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# Superconducting Vortex-Antivortex Pairs: Nucleation and Confinement in Magnetically Coupled Superconductor-Ferromagnet Hybrids

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http://dx.doi.org/10.5772/65954

#### Abstract

Superconducting vortices are a well known class of vortices, each of them carrying a single magnetic flux quantum. In this chapter the authors present the results of low temperature Magnetic Force Microscopy experiments to investigate the nucleation and dynamics of superconducting vortices in magnetically coupled Superconductor/ Ferromagnet (S/F) heterostructures made by Nb/Py. It is here shown that by controlling the thicknesses of both S and F layer, the formation of spontaneous vortex-antivortex pairs (V-AV) can be favored and their confinement and mobility can be tuned. The experimental results are compared with two theoretical models dealing with the spontaneous nucleation of V/AV pairs in the limits of S thickness respectively greater and smaller than the London penetration depth. It is shown that vortex nucleation and confinement is regulated by the intensity of the out-of-plane component of the magnetization with respect to a critical magnetization set by the thickness of both S and F layers. Additionally, external field cooling processes were used to probe in-field vortex nucleation and V-AV unbalancing, whereas the sweeping of an external magnetic field when below the superconducting critical temperature was used to force the vortex into motion, probing the vortex mobility/rigidity and the vortex avalanche events.

Keywords: superconductivity, ferromagnetism, vortex-antivortex



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# 1. Introduction

Superconductivity has a great potential to play a significant role in the development of new clean energy technologies by minimizing the losses in electrical current transport. However, the widespread use of superconducting materials is still limited by a few critical parameters, such as critical current, critical magnetic field, and critical temperature and, therefore, the active research on new superconducting technologies is underway.

Conventional superconductors are well described in the framework of the Ginzburg-Landau (G-L) theory [1]. Within G-L theory, the introduction of the G-L parameter  $k = \frac{\xi}{\lambda}$  leads to differentiation between type I and type II superconductors for cases when  $k > \frac{1}{\sqrt{2}}$  and  $k < \frac{1}{\sqrt{2}}$ respectively. The two characteristic superconducting length scales, the coherence length ( $\xi$ ) and penetration depth ( $\lambda$ ), are intimately related to the superconducting material. The superconducting order parameter describing the local density of the superconducting Cooper pairs varies at length scale defined by the coherence length, while the penetration depth characterizes the distance at which the external magnetic field is exponentially screened from the interior of a superconducting sample (Meissner effect). In type II superconductors, the nucleation of quantized magnetic flux tubes, Abrikosov vortices, enables the persistence of the superconducting state in high applied magnetic fields up to the upper critical field  $H_{c2}$  at which the superconductivity is destroyed. The mixed state (or Shubnikov state) of type II superconductors persists in applied magnetic fields *H* between  $H_{c1} < H < H_{c2'}$  where  $H_{c1}(T)$ and  $H_{c2}(T)$  are the lower and upper critical fields, respectively, making these materials very technologically relevant. It has been theoretically predicted by Abrikosov [2] and later experimentally shown by Trauble and Essman [3] that the mixed state is a macroscopic quantum fluid having vortices each carrying a flux quantum  $\Phi_0 = \frac{h}{2e}$ , where *h* is the Planck constant and *e* is the electron charge. The vortex causes a local suppression of the superconducting order parameter on the length scale of  $\xi$  while  $\lambda$  measures the exponential decay of magnetic field and currents, when moving away from the vortex core. In 1957, Abrikosov [2] predicted a lattice arrangement of vortices (Figure 1) in order to minimize the energy of the system. The vortex lattice period d is set by the intensity of the external magnetic field Haccording to the relation  $d = \sqrt{\frac{2\Phi_0}{\sqrt{3}H}}$ . When an external current, exceeding the critical value, is applied to a superconductor in the mixed state, vortices are forced to move under the action of the Lorentz force causing energy dissipation. For this reason, control of the vortex dynamics is one of the main challenges for technological applications and fundamental science. In order

to restore a dissipation-free regime, the driving Lorentz force has to be counterbalanced by a pinning force. In this scenario, the technological applications of type II superconductors deal with the capability to create and control pinning centers that locally induce a suppression or reduction of the superconducting order parameter.

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**Figure 1.** Periodic arrangement of vortices in a type-II superconductor in an external applied magnetic field. Each vortex has a normal core, where the superconducting order parameter  $|\psi|^2$  (blue line) drops to zero on the scale of  $\xi$  while the magnetic field profile (red line) exponentially decays on the scale of  $\lambda$ .

Lattice defects, dopant inclusions or peculiar sample geometry, have been proposed in order to impose a pinning potential for the superconducting vortices [4]. An enhancement of the critical current has been reported by bulk processing of the superconductors to create pinning centers and by lithographic patterning of arrays of pinning centers. Magnetic pinning centers have also been widely used for enhancing vortex pinning properties since they locally suppress the superconducting order parameter (pair-breaking effect of the local magnetic moment) and magnetically attract vortex lines. Several methods of introducing magnetic pinning centers have been employed from deposition of magnetic nanoparticles to lithographically defining magnetic nanotextures on the superconducting layer [5–30]. Magnetically coupled superconductor/ferromagnet thin film heterostructures in which the magnetic domains in the ferromagnet act as pinning centers have been of great interest due to ease of fabrication, scalability for future applications, and due to basic fundamental physics governing the superconductivity in these hybrid systems [31–52].

In the past decades a lot of effort has been focused on developing experimental techniques for studying vortex matter at the nanoscale. Since a collective behavior of vortex dynamics can be extracted, for example, from electronic and heat transport and magnetic measurements [53–61], the real challenge lies in the capability to investigate single vortex, lattice arrangements, and local mechanism of motion with a high spatial resolution. An overall view on the vortex lattice, and its structural characteristics, can be provided by small-angle neutron scattering in the reciprocal space [62, 63], and by Bitter decoration [3, 64], time-resolved magneto-optic techniques [65–68], and holography electron microscopy [69] in real space. The first observation of isolated vortices was pioneered by Essman and Trauble [3] in 1967. In a low magnetic field, they used small magnetic particles to decorate the surfaces of different superconductors in order to get information on the arrangement of vortices in the vortex lattice. By using this technique, large areas hundreds of microns square of the sample surface can be investigated by taking a snapshot of the lattice. More recently, real space imaging of superconducting vortices has been obtained by using scanning probe microscopy [70] and scanning Hall probe

microscopy [71, 72], with a submicron spatial resolution, have been successfully used to study the geometries, dynamics, and interactions of vortices in different systems. On the other hand, scanning tunneling microscopy (STM), with a subnanometric resolution, is the only technique able to image individual vortex cores by spatially mapping the amplitude of the order parameter [73–75]. The STM method is sensitive to the electronic properties of the sample surface and thus requires clean and flat surfaces. Although it provides a unique opportunity to image vortices at high magnetic fields (due to sensitivity to the order parameter rather than the magnetic profiling), STM technique cannot distinguish between the polarity of the vortices. On the other hand, magnetic force microscopy (MFM) provides information about the vortex polarity and requires less stringent surface quality, albeit the method is constrained to low enough magnetic field as to distinguish the magnetic profiles of individual flux quantum [22, 23, 47–50, 76–79]. MFM measures the force between a magnetic tip and the local magnetic moment on the surface of the investigated sample. In vacuum, the MFM operates in the socalled noncontact regime in which nonmagnetic short range tip-sample interactions are undetected. Being directly sensitive to the strength and direction of the stray field, MFM provides information that is not easily available elsewhere.

Recently, low temperature magnetic force microscopy experiments (MFM) have been performed by the authors to observe "spontaneous" vortex-antivortex pairs (V-AV), appearing in the absence of an external magnetic field, in magnetically coupled superconductor/ferromagnet (S/F) heterostructures made by niobium and permalloy (Py, Ni<sub>80</sub>Fe<sub>20</sub>) [47–50]. A thin film of SiO<sub>2</sub> separates S and F layers in order to prevent proximity effect [80]. Since the Curie temperature  $T_c$  of Py is much higher than the superconducting critical temperature  $T_s$ , a field cooling of Nb in the spatially non-uniform Py stray field occurs. The formation of quantum fluxes with opposite polarities, vortices and antivortices (V-AV), is a consequence of the peculiar stripe-like magnetic configuration of the Py. The magnetization vector in the Py film is slightly canted with respect to the film's plane so that the stray field, coming out from the Py surface due to the small and alternating out-of-plane components, causes the nucleation of V-AV pairs and guides their motion along such magnetic channels. Here, we detail MFM experimental results on spontaneous V-AV formation in Nb/Py bilayers as well as on vortex dynamics.

# 2. Theoretical models

In this chapter, the MFM results will be discussed in quantitative comparison with two different theoretical models dealing with the two opposite limits of superconductor film thickness greater [51] and smaller [52] than the penetration depth,  $\frac{d_s}{\lambda} > 1$  and  $\frac{d_s}{\lambda} < 1$ , respectively. In the framework of S/F bilayers in which the ferromagnet exhibits alternating up-and-down out-of-plane magnetization vectors  $\pm M_{0'}$  the magnetization values required for spontaneous vortex nucleation were deduced, for given values of stripe domain width *w* of F layer, superconducting penetration depth  $\lambda$  and thickness  $d_s$  of S film. In agreement with the

models, in the magnetically coupled Nb/Py system the vortex formation is due to the  $\pm M_0$  outof-plane components of Py magnetization. Hereinafter, we will define V or AV as the quantum fluxes formed on the top of  $-M_0$  or  $+M_0$  domains, respectively. Within the considered models [51, 52], by minimizing the total free energy of the S/F system, the critical magnetizations needed to nucleate the first V-AV pair can be deduced. In the limit of  $\frac{w}{\lambda} > 1$  and  $\frac{d_s}{\lambda} > 1$ , the model of Laiho et al. has been taken into account [51]. The threshold magnetization values required to nucleate the first pair of spontaneous straight vortices  $M_{cs}$  (Figure 2a), which pierces through the superconducting film, or the first vortex semiloop  $M_{cl}$  (Figure 2b), which

is bent within the superconducting film, result in  $M_{cs} = 0.2 \frac{d_s}{w} H_{c1}$  and  $M_{cl} = \frac{H_{c1}}{8 \ln (4w/\pi\lambda)}$ , respectively. If  $M_{cl} > M_{cs'}$  the formation of straight vortices is energetically favorable, and vice versa. The energy profiles of a straight (red) and semiloop (blue) vortex are shown in **Figure 2(c)** indicating that the energy minimum of the straight vortex is achieved in the middle of the magnetic stripe domain, whereas the semiloop vortex crosses over the stripe domain wall.



**Figure 2.** 3D sketch of magnetically coupled S/F heterostructure with the F layer in the stripe regime. (a) In black and white the screening supercurrents are shown, straight vortices and antivortices piercing the superconducting layer are represented by straight arrows. (b) Semiloop vortices close inside the superconducting layer. (c) Plot of the position-dependent energy of straight vortex (red) and semiloop vortex (blue) across a stripe.

On the other hand, in the opposite limit of  $\frac{w}{\lambda} > 1$  and  $\frac{d_s}{\lambda} < 1$ , the model of Genkin et al. has to be taken into account [52], where the threshold magnetization for spontaneous straight vortex nucleation results in  $M_c = \ln\left(\frac{\lambda_{\text{eff}}}{\xi}\right)\left(\frac{\Phi_0}{(4\pi)^2}\right)\frac{1}{w\lambda_{\text{eff}}}$ , with  $\lambda_{eff}(T) = \lambda(T) \coth\left(\frac{d_s}{\lambda(T)}\right)$  [81].

Spontaneous vortex formation will thus be energetically regulated by the threshold condition  $M_0 > M_{c(s,l)}$ .

Close to the superconducting critical temperature  $T_{s'}$  the superconducting lower critical field is almost zero, and the critical magnetizations for the nucleation of spontaneous vortices are

lower than  $M_0$ . As a consequence, at  $T \sim T_{S'}$  the threshold condition  $M_0 > M_{c(s,l)}$  is always satisfied and spontaneous vortices could be formed in the Nb layer but, being  $\lambda$  divergent,  $\frac{w}{\lambda} < 1$  and there is no net magnetization within the vortex area. By further decreasing the temperature,  $H_{c1}(T)$  increases with a corresponding increase in  $M_{c(s,l)}$ . For this reason,  $M_0 < M_{c(s,l)}$  can occur and vortices can move out from the superconducting layer. Vortices escape from the S layer when  $M_0 < M_{c(s,l)}$  and  $U_{SV} < U_{BL'}$  i.e., the energy required to pin a vortex  $U_{SV} = \frac{1}{4\pi} H_{c1} \Phi_0 d_s$  is much lower than the Bean-Livingstone barrier  $U_{BL} = \frac{(\pi + 2)}{4\pi} H_{c1} \Phi_0 \lambda$  [82]. This "escape condition"  $U_{SV} < U_{BL}$  is always verified for the semiloops while it is satisfied by the straight vortices only when  $d_s < (\pi + 2)\lambda$  [51].

#### 3. S/F heterostructures

In this chapter, we focus on Nb/SiO<sub>2</sub>/Py heterostructures with 1 and 2  $\mu$ m thick Py layers and Nb thickness in the range of 50–360 nm. In all cases, a 10 nm thin insulating SiO<sub>2</sub> was placed between the S and F layers in order to have only a magnetic coupling between Nb and Py. The choice of the insulating material is not as crucial as the choice of its thickness. It should be thick enough to prevent electrical proximity effects, which are, in general, short-range (Å to few nanometers) but not as much to reduce the magnetic coupling between F and S layers, which is a long-range interaction. The experiments brought us to the conclusion that 10 nm of SiO<sub>2</sub> is sufficient to reach such a goal. All the heterostructures were made by sputtering deposition as described in [47–50].

Nb films were characterized by both transport and magnetic measurements, showing a superconducting critical temperature of  $T_s = (8.8 \pm 0.1)K$ . From transport measurements [47] and by using the dirty limit expression as derived by Gorkov [83] and Kes and Tsuei [84],  $\xi(0K) = 12$ nm and  $\lambda(0K) = 61$ nm were inferred. As a consequence, the superconducting lower critical field was calculated to be  $H_{c1}(0K) = 7200e$ . At the MFM measurement temperature of 6 K  $\xi(6K) = 21$  nm,  $\lambda(6K) = 68$  nm, and  $H_{c1}(6K) = 418$  0e have been derived.

Py is a ferromagnetic material where competing magnetic energies (magnetostatic, exchange, magneto-elastic, domain wall, and anisotropy) determine the domain configurations. In thin films, periodic stripe-like domains occur above a critical thickness of  $t_c = 2\pi \frac{A}{K_u}$  [85], where A is the exchange constant and  $K_u$  is the perpendicular anisotropy constant [86, 87]. We remark that  $K_u$  and consequently the critical thickness  $t_c$  can be strongly affected by the deposition parameters [88]. In our case, by considering the typical value  $A = 1 \times 10^{-6} \text{erg/cm}$ ,  $t_c = 100 \div 300 nm$  is calculated. Stripe domains appear as a consequence of a slight magnetization canting with respect to the overall in-plane orientation. The small out-of-plane

components  $(\pm M_0)$  point alternatively in upward and downward directions across adjacent stripes. The width *w* of the stripes can be controlled by the Py thickness  $d_m$  following the phenomenological relation  $w = \alpha \sqrt{d_m}$  [87].



**Figure 3.** In frequency modulation-MFM mode, a resonating cantilever scans the sample surface line by line. The stray field coming out from it shifts the resonance frequency of the cantilever. The frequency increases (decreases) in an attractive (repulsive) regime.

In our experiments, the choice of Py as ferromagnetic material is thus due to the high control we can exert on *w* by changing the ferromagnetic thickness  $d_m$ . Indeed, *w* affects the value of the critical magnetization  $M_{cs}$ . For the same reason, several values of the superconducting thickness  $d_s$  have been considered in order to tune  $M_{cs'}$  by keeping the same superconducting

material. Indeed, once the S layer is set, the superconducting critical field  $H_{c1} = \frac{\Phi_0}{4\pi\lambda^2} \ln\left(\frac{\lambda}{\xi}\right)$ ,

which depends on the London penetration depth  $\lambda$  and on the coherence length  $\xi$ , is automatically fixed.

The magnetic properties of the Nb/Py hybrids were analyzed by means of a vibrating sample magnetometer insert of a Quantum Design PPMS and a cryogenic ultrahigh vacuum scanning force microscope equipped with a magnetic tip and operating in frequency modulation-magnetic force microscopy (FM-MFM) mode. **Figure 3** illustrates the working principle of the FM-MFM technique: the frequency shift  $df = f - f_0$  of the resonating cantilever, due to the stray field coming out from the sample, is acquired line by line (*f* is the oscillation frequency measured during tip-sample interaction and  $f_0$  is the free frequency). MFM maps were obtained by scanning at constant tip-sample heights and the attractive/repulsive tip-sample interaction is mapped using color contrast. For each sample, we scanned several different areas of the surface to get good statistics. During the measurements the temperature stability was within 0.01 K. We used a commercial Si cantilever, covered with ferromagnetic Co/Cr film,

with resonance frequency  $f_0 \approx 75 kHz$ , elastic constant  $k \approx 2.8 N/m_{\nu}$  and nominal low magnetic moment  $\mu \approx 0.3 \times 10^{-13} emu$ .

The tip coercivity  $H_{c,tip} = 550 \div 6000e$  was deduced from the inversion of the magnetic contrast by applying an external magnetic field. Before measuring, the tips were magnetized in downward direction along their longitudinal axis. As a consequence, an attractive (repulsive) force appears as a dark (bright) contrast region in the MFM maps. In the presence of vortices, V (AV) appears as darker (brighter) spots on the magnetic background. We remark that the tip-sample distance plays a key role during the MFM experiments. When crossing  $T_{s'}$  the cantilever was pulled away from the sample surface (about 10 µm), to minimize the influence of the tip's stray field on V-AV nucleation and spatial configuration.

Typical magnetic hysteresis loop of Py-1  $\mu$ m and Py-1  $\mu$ m, in perpendicular (**Figure 4a** and **b**) and parallel (**Figure 4c** and **d**) configurations with respect to the film plane are reported in **Figure 4**.

By measuring the saturation fields  $H_{s\parallel} = 130 \div 1600e$  and  $H_{s\perp} = 11 \div 12k0e$ , the saturation magnetization  $M_s$  as well as the uniaxial anisotropy constant  $K_u$  can be estimated, resulting in  $M_s = \frac{H_s \perp + H_s \parallel}{4\pi} \approx 900G$  and  $K_u = \frac{H_s \parallel^M s}{2} \approx 6.3 \times 10^{4erg} / cm^3$  [89]. The comparison between parallel and perpendicular saturation field values confirms the presence of an easy axis mainly oriented in the film's plane, whereas the ratio  $\frac{K_u}{K_d} < 1$  (here,  $K_d = 2\pi M_s^2$  is the stray field energy density) indicates a weak perpendicular anisotropy.



**Figure 4.** (a) Magnetic hysteresis loop of 1  $\mu$ m-Py in perpendicular applied magnetic field. Top left corner: MFM map of Nb(100 nm)/Py(1  $\mu$ m) at *T* = 12 K and tip-Py separation of *h* = 140 nm. Bottom right corner: FFT of the MFM map. (b) Magnetic hysteresis loop of 2  $\mu$ m-Py in perpendicular applied magnetic field. Top corner: MFM map of Nb(200 nm)/Py(2  $\mu$ m) at *T* = 12 K and *h* = 180 nm. Bottom right corner: FFT of the MFM map. (c) and (d) Magnetic hysteresis loop of 1 and 2  $\mu$ m-Py in parallel applied magnetic field.

The MFM maps shown in the insets of **Figure 4(a)** and **(b)**, respectively, on Nb/Py(1 µm) and Nb/Py(2 µm) samples, were taken at *T* = 12 K with relative separation of the tip from the Py surface of 140 and 380 nm, respectively. Before the MFM experiments, the magnetic stripes were oriented along a preferred direction by applying an in-plane external magnetic field greater than  $H_{s\parallel}$ . Frequency spans of the MFM images of 1.1 Hz in Nb/Py(1 µm) and 1.7 Hz in Nb/Py(2 µm), even though the tip-Py(2 µm) separation is higher, indicates that the magnetic signal coming out from 2 µm-Py sample surface is definitely stronger than 2 µm-Py. In addition to this, in the 2 µm ferromagnetic layer, not only the stripe conformation is much more straight and regular, but also the magnetic roughness along the single stripe is significantly lower, as measured by a frequency shift of around 0.16 Hz in 2 µm-Py and 0.4 Hz in 1 µm-Py layer. From the fast Fourier transform (FFT) analysis of the MFM maps, acquired in different areas of the sample surface, an average stripe width of 490 nm ± 2% and 790 nm ± 4%, for the 1 µm-Py and 2 µm-Py layers, respectively, can be inferred, confirming theoretical expectations  $w = \alpha \sqrt{d_m}$  [87].

In order to quantitatively compare the MFM results with the theoretical threshold conditions for vortex nucleation, the thickness  $d_s$  of Nb films and the magnetic domain width w were tuned by changing thin film deposition rate and time and measured by statistical analyses of MFM maps by FFT. Moreover, the knowledge of  $\xi(6K)$  and  $\lambda(6K)$ , derived from transport and magnetic measurements, allows the estimate of  $H_{c1}(6K)$ .

Nb (nm)	Py (µm)	$w[nm]+\Delta w$	w/λ	$d_{\rm s}/\lambda$	<i>M</i> <sub>cs</sub> (6K) (G)	<i>M</i> <sub>cl</sub> (6K) (G)	<i>M</i> <sub>c</sub> (6K) (G)
50	1.0	$490 \pm 2\%$	7	0.74	_	_	15.9
100				1.5	15.1	32.6	-
150				2.2	24.9	33.9	-
200				2.9	33.9	34.2	-
360				5.3	61.5	34.3	-
120	2.0	$790 \pm 4\%$	12	1.8	11.9	26.1	-
200				2.9	21.1	25.2	_

By increasing Py thickness, the stripe width increases following a square root dependence. Note that  $M_{cs}$  is always lower than  $M_{cl'}$  except for Nb(360nm)/Py(1  $\mu$ m).

**Table 1.** Characteristic parameters of measured Nb/Py bilayers and relative critical magnetization values.

In **Table 1**, the thickness of the superconducting films and the magnetic domain width are compared to the magnetic size of the vortex and the strength of the critical magnetizations is derived. For all the analyzed hybrids, the ratio  $\frac{w}{\lambda}$  is always greater than 1. On the other hand, we point out that the Nb(50 nm)/Py(1 µm) sample, being within the limit  $\frac{d_s}{\lambda} < 1$ , satisfies the conditions of model [52], whereas all other samples are in agreement with model [51], having  $\frac{d_s}{\lambda} > 1$ . From **Table 1**, one can note that the formation of spontaneous straight vortices is

energetically favored in most cases, since it results  $M_{cs} < M_{cl}$ . Only in the case of Nb(360 nm)/ Py(1 µm) the semiloop vortices are expected. At the MFM imaging temperature, vortices will stay in the superconducting layer if the intensity of the out-of-plane components  $M_0$  of Py magnetization is enough to sustain them (the temperature-dependent threshold conditions are satisfied). If under threshold, they can still be confined in the superconductor when the energy of the single vortex  $U_{sv}$  is higher than the energy of the Bean-Livingston barrier  $U_{BL'}$  so that the escape condition is not satisfied.



**Figure 5.** Temperature-dependence of the ratio  $\frac{U_{SV}}{U_{BL}}$  in the studied Nb/Py heterostructures.

In **Figure 5**, we report the plot of the ratio  $\frac{U_{sv}}{U_{BL}}$  as a function of the temperature for all of our samples, showing that spontaneous V-AV formation is only regulated by  $M_0$  value, thus ruling out Bean-Livingston confinement. By comparing the calculated values of the critical magnetizations of 15.9 and 15.1 G for Nb(50 nm)/Py(1 µm) and Nb(100 nm)/Py(1 µm), respectively, with the measured value  $M_0 = 16G$  [47, 90, 91], we expect a spontaneous formation of V-AV in both samples, even though the threshold values are very close to the measured  $M_0$ .

#### 4. Superconducting vortex nucleation

In **Figure 6(a)** and **(b)**, the MFM images of Nb(100 nm)/Py(1 um) and Nb(50 nm)/Py(1 um) below the superconducting critical temperature are shown. As expected, Nb(100 nm)/Py(1  $\mu$ m) (**Figure 6a**) forms spontaneous vortices and antivortices in the center of the oppositely polarized stripes, with a vortex polarity collinear with the magnetization of the underlying stripe domain [50]. In a scan area of 3.8  $\mu$ m × 3.8  $\mu$ m, we observe a small vortex density, with

unequal number of vortices and antivortices, with "up" polarity vortices dominating. To gain further insight into the imbalanced vortex–antivortex phenomenon, field cooling (FC) measurements in both positive and negative magnetic fields were performed. In general, a change in the relative density of vortices and antivortices is always expected after a field cooling process. Indeed, the effect of the external magnetic field on the magnetization vectors is to enhance the collinear magnetization components and compensate (partially or totally) the anticollinear ones. When the external magnetic field totally compensates the stray field of the ferromagnet, vortices will not nucleate on the top of them, as it happens in **Figure 6(c)**. If the stripe stray field is only partially compensated, vortices might be still induced. However, their density will result lower with respect to the vortex family nucleated on stripes collinear to the external field.



**Figure 6.** MFM maps in zero field cooling of (a) Nb(100 nm)/Py(1  $\mu$ m), *T* = 6 K, *h* = 130 nm; (b) Nb(50 nm)/Py(1  $\mu$ m), *T* = 6 K, *h* = 200 nm. MFM of Nb(100 nm)/Py(1  $\mu$ m) at *T* = 6 K, and *h* = 180 nm field cooled in (c) H = +6 Oe, (d) H = -27 Oe.

**Figure 6(c)** shows a MFM image acquired after a FC in H = +6 Oe. Antivortices appear above the proper stripes whereas no vortices are present above oppositely polarized magnetic stripes. On the other hand, the map acquired after a FC in higher negative field H = -27 Oe (**Figure 6d**) still shows the presence of both V and AV, even though the density of vortices with the same polarity as the external applied field becomes higher. The absence of vortices after a field cooling in H = 6 G (**Figure 6c**) and the presence of antivortices after a field cooling in H = -27G (**Figure 6d**) suggest the local unbalancing of Py out-of-plane magnetization components. Indeed, while a positive field of 6 G is enough to compensate the "negative stripe," preventing vortex formation, a negative field of 27 G has only a partial effect, still letting some antivortices to pierce the Nb film. This result points out a local residual out-ofplane magnetization of -16.5 G. No clear evidence of spontaneous V-AV formation was shown by Nb(50 nm)/Py(1  $\mu$ m) but instabilities or jumps in the MFM image (marked with arrows in **Figure 6b**), and contrast modulation along the stripes were measured. In reference [50], it was shown that these jumps, which always appear in the direction of the fast-scan axis, are due to the interaction of the vortex with the magnetic tip itself. Jumps due to the vortex motion are also visible in **Figure 6(c)** and **(d)** and their geometrical confinement inside the stripes is proof of the role of the Py out-of-plane component as a strong magnetic pinning source acting against the possibility for the vortices to move perpendicularly to the stripe domains, by crossing the domain wall barrier. The behavior below the superconducting critical temperature of the thickest superconductor samples, Nb(360 nm)/Py(1  $\mu$ m), Nb(200 nm)/Py(1  $\mu$ m), and Nb(150 nm)/Py(1  $\mu$ m) is presented in **Figure 7** [50].



**Figure 7.** MFM maps in zero field cooling of (a) Nb(360 nm)/Py(1  $\mu$ m), T = 6 K, h = 110 nm; (b) Nb(200 nm)/Py(1  $\mu$ m), T = 6 K, h = 60 nm; (c) Nb(150 nm)/Py(1  $\mu$ m), T = 6 K, h = 110 nm. MFM maps of (d) Nb(360 nm)/Py(1  $\mu$ m), (e) Nb(200 nm)/Py(1  $\mu$ m), and (f) Nb(150 nm)/Py(1  $\mu$ m) field cooled respectively in H = -16 Oe, H = -11.5 Oe, and H = +10 Oe.

The Nb diamagnetism causes the attenuation of the stripe contrast as the thickness of the superconducting layer grows. Keeping the tip-sample separation fixed at h = 110 nm above the Nb surface, a low magnetic contrast is observed in the thickest sample (Nb 360 nm, **Figure 7a**), whereas the magnetic stripes appear visible whenever the Nb thickness is at or below 150 nm (**Figure 7c**). Clearly, a more efficient screening of the Py out-of-plane magnetization component occurs in the thickest superconducting layer. In Nb(200 nm)/Py, **Figure 7(b**), we reduced the tip-sample separation to h = 60 nm in order to gain sensitivity. All attempts to unveil spontaneous V-AV in Nb(360–200–150 nm)/Py(1 µm) failed, thus confirming the agreement between the theoretical model [51] and the measured value of local magnetization of 16 G, estimated from the transport measurements. Since the stray field from 1 um-Py film was not sufficient by itself to induce vortices, these Nb/Py samples were cooled down in an

out-of-plane external applied magnetic field. In **Figure 7(d)–(f)**, three MFM maps acquired at T = 6 K on Nb(360 nm)/Py(1 µm), Nb(200 nm)/Py(1 µm), and Nb(150 nm)/Py(1 µm), respectively, field cooled in H = -16Oe, H= -11.5 Oe, and H = +10O e, are reported. As expected, only the vortices parallel to the external field direction are created. In **Figure 7(f)**, the intensity of the field was tuned in order to get a vortex-vortex distance matching the formation of a triangular (or hexagonal) vortex lattice, matching the stripe confinement.

Finally, the formation of spontaneous V-AV pairs due to thicker Py layer was demonstrated in Nb(200 nm)/Py(2 µm) (**Figure 8a**)and Nb(120 nm)/Py(2 µm) (**Figure 8b**). The experimental evidence of spontaneous V-AV nucleation and its comparison with the model [51], to which these samples within the limit  $\frac{d_s}{\lambda} > 1$  refer, allow us to infer the lower limit of the 2 µm-Py out-of-plane component value resulting in  $|M_{0-Py2\mu m}| > 21.1G$ .



**Figure 8.** MFM maps in zero field cooling of (a) Nb(200 nm)/Py(2 μm), *T* = 6 K, *h* = 180 nm; (b) Nb(120 nm)/Py(2 μm), *T* = 6 K, *h* = 180 nm; (c) MFM map of Nb(200 nm)/Py(2 μm) field cooled in H = +60 Oe, *h* = 150 nm.

In these samples we observe a high and almost uniform vortex density along the stripes as well as the tendency for spontaneous vortices and antivortices to be paired with each other. We correlate these experimental results to the stronger magnetic template, together with wider magnetic stripe domains, and the thickest superconducting layer. As compared to the 1  $\mu$ m-Py layer samples, the stripe conformation in 2  $\mu$ m-Py samples is more straight and regular, the magnetic signal coming out from the surface is stronger and the magnetic roughness along the single stripe is smaller, thus highlighting a much more uniform canting of the ferromagnet's magnetization. The frequency signal of the vortex compared to the stripe's magnetic background is 0.97 mHz in Nb(200 nm)/Py(2  $\mu$ m), 0.3 mHz in Nb(120 nm)/Py(2  $\mu$ m), and 0.4 mHz in Nb(100 nm)/Py(1  $\mu$ m), indicating that, as expected, superconducting leaks occur in the thinnest samples.

We speculate that the decoupling of V-AV pairs in Nb(100 nm)/Py(1  $\mu$ m) may be affected by the tendency of the magnetic field lines coming out from a vortex to close inside the leak, instead of the paired antivortex. As well as by the presence of smaller magnetic stripe domains so that any inhomogeneity in the stripe width induces very inhomogeneous vortex density. In **Figure 8(c)**, the low temperature MFM map of Nb(200 nm)/Py(2  $\mu$ m) after a field cooling in H = -60 Oe is shown. The strength of the field is not enough to completely compensate the + $M_0$ 

magnetic domain and, as a consequence, both the families of vortices and antivortices are still in the sample, albeit in different numbers. We notice a very high vortex mobility inferred from the frequent "vortex jumps" facilitated by the scanning magnetic tip.

#### 5. Superconducting vortex dynamics

In Figure 9, the comparison between vortex motion in Nb(360 nm)/Py(1 µm) and Nb(200 nm)/ Py(2 µm) under the sweeping of the magnetic field is reported. Figure 9(a)-(d) shows the behavior of vortices in Nb(360 nm)/Py(1  $\mu$ m), after a field cooling in H = -21 Oe and by sweeping the field up to positive values. After an initial phase, where the vortex configuration appeared rigid, we noticed that few vortices start moving and, at H = +80 Oe (Figure 9a), a nonuniform spatial distribution of the vortex density takes place. As a consequence of jamming events or influenced by the intrinsic pinning, anomalous accumulations of vortices can occur. We speculate that the presence of few higher-energy pinning centers acts as an obstacle against the possibility for other vortices to move along a stripe. By further increasing the external field pressure, a switching event happened at H = +122 Oe, that was captured in Figure 9(b), and an antivortex avalanche enters during the external magnetic field sweep. The regular vortex pattern present in the lower half of Figure 9(b) that was recorded before the avalanche is suddenly destroyed and a disordered flux distribution sets up in the upper half of the Figure 9(b). From this point, we kept the field constant and we imaged the vortex arrangement that appeared not to match the Py stripe pattern (Figure 9c). We found that the antivortex disorder (with respect to the underlying magnetic background) remains present while the external field is reduced to zero (Figure 9d). To check if the disordered vortex pattern was not due to any modification of the Py stripes, the sample was consequently warmed up above the superconducting critical temperature of the Nb, and the stripe domains proved to remain unchanged from the original configuration.



**Figure 9.** (a–d) MFM maps of Nb(360 nm)/Py(1  $\mu$ m) at *T* = 6 K, *h* = 110 nm. After a field cooling in H = –21G, the field has been swept up +80 Oe (a), from +80 Oe to +122 G (b), kept constant at +122 Oe (c), brought down to 0 G (d). (e)–(h) MFM maps of Nb(200 nm)/Py(2  $\mu$ m) at *T* = 6 K, *h* = 250 nm. The field has been applied below *T*<sub>s</sub> and swept from 0 to –600 G.

The scenario of the vortex dynamic is completely different in Nb(200 nm)/Py(2  $\mu$ m), where spontaneous vortices appear below the superconducting critical temperature. In this case, there is no need to cool down the sample in a negative (positive) magnetic field and then sweep it to the opposite polarity, since both vortices and antivortices are already in the sample. The extremely high mobility of the spontaneous vortices was imaged by keeping the fast-scan axis as parallel as possible to the stripes in order to follow the vortex motion. **Figure 9(e)–(h)** shows the MFM maps acquired while the field is sweeping respectively from –60 to –94 Oe, from –159 to –191 Oe, from –289 to –323 Oe, and from –483 to –516 Oe and, due to the continuous motion of the vortices under the tip apex, it was not possible to get a clear image of a single vortex. By sweeping the magnetic field down to -600 G, no occurrences of avalanches were recorded.

#### 6. Magnetization measurements

Temperature-dependent low-field magnetization M(T) curves have been acquired on Nb/Py samples in zero-field-cooling (ZFC) and field-cooling (FC) processes as follows. The samples were first cooled down to 5 K in zero magnetic field, then a small field was applied and the ZFC curve was obtained by measuring the magnetization as a function of the temperature during the warming of the samples up to 10 K.



**Figure 10.** (a) ZFC magnetization measured as function of *T* during the warming-up of Nb(200 nm)/Py(2  $\mu$ m) e in 20 Oe, perpendicular to the film plane. In the inset: ZFC and FC magnetization curves. (b) Field dependence of the difference  $\Delta M$  between the upper demagnetization branch and the lower magnetization branch of the loops in Nb(200 nm)/Py(2  $\mu$ m) and Nb(120 nm)/Py(2  $\mu$ m) normalized to the Nb layer thickness  $d_s$ . In the inset: hysteresis loops for both samples, at *T* = 6 K without the contribution of the Py film, in the perpendicular field configuration.

After that, the FC curve was measured while cooling the sample down to 5 K, in the presence of the applied magnetic field. In the inset of **Figure 10(a)**, we report both the ZFC and FC curves measured in applied magnetic field of 20 Oe perpendicular to the film plane for the sample Nb(200 nm)/Py (2  $\mu$ m). The ZFC curve, in the main graph of **Figure 10(a)**, shows the characteristic behavior of a superconducting M(T), with the shielding of the magnetic field starting just below the superconducting critical temperature. From the magnetic hysteresis loops measured below  $T_s$  in the external field perpendicular to the film plane, for samples with the same Py thickness (2  $\mu$ m), but different Nb thickness (200 and 120 nm), the value of the critical current density  $j_c$  remains the same. This indicates that the vortex pinning is dominated by the underlying ferromagnetic layer rather than by an intrinsic pinning in Nb films.

The magnetic response of the Nb layer at T = 6 K, shown in the inset of **Figure 10(b)**, was determined by subtracting from the M(H) measured at  $T < T_s$  the same curve measured at  $T > T_s$ . From the hysteresis loops of the SC layers, one can evaluate the critical current density by calculating the ratio  $\frac{\Delta M}{d_{s'}}$  where  $\Delta M$  is the difference between the upper demagnetization and the lower magnetization branches of the loops and  $d_s$  is the Nb layer thickness. As shown in **Figure 10(b)**, the  $\frac{\Delta M}{d_s}$  curves are perfectly overlapping.

#### 7. Conclusions

In this work, we studied vortex-antivortex formation in magnetically coupled Nb/Py bilayers, by varying both the superconducting and ferromagnetic thicknesses. By studying the magnetostatic interaction between S and F layers satisfying the constraint  $\frac{w}{\lambda} > 1$ , the threshold condition  $M_0 > M_{c(s,l)}$  to form spontaneous V-AV (straight or semiloops) was experimentally shown and compared to theoretical predictions. In cases for both superconducting layer thickness greater or smaller than the penetration depth,  $\frac{d_s}{\lambda} > 1$  or  $\frac{d_s}{\lambda} < 1$ , the temperature-dependent expressions of the critical magnetizations  $M_{c(s,l)}(T)$  were found and experimentally analyzed. By analyzing the temperature behavior of  $M_{c(s,l)}(T)$ , one can deduce that vortices are always formed right below the superconducting critical temperature  $T_{s'}$  where  $M_{c(s,l)}(T_s) = 0$ . As the temperature decreases,  $M_{c(s,l)}(T)$  increases and the threshold condition can result to be no longer satisfied, allowing the exit of the superconducting vortices from the S layer whenever the escape condition  $d_s < < (\pi + 2)\lambda$  is respected. We proved that our samples always satisfy the escape condition, addressing the occurrences of spontaneous V-AV formation to a  $M_0$  value higher than  $M_{c(s, l)}(T)$ . By referring to [52] for Nb(50 nm)/Py(1  $\mu$ m) and to [51] for Nb(100 nm)/Py(1 µm), we should expect spontaneous V-AVs if  $M_{0-Pv(1\mu m)} > 15.9G$  and  $M_{0-Pv(1\mu m)} > 15.1G$ , respectively. From transport measurements [47, 90, 91],  $M_{0-Py(1\mu m)} \approx 16G$  was estimated and from the imaging of spontaneous V-AVs in Nb(100 nm)/Py(1  $\mu$ m),  $M_{0-Py(1\mu m)} > 15.1G$  was confirmed by MFM. On the other hand, the vortex nucleation in Nb(50 nm)/Py(1 µm) still leaves some open questions. A strong indication of the vortex nucleation in this sample is the presence of jumps appearing in the MFM map only below  $T_s$ . These jumps are the signatures of the interaction between the magnetic tip and the superconducting vortex. It is not surprising to find clearer evidence of spontaneous V-AVs in Nb(100 nm)/Py(1 µm) rather than in Nb(50 nm)/Py(1 µm). In fact, when the penetration depth  $\lambda(T)$  starts being greater than the superconducting thickness, it has to be corrected into  $\lambda_{eff}(T) = \lambda(T) \coth\left(\frac{a_s}{\lambda(T)}\right)$  [81]. As a consequence, as  $\lambda_{eff}(T) > \lambda(T)$ , the vortices swell and a greater  $M_0$  value is required to accommodate them on the stripes. From a theoretical point of view, this results in using model [52] instead of [51] that fails to satisfy the validity condition  $\frac{a_s}{\lambda} > 1$ . In **Figure 11**, we report the behavior of  $M_c(d_s)$  for the two models, together with the dependence of  $\lambda_{eff}$  on the superconducting thickness. In the framework of model [52], any further reduction in the superconducting thickness, due for example to the presence of few oxide layers, will favor V-AV formation, provided that the condition  $\frac{w}{\lambda} < 1$  is satisfied. In the case of Py(1  $\mu$ m), where  $w \approx 490$  nm, the thinnest superconducting layer satisfying the model results to be ideally 10 nm thick.



**Figure 11.** The behavior of  $M_c(d_s)$ ,  $M_{cs}(d_s)$ , and  $\lambda_{eff}(d_s)$  at T = 6 K is reported. The intersection points between black dashed line-  $M_c$  and black-dashed line-  $M_{cs}$  draw, respectively, the lower limit of the model [51] and the upper limit of [52]. The intersection point between the red dashed lines draws the lower limit of the model [52], resulting in  $d_s \approx 10nm$  in the studied S/F system.

In summary, we were able to estimate the value of ferromagnet's spontaneous out-of-plane magnetization  $M_0$  based on our MFM results. We have shown experimentally the robustness of the two theoretical models describing spontaneous vortex formation in the S/F bilayer. The field cooled experiments demonstrate that either vortices or antivortices, depending on the sign of the external field, can be formed in the samples that lack sufficient magnetization to form spontaneous V-AV pairs. The zero-field cooled experiments on samples fulfilling the condition for spontaneous V-AV formation show that the V-AV population density can be unbalanced. We also studied the dynamics of vortex and antivortex lattice under a changing applied magnetic field. Different behavior was observed in the case of spontaneous V-AVs compared to the case of Vs (or AVs) formed in external field cooling. After a field cooling in a negative static field, Nb(360 nm)/Py(1 µm), in the under-threshold regime, correctly shows Vs populating the proper stripes. This vortex configuration appears rigid when the field is swept from negative to positive values, up to 122 Oe, when an avalanche of antivortices penetrates the superconducting layer. In fact, once this critical field is reached, vortices are locally driven out and antivortices completely penetrate inside the sample, regardless of underlying magnetic template. We directly imaged already the vortex lattice before and after an avalanche in the Nb(200 nm)/Py(1 µm) sample [48]. In that case, even though the antivortices suddenly penetrated the Nb layer, the magnetic confinement imposed by the Py stripe domains was still visible. We attribute the antivortex dislocation occurring in Nb(360 nm)/  $Py(1 \mu m)$  to the stronger bulk pinning in the thicker Nb films: the influence of the magnetic template on the Nb surface decreases as much as the Nb thickness increases and, in a thick superconducting layer, during as abrupt a phenomenon such as the avalanche, the antivortices can assume a disordered configuration. On the other hand, if the magnetic field is swept in the presence of spontaneous V-AVs, a completely different vortex dynamic occurs. No avalanches were observed in the Nb(200 nm)/Py(2 µm) sample by ramping the external magnetic field from 0 to -600 Oe, but a continuous motion of V-AVs occurs, as revealed in the MFM data. The magnetic template guided the vortices along the magnetic channels, preventing them from crossing the stripes.

# Acknowledgements

Work at Temple University was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award DE-SC0004556. Work at Argonne National Laboratory was supported by UChicago Argonne, LLC, Operator of Argonne National Laboratory ('Argonne'). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. We also acknowledge the support of the MIUR (Italian Ministry for Higher Education and Research) under the project "Rientro dei cervelli."

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