), Fig. S3). Because of elliptical distortions present in this experimental image, we could not state with confidence that this phase image corresponds to a chiral bobber. We, therefore, do not draw this conclusion in the present work. In agreement with the theoretical prediction, we did not experimentally observe a skyrmion state, whose energy is even higher than that of the dipole string and the chiral bobber.

Small discrepancies between the measured and calculated phase shift differences can be attributed to imperfections of the sample geometry and local deviations in the thickness of the damaged layer in the real sample.<sup>34,35</sup> In addition, material parameters, such as  $M_s$  and  $A$ , in the damaged layer may not be identical to those for B20-type FeGe. The damaged layer may contain  $\text{Ga}^+$  ions and oxygen. Its stoichiometric composition may also deviate slightly from that of B20-type FeGe.<sup>36</sup> However, we can exclude the possibility that the damaged layer is nonmagnetic. Details of the corresponding calculations are provided in the [supplementary material](#), Fig. S4. As the thickness of such a FIB-damaged layer is typically 4–10 nm,<sup>35,36</sup> our estimation of  $t = 6$  nm fits this range.

#### IV. CONCLUSIONS

Experimental electron optical phase shift images of a nanocylinder of B20-type FeGe recorded using off-axis electron holography have been interpreted using a micromagnetic model

that takes into account the presence of a surface damaged layer, resulting from the use of FIB milling during sample preparation. On the assumption that the damaged layer is disordered and effectively behaves as amorphous FeGe, we obtain quantitative agreement between experimental and theoretical images for an estimated thickness of the damaged layer of ~6 nm.

One of the experimentally observed states is identified as a dipole string (or toron)—a spin configuration that is composed of two Bloch points of opposite topological charge. To the best of our knowledge, this is the first direct observation of such a state. An alternative approach for stabilizing a dipole string state was presented by Liu *et al.*,<sup>22</sup> who suggested that it can be stabilized in a nanocylinder of a chiral magnet by perpendicular pinning of the magnetization at the top and bottom bases of the cylinder. Such pinning was assumed to be introduced by an additional magnetic layer with strong perpendicular magnetic anisotropy sputtered onto the bases of the nanocylinder. Instead, we show how a dipole string can be stabilized naturally by effectively softening the surface layer by introducing damage by FIB milling. Moreover, we show that even a small variation in the damage layer thickness can change the energy balance across a large diversity of possible magnetic states. Further studies of the properties of such damaged layers may suggest practical applications for the reliable control of different magnetic states in magnetic nanostructures.

#### SUPPLEMENTARY MATERIAL

See the [supplementary material](#) for Figs. S1–S4 and the Mumax script.

#### ACKNOWLEDGMENTS

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#### AUTHOR DECLARATIONS

##### Conflict of Interest

The authors have no conflicts to disclose.

##### Author Contributions

**Andrii S. Savchenko:** Data curation (equal); Formal analysis (lead); Investigation (lead); Visualization (lead); Writing – original draft (lead). **Fengshan Zheng:** Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal). **Nikolai S. Kiselev:** Conceptualization (lead); Formal analysis (supporting); Investigation (supporting); Methodology (lead); Software (supporting); Supervision (equal); Writing – review & editing (lead). **Luyan Yang:** Data curation (equal); Formal analysis (equal). **Filipp N. Rybakov:** Methodology (lead); Software (lead). **Stefan Blügel:** Funding acquisition (equal); Project administration (equal); Resources (equal). **Rafal E. Dunin-Borkowski:** Funding acquisition (lead); Project administration (lead); Resources (lead); Supervision (equal); Writing – review & editing (supporting).

## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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