

Room temperature magnetic vortices in the van der Waals magnet  $\text{Fe}_5\text{GeTe}_2$ Elias Sfeir,<sup>1</sup> Carolin Schrader,<sup>1</sup> Florentin Fabre<sup>1</sup>, Jules Courtin<sup>2</sup>, Céline Vergnaud,<sup>2</sup> Alain Marty,<sup>2</sup> Matthieu Jamet,<sup>2</sup> Frédéric Bonell<sup>2</sup>, Isabelle Robert-Philip<sup>1</sup>, Vincent Jacques,<sup>1</sup> and Aurore Finco<sup>1,\*</sup><sup>1</sup>*Laboratoire Charles Coulomb, Université de Montpellier, CNRS, Montpellier, France*<sup>2</sup>*Université Grenoble Alpes, CNRS, CEA, SPINTEC, 38054 Grenoble, France*

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We investigate the effect of confinement on the magnetic state of a 12-nm-thick  $\text{Fe}_5\text{GeTe}_2$  layer grown by molecular beam epitaxy. We use quantitative scanning nitrogen-vacancy magnetometry to locally extract the magnetization in rectangular uniformly in-plane magnetized microstructures, showing no enhancement of the Curie temperature compared to magnetization measurements performed before patterning the film, in contrast to previous results obtained on thick  $\text{Fe}_3\text{GeTe}_2$  flakes. Under the application of a weak out-of-plane magnetic field, we observe the stabilization of magnetic vortices at room temperature in micrometric squares. Finally, we highlight the effect of the size of the patterned microdisks and microsquares on the stabilization of the vortices using experiments and micromagnetic simulations. Our work thus proposes and demonstrates a way to stabilize noncollinear textures at room temperature in a van der Waals magnet using confinement.

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## I. INTRODUCTION

Among the broad diversity of van der Waals magnets discovered in recent years, very few possess a Curie temperature  $T_C$  above room temperature [1], which is a crucial requirement for technological applications. In particular,  $\text{Fe}_5\text{GeTe}_2$  possesses a  $T_C$  of about 300 K in bulk [2–4], which remains high (>200 K) in few-layers-thick films or flakes [2,5–8]. It was also shown that its Curie temperature can be significantly enhanced above room temperature by substituting Fe with Co [9] or Ni [10]. This material is particularly interesting, as several groups have reported the observation or the prediction of magnetic bubbles, merons, and skyrmions [11–18], with discussions about the possible presence of a sizable bulk Dzyaloshinskii-Moriya interaction (DMI) [19,20] and about the role of oxidation in their stabilization [21,22]. The magnetic anisotropy in  $\text{Fe}_5\text{GeTe}_2$  flakes has also been found to significantly vary [23]. Therefore, the nucleation and stabilization of these complex magnetic textures remains only partially controlled.

A solution could be to take advantage of dipolar energy and use confinement in order to stabilize whirling textures in micromagnetic structures. The effect of such patterning on  $\text{Fe}_5\text{GeTe}_2$  has not been studied yet, but on the parent compound  $\text{Fe}_3\text{GeTe}_2$ , surprising effects have been observed. Li *et al.* [24] have reported X-ray magnetic circular dichroism and photoelectron emission microscopy experiments showing the presence of a spin reorientation transition from out of plane to in plane at 230 K in patterned  $\text{Fe}_3\text{GeTe}_2$  flakes, together with a dramatic increase of  $T_C$  above room temperature. The resulting microstructures thus exhibit a vortex state at 300 K. As pointed out by Li *et al.*, it remains unclear whether this strong variation of magnetic properties origi-

ates from size effects or from a chemical alteration of the material during microfabrication. Here, we investigate the influence of patterning on the magnetic state of a  $\text{Fe}_5\text{GeTe}_2$  thin film grown by molecular beam epitaxy (MBE) [7], which is thus already ferromagnetic at room temperature. In particular, we study the effect of confinement on the stabilization of noncollinear magnetic textures such as vortices. In order to perform this analysis and to obtain local information about  $T_C$  by extracting the value of the magnetization in different microstructures, we use scanning nitrogen-vacancy (NV) center magnetometry [25]. This technique allows us to map quantitatively the magnetic stray field produced by the  $\text{Fe}_5\text{GeTe}_2$  microstructures, nonperturbatively and under ambient conditions. All the measurements presented in this work have been performed with a commercial scanning NV center microscope (ProteusQ, Qnami).

## II. METHODS

Figure 1(a) presents a sketch of the experiment. Our sample is a 11.8-nm-thick  $\text{Fe}_5\text{GeTe}_2$  film grown by MBE on  $\text{Al}_2\text{O}_3(0001)$  by coevaporation of Fe, Ge, and Te with a flux ratio  $\text{Fe}:\text{Ge} = 5$  and a slight excess of Te. The substrate temperature was 450 °C, as measured with a thermocouple located close to the sample holder. The growth was performed at a rate of 1 monolayer per minute, followed by annealing under Te flux up to 580 °C for 5 min. The sample was then cooled down to room temperature at a rate of 5–10 K min<sup>−1</sup>, then capped with a protective 3-nm-thick Al layer that naturally oxidized in air [7]. The film has been subsequently patterned with electron beam lithography and Ar ion etching in order to create microstructures of various sizes and shapes (squares, rectangles, and disks). Macroscopic characterization of a typical MBE-grown film of the same thickness before patterning using Superconducting Quantum Interference Device (SQUID) magnetometry is presented in

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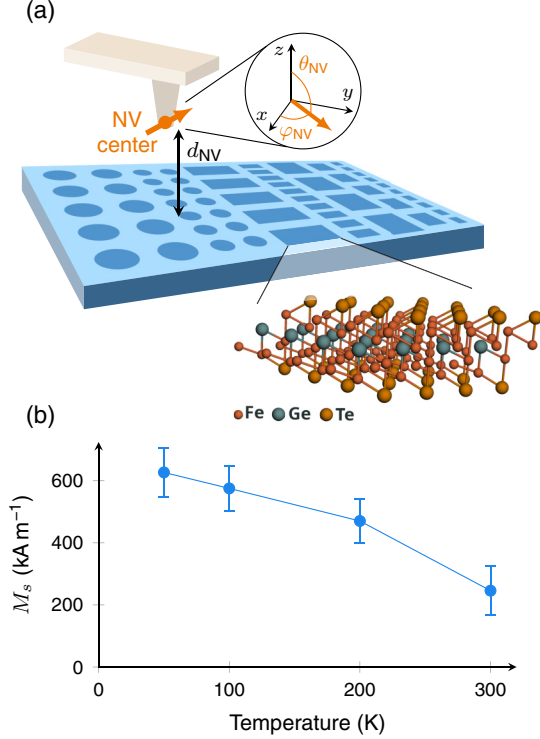


FIG. 1. (a) Sketch of the experiment. We use a diamond scanning probe to perform NV magnetometry experiments on a microstructured  $\text{Fe}_5\text{GeTe}_2$  film. The top inset defines the angles  $\theta_{\text{NV}}$  and  $\varphi_{\text{NV}}$  which describe the orientation of the NV center in the laboratory frame. The bottom inset displays the structure of a monolayer of  $\text{Fe}_5\text{GeTe}_2$ , drawn using the pyXtal library [26] and a cif file from the Computational 2D Materials Database [27,28]. (b) Typical SQUID measurement of  $M_s$  in a 12-nm-thick MBE grown film, indicating a Curie temperature above 300 K before patterning.

Fig. 1(b) and indicates that the film is magnetized in plane and exhibits a Curie temperature slightly above room temperature, with a saturation magnetization  $M_s = 246 \pm 80 \text{ kA m}^{-1}$  at 300 K. A temperature-dependent magneto-optical Kerr effect measurement performed on the patterned sample before microfabrication is shown in the Supplemental Material [29] and confirms that the film is ferromagnetic at 300 K.

### III. RESULTS

In a first series of measurements presented in Fig. 2, we perform scanning NV center magnetometry on microstructures of different sizes. The sample was initially in a demagnetized state since the microfabrication process involved an annealing above  $T_C$ . We use a permanent magnet to apply a tilted external magnetic field of 5.4 mT roughly aligned with the quantization axis of the NV center, which is described by the angles  $\theta_{\text{NV}} \simeq 125^\circ$  and  $\varphi_{\text{NV}} \simeq 87^\circ$  defined in Fig. 1(a). The field is sufficient to obtain rectangular microstructures hosting a single uniform magnetic domain, and its out-of-plane component is low enough to avoid tilting the magnetization  $\vec{M}$  out of the  $ab$  plane. In this case, we expect to detect magnetic stray field at the edges orthogonal to  $\vec{M}$ , and no field at the edges parallel to  $\vec{M}$ . This corresponds perfectly

to the experimental data shown in Fig. 2 for four different rectangular microstructures.

Since our measurements are quantitative, we can compute the saturation magnetization  $M_s$  from each map, by fitting the stray field to its analytical expression. To achieve this, we first extract a stray field profile across the edges which produce stray field. The positions of these line profiles are indicated by the black lines on the images in Fig. 2. With scanning NV center magnetometry, we measure the component  $B_{\text{NV}}$  of the stray field, which is its projection along the quantization axis of the NV center and can therefore be written as

$$B_{\text{NV}} = \sin \theta_{\text{NV}} \cos \varphi_{\text{NV}} B_x + \sin \theta_{\text{NV}} \sin \varphi_{\text{NV}} B_y + \cos \theta_{\text{NV}} B_z. \quad (1)$$

The magnetic stray field produced at an edge parallel to the  $y$  axis and located at  $x = x_0$ , with  $\vec{M}$  orthogonal to the edge and measured at a height  $d_{\text{NV}}$ , can be written as [29]

$$B_x(x) = \frac{\mu_0 M_s}{2\pi} \left[ \arctan \left( \frac{d_{\text{NV}} - \frac{t}{2}}{x - x_0} \right) - \arctan \left( \frac{d_{\text{NV}} + \frac{t}{2}}{x - x_0} \right) \right],$$

$$B_y(x) = 0,$$

$$B_z(x) = \frac{\mu_0 M_s}{4\pi} \left\{ \log \left[ \left( d_{\text{NV}} - \frac{t}{2} \right)^2 + (x - x_0)^2 \right] - \log \left[ \left( d_{\text{NV}} + \frac{t}{2} \right)^2 + (x - x_0)^2 \right] \right\}, \quad (2)$$

where  $t$  is the film thickness. We can fit the experimental line profiles to a combination of Eqs. (1) and (2), considering two opposite edges in each profile. Although the distance  $d_{\text{NV}}$  between the NV center and the surface as well as the angles  $\theta_{\text{NV}}$  and  $\varphi_{\text{NV}}$  can be calibrated independently, we consider  $d_{\text{NV}}$  as a free parameter in the fits because of the presence of resist residues on the surface of the sample. These fits are plotted in the graphs at the bottom of Fig. 2, where the resulting value of  $M_s$  is also indicated. The main conclusion from this analysis is that  $M_s$  does not vary significantly with the size of the microstructure. However, we note that it seems that  $M_s$  decreases slightly when the area of the rectangle decreases, although the values do not deviate much from the error bars. This could be related to moderate damage made to the film during the patterning procedure. Since the average value that we obtain,  $M_s = 202 \pm 12 \text{ kA m}^{-1}$ , is in good agreement with the value of  $M_s$  measured by SQUID at 300 K, we conclude that the microstructuring does not help increasing  $T_C$  in our experiment. In particular, we do not observe the strong enhancement reported in Ref. [24] for  $\text{Fe}_3\text{GeTe}_2$ . This suggests that these results were probably the consequence of a chemical alteration of  $\text{Fe}_5\text{GeTe}_2$  by Ga implantation. It has indeed recently been shown that replacing Ge with Ga leads to an increase of  $T_C$  up to 350–380 K in  $\text{Fe}_3\text{GeTe}_2$  [30] and  $\text{Fe}_5\text{GeTe}_2$  [31] and to the observation of skyrmions in thick flakes of  $\text{Fe}_3\text{GaTe}_2$  [32,33]. Theoretical investigations attribute this effect to higher-order exchange coefficients which are positive in  $\text{Fe}_3\text{GaTe}_2$  in contrast to  $\text{Fe}_3\text{GeTe}_2$  in which they are negative [34].

In a second series of measurements, we reduce the applied magnetic field to 3.6 mT out of plane. This field allows us to determine the sign of the measured magnetic field during

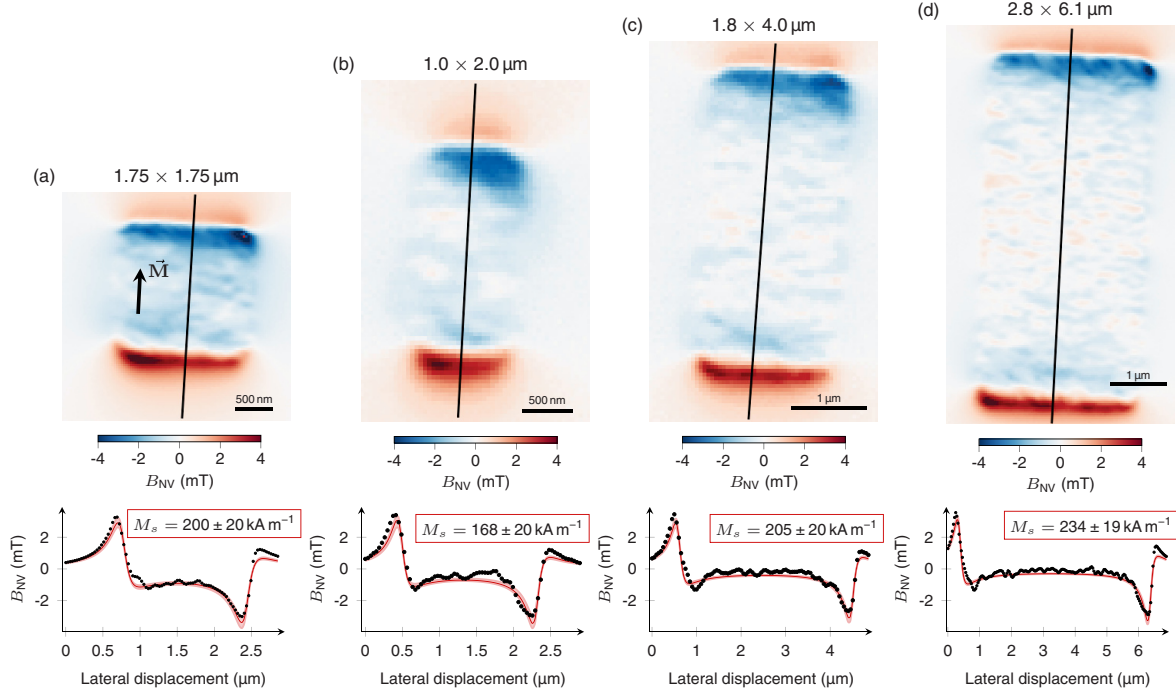


FIG. 2. Measurement of  $M_s$  in microstructures of different sizes. The maps show the measured stray magnetic field under a tilted external magnetic field of 5.4 mT, allowing us to stabilize a single domain in each rectangle. The plots below each map show the profiles extracted along the black lines and the corresponding fits with Eqs. (1) and (2), together with the extracted  $M_s$  value.

the NV magnetometry scans [35] and has a limited effect on the magnetic texture inside the  $\text{Fe}_5\text{GeTe}_2$  microstructures. Similarly to what was observed in the images of the  $\text{Fe}_3\text{GeTe}_2$  microstructures in Ref. [24], our magnetic stray field maps reveal the presence of magnetic vortices in some of the square structures [see Figs. 3(a) and 3(b)]. Indeed, we detect a flower-shaped stray field pattern, which is typical for vortices [36], in squares of different sizes and orientations with respect to the crystal structure of  $\text{Fe}_5\text{GeTe}_2$ .

However, as the vortex cores are not visible in our images, we performed additional micromagnetic simulations to really ensure that we are observing vortices. We used the Python package Ubermag [37] with OOMMF [38] to compute the relaxed magnetic configurations in the two squares considered in Fig. 3, and computed the associated magnetic stray field map at the NV center height using a scalar potential method

TABLE I. Parameters used in the micromagnetic simulations and the subsequent stray field calculations.

Parameter	Value
NV height $d_{\text{NV}}$	120 nm
NV polar angle $\theta_{\text{NV}}$	125°
NV azimuthal angle $\varphi_{\text{NV}}$	87°
Exchange stiffness $A$	10 pJ m <sup>-1</sup>
Uniaxial anisotropy $K_u$	-20 kJ m <sup>-3</sup>
Film thickness $t$	11.8 nm
Saturation magnetization $M_s$	202 kA m <sup>-1</sup>
$M_s$ disorder, patch size	~50 nm
$M_s$ disorder, variation amplitude	± 20%

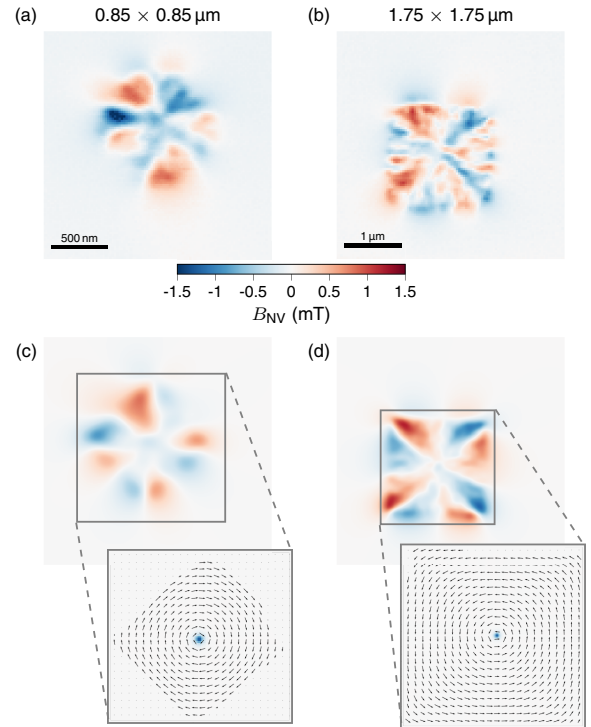


FIG. 3. (a), (b) Measured stray field maps of vortices in square microstructures. (c), (d) Computed stray field maps resulting from the magnetic state shown in the insets. The insets show the magnetization configurations obtained from micromagnetic calculations with the parameters from Table I, with the blue color indicating the core of the vortices where the magnetization tilts out of plane.

[39]. We write the magnetic field as the gradient of a scalar potential  $\phi$  verifying the Poisson equation below [Eq. (3)], which we solve in Fourier space for computational efficiency.

$$\Delta\phi = \nabla \cdot \vec{M}. \quad (3)$$

We extracted the shape of the microstructures from the topography measurements, and used the parameters gathered in Table I. We chose a value for the exchange stiffness which corresponds to the order of magnitude given in the literature,  $10 \text{ pJ m}^{-1}$  [13,16,17]. Macroscopic measurements of the uniaxial anisotropy  $K_u$  shown in Fig. S3(a) [29] indicate that it is negative and vanishes around room temperature. Using the slightly positive value of  $18 \pm 30 \text{ kJ m}^{-3}$  measured at 300 K results in the presence of a large and clearly visible vortex core in the center of the square microstructure (see Fig. S4), which does not match with our experiments. We have therefore varied  $K_u$  below this value in order to get images which are in better agreement with experimental data, and used  $K_u = -20 \text{ kJ m}^{-3}$ . Using this value, the vortex core cannot be distinguished in the stray field images, in agreement with our measurements. In addition, although the presence of DMI has been demonstrated in our films [19], we do not include it in our simulation, as the effective in-plane anisotropy does not favor the stabilization of skyrmionic states. We also added some disorder in the simulations in order to mimic local variations of thickness or of  $M_s$  by defining random grains with a Voronoi tessellation and attributing them a random  $M_s$  value, with a variation of  $\pm 20\%$  around the measured mean value  $M_s = 202 \text{ kA m}^{-1}$ . Note that this  $M_s$  distribution is not directly related to the measurement uncertainty on the  $M_s$  value in Fig. 2, which provides an average value on the microstructure, but describes very local variations resulting from disorder and thickness fluctuations. Structural characterization of our films [7] indeed shows the presence of domains, a few tens of nm in size, epitaxially oriented but with a finite mosaic spread of  $\sim 3^\circ$ . Atomic steps are also present on the surface, indicating local thickness variations on the scale of our microstructures.

The NV-to-sample distance  $d_{\text{NV}}$  was assumed rather large, 120 nm, in agreement with the fits shown in Fig. 2 (see actual values in Supplemental Material [29]). As expected, this value is higher than the one measured in the calibration because of the resist residues. The NV orientation  $\theta_{\text{NV}}$ ,  $\varphi_{\text{NV}}$  was determined from the calibration measurement (Fig. S2 [29]). Using this  $K_u$  value and including disorder, we obtain stray field maps in qualitative good agreement with the experimental ones. We therefore conclude that we are actually observing vortices in square  $\text{Fe}_5\text{GeTe}_2$  microstructures at room temperature.

We performed similar experiments in disk-shaped microstructures, and observed either magnetic single domains in very small disks (diameter below 500 nm), or complicated flower-shaped patterns most probably resulting from disorder (see Fig. S4 [29]). In the disks, in the absence of disorder, we only expect to measure stray field at the vortex core [40,41], and this field is very weak, whereas with disorder, a stray field pattern resulting exclusively from this disorder and masking the vortex core can be observed. As a consequence, our experimental data do not allow us to decide experimentally if vortices are present. Our simulations including disorder

and a vortex in the center of the disks show nevertheless a reasonable agreement with the experimental data.

Our experimental investigation of microstructures of different sizes (500 nm, 1  $\mu\text{m}$ , 2  $\mu\text{m}$ ) indicates that there is a higher probability to observe one or several vortices in the large structures than in the small ones. In particular, we found several disks of diameter below 500 nm without any vortex, such as the one shown in the left inset of Fig. 4, while the large rectangles such as the one shown in Fig. 2(d) can exhibit several vortices in a low out-of-plane field (see Supplemental Material [29]). We investigated this question further in squares and disks using micromagnetic simulations, without disorder and external magnetic field applied. We considered squares of various sizes, from 100 nm to 2  $\mu\text{m}$  as well as disks with diameters ranging from 100 nm to 2  $\mu\text{m}$ . As an initial state, we use a completely random orientation of the magnetization in each cell. We performed the simulation with 20 different random initial configurations for each size, and counted the number of realizations in which one or several vortices or antivortices are present at the end of the relaxation. Our results are gathered in Fig. 4. The same trend, in agreement with our experiments, is observed for the disks and the squares. In the very small structures of typical size 100 nm, we never find a vortex. When the size increases, the probability to observe a vortex increases, and for a size above 1.5  $\mu\text{m}$ , we systematically end up with a complicated magnetic state consisting in several vortices and/or antivortices.

#### IV. DISCUSSION

This size effect has been studied previously experimentally, analytically and numerically [42–47] in disks and squares of soft ferromagnets. The critical disk radius separating the vortex state from a roughly uniformly in-plane magnetized state in microdisks depends on the exchange stiffness, the saturation magnetization, and on the aspect ratio of the disks. In particular, most of the analytical calculations [44] have been performed for disks with a thickness larger than the exchange length [48]:

$$l_{\text{ex}} = \sqrt{\frac{2A}{\mu_0 M_s^2}}. \quad (4)$$

In our case,  $l_{\text{ex}} \simeq 20 \text{ nm}$ , and therefore our disks are significantly thinner than  $l_{\text{ex}}$ , meaning that a two-dimensional (2D) model is more relevant [46]. It appears that for very thin films, the critical radius allowing the stabilization of vortices in a disk becomes very large [47], in agreement with our results presented in Fig. 4. We also note that the diameter of the core of the vortices in our simulations, which we obtained by fitting a two-dimensional gaussian function to the simulated maps of  $M_z$ , lies between 60 and 55 nm, again much larger than  $l_{\text{ex}}$ . Comparing this large core size with the dimensions of our simulated disks and squares, it appears that, as shown in Fig. 4, the 100-nm structures are too small to allow for the proper stabilization of a vortex.

To conclude, using quantitative scanning NV center magnetometry, we demonstrated that microstructuring does not significantly affect the Curie temperature in MBE-grown  $\text{Fe}_5\text{GeTe}_2$  ultrathin films. However, by comparing experimental data with micromagnetic simulations, we revealed the



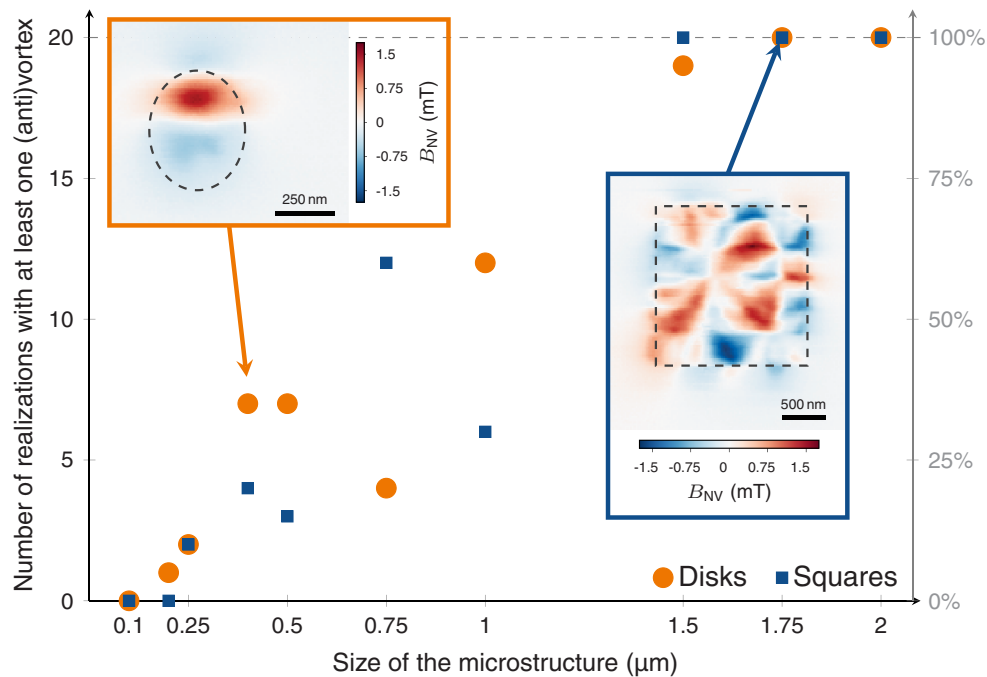


FIG. 4. Graph gathering the results of micromagnetic simulations of the magnetic state in disks and squares of various sizes, in the absence of external magnetic field and starting from random configurations. For each size and shape, 20 disorder configurations were considered. In the larger structures ( $>1.5 \mu\text{m}$ ), several vortices and/or antivortices were systematically found. The insets show experimental data of a monodomain disk of diameter  $0.4 \mu\text{m}$  and a multivortex square of size  $1.75 \mu\text{m}$ , measured under a small out-of-plane magnetic field in order to bias the NV center.

presence of magnetic vortices in microstructures. We thus demonstrated here a way to control the formation of non-collinear magnetic textures at room temperature in  $\text{Fe}_5\text{GeTe}_2$ , opening perspectives to study the rich physics and potential applications of these objects in the context of 2D and van der Waals materials.

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#### DATA AVAILABILITY

The data that support the findings of this article are openly available [49].

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