Efficient artificial ordered vortex pinning in high-T$_C$ superconductors via masked ion irradiation

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Abstract:
Superconducting flux quanta in nanostructured YBa$_2$Cu$_3$O$_{7-\delta}$ films constitute a model to study the general problem of interacting particles in a potential-energy landscape. Using O$^+$ ion irradiation through a mask defined via electron-beam lithography, ordered nanometric regions with a depressed local critical temperature can be defined to precisely engineer the vortex energy landscape. A detailed characterization of this artificial pinning’s strength with temperature and vortex velocity is given, as well as a study of the vortex phase diagram and the associated thermodynamic phase transition. Thanks to the ability to modulate the superconducting condensate at the nanoscale, a new mechanism to reversibly switch the vortex energy landscape’s geometry using temperature as a control knob is developed. This is shown through the thermal switching of a geometrically frustrated array into a square periodic one. Finally, asymmetric vortex motion under a symmetric drive is studied using asymmetric pinning sites.

Résumé:
L’étude de réseaux de vortex dans un film d’YBa$_2$Cu$_3$O$_7$, un supraconducteur à haute température critique, représente un bon modèle pour comprendre les interactions entre deux particules dans un paysage d’énergie potentielle. En irradiant le matériau supraconducteur avec des ions oxygène, on affaiblit localement la température de transition supraconductrice du matériau. En combinant cette technologie à la lithographie électronique, on peut moduler spatialement les propriétés électroniques du supraconducteur à l’échelle nanométrique et de la sorte contrôler le paysage d’énergie vu par les vortex. Dans cette thèse, nous réalisons une étude sur l’évolution de ce paysage en énergie en fonction de la température et de la vitesse des vortex suivant la force de piégeage. En effet, on a pu observer que la géométrie du paysage d'énergie pour des vortex magnétiques variait radicalement en fonction de la température. En prenant cette dernière comme paramètre de contrôle, on est capable de modifier la configuration du réseau en passant d’un réseau géométriquement frustré à un réseau carrée. De plus, nous avons pu reconstituer le diagramme de phase des vortex et les transitions de phases thermodynamique associées. Pour finir, nous avons aussi étudié le mouvement asymétrique des vortex soumis à une force symétrique dans un réseau de potentiel asymétrique.
Preface:

The work presented here was conducted in the Unité Mixte de Physique CNRS/Thales in Palaiseau, France, under the direction of Javier Briático and Javier E. Villegas.

This manuscript was conceived to be self-contained and it is divided in the following manner:

Chapter 1 provides with a brief introduction to superconductivity, vortices and vortex pinning. In Chapter 2 a description of the experimental techniques employed is given: the processes and methods for fabrication and structural characterization of the samples are explained in 2.1 and 2.2, while 2.3 and 2.4 offer a description of the different lithography processes and the electrical transport measuring techniques. Chapter 3 introduces the artificial periodic pinning used in this work and presents a vortex velocity analysis in this system, and Chapter 4 studies the interplay between intrinsic and artificial periodic pinning. Chapter 5 demonstrates the thermal switching mechanism of the energy landscape’s geometry, and its application to the exploration of vortex ice states. And finally, Chapter 6 describes the work using asymmetric pinning defects to obtain a ratchet effect.
Acknowledgements:

I am immensely lucky to have reached this point in my life in which I am about to become a doctor. Of course, I owe this to a number of people that have been hoisting me up at each stage and without whose help I could have never done this.

I would like to start thanking the Fundación Pedro Barrié de la Maza, from Galicia, Spain. The scholarship that they awarded me is what allowed me to become a PhD student. I literally could not have done it without them.

Thanks to all my teachers, all of them, from kindergarten to university. They have all made me advance by stimulating my curiosity and, not less important, managing to keep me under control. My classmates, who made bearing those teachers a bit easier with various and sometimes very creative strategies. I have been lucky enough that some of those classmates are still my best friends, with which I have shared many hours of “detention” in school. El Manco, I want to thank him because, for all that I have helped him, he has helped me more. The same goes for Gusi, who is pretty thin but gets all the women. All three we make a trio calavera because we keep each other’s balance like a three-legged stool. I couldn’t have made it without you both. I also want to thank my friends from university: Yago, Calderón and Aurelio. We have spent uncountable hours in the library dealing with seemingly impossible exercises which through shared work and sheer stubbornness turned into child’s play the day of the exam.

I must admit that in my last year of university (in the [ ], Miami) my intention was to stay in the U.S. for my PhD. Luckily, no university wanted me! So I had to turn to my option of last resort: the Unité Mixte the Physique CNRS/Thales. I had spent a summer there in 2009 thanks to the intervention of José Edelstein, friend of Javier Briático, and, apparently, they had liked me for they accepted me as a PhD student. That or they were desperate to find students… In any case I started working with Javier Villegas, who in 2009 hadn’t even looked me in the face, and Javier Briático. And the truth is that these past three years have been sooo good. It is amazing that everybody here is nice and friendly, and very good at what they do. Here I have learnt, suffered and laughed so much. This was especially true while Javier Villegas
and I tried to get vortex papers published (it is not à la mode), although I think it is fair to say that we make a good team, judging by the results.

I cannot in any way begin to describe how much I rely on my family. I know it and they know it. It is a driving force for which I could never be thankful enough, but not for lack of trying: thank you time and again.

And then there is a special person who waits for me at home every night that I arrive late with something wonderful prepared (a delicious salmon with potatoes is her specialty), unless she is feeling tired and just wants to have popcorn for dinner… She is my voice of reason and an infinite font of cuteness. I owe her so much. Thank you for these past three plus years and let’s try remembering our anniversary at some point.

Lastly, I would like to issue a warning. This thesis was written in its majority in the R.E.R. B of Paris, so divers incidents et pannes de signalisation ont perturbé l’écriture de cette thèse.
Index:

Abstract ........................................................................................................................................... i
Preface ............................................................................................................................................... ii
Acknowledments ........................................................................................................................ iii

1. Introduction to vortices and vortex pinning ............................................................... 1
   1.1 The mixed state in type-II superconductors ............................................... 4
   1.2 Vortex dynamics .............................................................................................. 5
   1.3 Vortex phase diagram ..................................................................................... 5
   1.4 Artificial vortex pinning ............................................................................... 7

2. Experimental techniques ............................................................................................ 11
   2.1 Fabrication of samples .............................................................................. 12
   2.2 Structural characterization .......................................................................... 14
      2.2.1 X-ray diffraction ............................................................................. 15
      2.2.2 Scanning electron microscopy ................................................... 17
      2.2.3 Atomic force microscopy .......................................................... 19
      2.2.4 Electrical transport measurement of $T_C$ ...................................... 20
   2.3 Lithography and $O^+$ ion irradiation ........................................................... 21
      2.3.1 Optical lithography ........................................................................ 21
      2.3.2 Electron-beam lithography ......................................................... 23
      2.3.3 $O^+$ ion irradiation ..................................................................... 24
   2.4 Electrical characterization ........................................................................... 25

3. Efficient artificial ordered pinning in high-$T_C$ superconductors .................... 29
   3.1 Introduction .................................................................................................... 30
      3.1.1 Artificial ordered pinning in low-$T_C$ superconductors .............. 31
      3.1.1 Artificial ordered pinning in high-$T_C$ superconductors .......... 32
   3.2 Masked $O^+$ ion irradiation ...................................................................... 35
      3.2.1 Simulation of the $O^+$ ion damage ............................................... 36
      3.2.2 Simulation of the critical temperature $T_C$ .................................. 37
   3.3 Pinning properties of masked $O^+$ ion irradiated pinning arrays ............ 39
   3.4 Vortex velocity analysis in a periodic pinning array .................................. 40
      3.4.1 Introduction to velocity analysis ..................................................... 40
1. Introduction to vortices and vortex pinning

1.1 The mixed state in type-II superconductors

1.2 Vortex dynamics

1.3 Vortex phase diagram

1.4 Artificial vortex pinning
1. Introduction to vortices and vortex pinning

A superconductor is a material characterized by a sudden drop to zero in its electrical resistance when cooled below a certain temperature $T_C$, called the critical temperature. In the superconducting state the electrons in the material form a condensate, in which the electrical current is carried by pairs of electrons with opposite spin called Cooper pairs.

A superconductor could be thought of just as a perfect conductor. However, there exists a fundamental difference in the presence of an applied magnetic field. A perfect conductor will keep the magnetic field in its interior constant, regardless of any variations in the applied field. This can be easily demonstrated. For a perfect conductor, the resistivity $\rho = 0$. According to Ohm’s law: $\vec{E} = \rho \vec{J}$ $\Rightarrow$ $\vec{E} = 0$, where $\vec{E}$ is the electric field and $\vec{J}$ the electric current density. Using Maxwell’s equation for the electric field curl:

$$\nabla \wedge \vec{E} = -\frac{\partial \vec{B}}{\partial t} = 0 \Rightarrow \vec{B} = \text{constant}$$

In contrast, a superconductor in the presence of a magnetic field will act as a perfect diamagnetic material. This means that it will generate a magnetic field of equal magnitude but opposite direction to the applied one so that the resulting magnetic field in the interior of the superconductor is zero at all times. This response prevents the magnetic-field-induced Cooper pair breaking, due to the tendency of electronic spins to flip and point along the magnetic field.

The perfect diamagnetism seen in the superconductor is the result of a quantum-mechanical phenomenon called the Meissner-Ochsenfeld effect. It consists on the appearance of supercurrents that generate the opposing magnetic field. These screening currents flow in a thin layer on the surface of the superconductor of dimension $\lambda$, the penetration depth, over which the total magnetic field decays exponentially to zero. In the same way, the superconducting state does not abruptly appear at the edge of the superconductor. There is also a thin layer in which the
1. INTRODUCTION

density of Cooper pairs $n_s$ increases exponentially, over a distance called the coherence length $\xi$.

These two characteristic distances, $\lambda$ and $\xi$, determine the properties of the superconductor. Crucially, they are the parameters on which the surface energy of normal/superconductor boundaries depends. The sign of the surface energy determines whether the presence of such boundaries is favorable. On one hand, the Cooper pair density decays over a distance $\xi$, and the longer this distance the greater the loss in superconducting condensation energy. On the other hand, the magnetic field decays over a distance $\lambda$, and the shorter this distance the greater the diamagnetic energy cost of expelling the field. Therefore, the sign of the surface energy will depend on the relation between both distances. In particular, defining $\kappa = \lambda / \xi$, if $\kappa < 1 / \sqrt{2}$ the surface energy is positive and it is energetically unfavorable to create such boundaries, so the magnetic field will be expelled from the interior of the superconductor. Contrarily, if $\kappa > 1 / \sqrt{2}$ the surface energy is negative and it is favorable to create such boundaries, so the superconductor will allow the penetration of the magnetic field in certain regions. This defines the difference between Type I ($\kappa < 1 / \sqrt{2}$) and Type II ($\kappa > 1 / \sqrt{2}$) superconductors (see Fig. 1).

![Type I](image1.png) ![Type II](image2.png)

Figure 1: Schematic variation of the magnetic field $B$ and the Cooper pair density $n_s$ in a normal-superconducting phase boundary. A ratio of $\lambda / \xi$ smaller than $1 / \sqrt{2}$ corresponds to a Type I superconductor while a greater ratio corresponds to a Type II superconductor. Adapted from ref. [1]
1.1 The mixed state in type-II superconductors

Type II superconductors still present a Meissner state but, upon increase of the magnetic field above the first critical field $H_{C1}$, these show an intermediate region—called the mixed state—up to the second critical field $H_{C2}$ [see Fig. 2(a)].

![Phase diagram for type II superconductors](image)

*Figure 2: a) Phase diagram for type II superconductors. At low temperature and fields the magnetic field is completely expelled in the Meissner phase. An increase of the field above $H_{C1}$ yields the mixed state where vortices are formed. For fields above $H_{C2}$ or temperature above $T_c$ the material turns to the normal state. Adapted from ref. [1] Inset: scanning tunneling microscopy image of the Abrikosov lattice. Adapted from ref. [2] b) Schematic of a vortex with its characteristic lengths indicated. The magnetic flux tube is represented in red. The superconducting screening currents are shown in blue circulating around the flux tube.*

In the mixed state the magnetic field penetrates the superconductor in small normal regions in the form of flux tubes surrounded by superconducting currents, called vortices [see Fig. 2(b)]. As it can be seen in the sketch, the vortex core is a cylinder of radius the coherence length $\xi$, over which the Cooper pair density $n_s$ decays to zero, while the magnetic field and the surrounding supercurrents extend over the penetration depth $\lambda$. The amount of magnetic flux each vortex carries is an integral multiple of the magnetic flux quantum $\phi_0 = \hbar/2e = 2.07 \times 10^{-15} Wb$. Vortices also feel a repulsive interaction with each other that provokes their ordering in a minimum energy configuration: a triangular lattice of spacing $a_\Delta = 1.075(\phi_0/B)^{1/2}$ called the Abrikosov lattice [inset of Fig. 2(a)]. The number of vortices per unit area is proportional to the applied field, $n_V = B/\phi_0$. 


1. INTRODUCTION

1.2 Vortex dynamics

In the presence of an applied electrical current vortices will feel a Lorentz force per unit vortex length $\vec{F}_L = \vec{j} \wedge \vec{\phi}_0$. Upon vortex displacement with velocity $\vec{v}$, a perpendicular electrical field is generated according to Lenz’s law: $\vec{E} = \vec{B} \wedge \vec{v}$. This means that, through Ohm’s law $\vec{E} = \rho \vec{j}$, a resistivity appears in the system. Therefore, vortex motion causes energy dissipation and the zero-resistance state is lost even if most parts of the material are still superconducting.

As it was previously discussed, the vortex core is in the normal state. This means that when a vortex is formed superconductivity is destroyed at the core, which implies an energy cost. However, if the superconductor presents a defect in which superconductivity has already been destroyed, the vortex will be pinned to it to reduce the system’s energy. Indeed, vortices see non-superconducting regions as potential-energy wells. These exert a pinning force $\vec{F}_p$ on vortices that will remain trapped unless depinned, for instance, by a greater Lorentz force. The minimum current density required to produce vortex depinning is called the critical current $J_C$.

It is also worth noting that the thermal energy in the system can depin or help depin–vortices through thermal fluctuations of the superconducting condensate or vortex thermal vibrations. This gives rise to the concept of thermal activation energy for flux motion.

1.3 Vortex phase diagram

Because vortices are an ensemble of interacting particles, they can be regarded as a type of matter whose properties change with external parameters such as temperature and magnetic field. A schematic phase diagram of the vortex matter for temperature and applied magnetic field is shown in Fig. 3(a). For a given field above $H_{C1}$, at low temperatures, spatial correlations are infinite in various dimensions and a solid phase is formed. This solid can be a crystalline vortex lattice or a glass depending on the presence of disorder due to pinning defects. Different types of glass phases have been observed, the vortex glass and the Bose glass the most relevant, each with an
associated scaling analysis of their thermodynamic transition. In the glass phase it is hard to depin vortices as a result of the necessity to depin the entire lattice due to infinite correlations. This elastic regime can be seen in voltage-current $V(I)$ [or electrical field-current density $E(J)$] characteristics that are non-linear, slope-decreasing and show vanishing resistance at low currents [blue curves in Fig. 3(b)]. The latter is due to the fact that high currents are needed to depin vortices and, thus, measure a voltage.

![Image](image_url)

**Figure 3:** a) Vortex phase diagram indicating the melting line $T_M$, which separates the vortex fluid from the vortex glass. Adapted from ref. [1] b) $E(J)$ isotherms colored depending on the vortex phase diagram situation. Note the $T_g$ line indicates the transition into a vortex glass. Adapted from ref. [3]

An increase in temperature above the melting temperature $T_M(H)$ melts the vortex solid and yields a vortex liquid in which correlations are reduced. This thermodynamic phase transition is of first order if it is a clean system and of second order otherwise. In the latter case, it is called a glass transition with a characteristic glass transition temperature $T_g$. Above $T_g$, the $V(I)$ characteristics show at low currents a linear behavior due to a constant resistance [red curves in Fig. 3(b)]. This corresponds to the thermally activated flux flow (TAFF), in which a fraction of the vortices are thermally depinned producing a plastic motion. As the current is increased and the remaining vortices are unpinned, the curves become non-linear signaling the change in resistance.
1.4 Artificial vortex pinning

The possibility to inhibit vortex dissipation through vortex pinning opened decades ago a quest for effective vortex pinning techniques that is still active nowadays. Both intrinsic and artificially created defects have been used varying their size, spatial distribution and strength. In general, pinning defects are categorized into two types: point and extended defects. One of the most efficient among the latter—especially concerning high-critical-temperature superconductors—are correlated defects: extended anisotropic structures distributed in the superconductor with equal orientation. This type of structures can be intrinsic defects, like twin boundaries, or can be introduced artificially using techniques such as heavy-ion irradiation, to create randomly distributed columnar defects. Each of these structures can usually accommodate one vortex line when the magnetic field is applied along the defect orientation. In cuprate superconductors, they have proved extremely efficient to reduce vortex motion dissipative effects. Other approaches that yield random distributions of pinning defects include the incorporation of nano-composites, which not only provide enhanced vortex pinning, but also efficiently reduce the anisotropy characteristic of high-temperature superconductors—thereby enlarging the range of magnetic field directions along which a critical current enhancement is obtained.

A comparatively less studied scenario is the possibility of introducing in the superconductor spatially ordered distributions of extended pinning defects—instead of random ones. In addition to a possible increase of the critical currents, ordered pinning allows for insights on the vortices’ collective behavior. For instance, at certain values of the applied magnetic field, the vortex lattice (originally triangular) deforms in order to geometrically match the ordered pinning array. In this manner, the pinning of all vortices is “synchronized”, which shows in the so-called field-matching effects. These are usually seen as sudden drops of the magneto-resistance (or as critical depinning current peaks) for values of the applied magnetic field in which the vortex density equals that of pinning defects. These effects can be used not only to control the distribution of vortices across the sample, but also as a tool to gain information about the superconducting system: the interplay between different
sources of pinning, the dimensionality of the vortex matter determined by correlations between vortices, the effects of thermal fluctuations, etc. In the present thesis ordered pinning will be used to investigate some of these problems.
1. INTRODUCTION

References:


2. Experimental techniques

2.1 Fabrication of samples

2.2 Structural characterization

2.2.1 X-ray diffraction

2.2.2 Scanning electron microscopy

2.2.3 Atomic force microscopy

2.2.4 Electrical transport measurement of $T_C$

2.3 Lithography and O$^+$ ion irradiation

2.3.1 Optical lithography

2.3.2 Electron-beam lithography

2.3.3 O$^+$ ion irradiation

2.4 Electrical characterization
2.1 Fabrication of samples

The samples used throughout this thesis consist of 50 nm-thick thin films of YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) grown on top of SrTiO$_3$ (STO) (001) substrates.

This high-$T_c$ cuprate superconductor has an orthorhombic layered structure. In each unit cell there are two CuO$_2$ layers separated by an yttrium atom in the center, one BaO layer after each CuO$_2$ layer, and finally a last layer with two CuO chains capping the unit cell on each side [see Fig. 1(a)]. The oxygen content, determined by $\delta$, is a crucial quantity that determines the structural and electrical properties of YBCO. For instance, as it can be seen in Fig. 1(b), the temperature of the superconducting transition varies enormously with the oxygen content. For $\delta \in (0,0.65]$ the material is superconductor whereas for $\delta > 0.65$ it becomes insulator. This is the reason why it is very important to precisely control and optimize the parameters of the growth process.

The technique used for the thin film growth is Pulsed Laser Deposition (PLD). The principle behind the PLD procedure is the creation, due to the impact of a laser on a ceramic target, of a photoionized plasma that undergoes an adiabatic expansion in

![Figure 1: a) Orthorhombic crystalline structure of YBa$_2$Cu$_3$O$_{7-\delta}$. b) Critical temperature as a function of oxygen content $\delta$. Above $\delta=0.65$ YBCO is no longer superconducting. Adapted from ref. [1]](image)
which it travels to be deposited on a heated substrate [see Fig. 2(a)] forming an ordered material.

The equipment employed consists of two ultra-high-vacuum chambers – the introduction and the deposition chambers – connected by a guillotine valve [see Fig. 2(b)]. Each chamber has its respective turbo-molecular pump that brings the pressure down to around $10^{-6}$ mbar. The introduction chamber has a four sample support and a transfer arm to perform multiple growths without the need to break the vacuum. The deposition chamber has a rotatable four-target support and a mobile sample support with a heating system. This chamber is coupled to a KrF laser with a wavelength of 248 nm. Laser pulses last 25 ns and are emitted with a 5 Hz frequency. A set of mirrors, lenses and collimators guide, shape and focalize the ray to obtain a homogeneous energy density distribution. In a periodical manner, the laser gas has to be renewed to keep the energy output and the beam profile constant. The power of the laser is measured using an external calorimeter before the deposition, and amounts to around 720 mW. This represents an energy density on the YBCO ceramic target of about 33000 J/m$^2$. The target sits in the deposition chamber on a 0.36 mbar oxygen atmosphere and the sample holder is heated to around 680ºC, 4 cm away from the target. The temperature of the substrate is measured using a pyrometer placed on one of the chamber's windows. During the deposition the target is moved with a programmed motor following a rectangular path to avoid over-ablation of one point, and the sample holder is rotated to ensure a homogeneous deposition. To obtain the desired 50 nm thickness the laser ablation lasts about 70 seconds, which implies a 0.7 nm/s growth rate.
The main control parameters for thin film growth are the substrate’s temperature, the target-substrate distance, the oxygen pressure and the laser’s energy. All parameters control various aspects as the deposition rate, the atom diffusion on the substrate, the roughness of the resulting film, etc. The optimization of these parameters requires the production of multiple test films and has to be repeated fairly often. In this case, the goal of this optimization was to obtain a YBCO film with both a good critical temperature and a low concentration of outgrowths and precipitates, which may produce strong intrinsic pinning.

The growth of samples was done with Rozenn Bernard in the Unité Mixte de Physique CNRS/Thales in Palaiseau, Paris.

2.2 Structural characterization of samples

As it was previously discussed, it is important to optimize the parameters in the deposition process to obtain the desired properties in the superconducting film. It is therefore necessary to characterize each film to be able to compare different growth conditions. The structural characterization of the samples was done using four different techniques: X-ray diffraction, scanning electron microscopy, atomic force
2. EXPERIMENTAL TECHNIQUES

microscopy and electrical transport measurements to determine the critical temperature.

2.2.1 X-ray diffraction

This characterization technique allows obtaining information about the films such as crystalline structure, roughness, thickness, etc., in a non-destructive manner.

The equipment used for diffractometry measurements was a Panalytical Empyrean, which uses an X-ray Cu source that emits radiation with two wavelengths $\lambda_1=1.54056$ Å and $\lambda_2=1.5439$ Å. A Ge (002) monochromator filters out $\lambda_2$ if necessary. The sample holder, the X-ray emitter and the detector (see Fig. 3) can be moved independently, which allows testing in many different configurations.

![Figure 3: Photograph of the Panalytical Empyrean with each key element labeled.](image)

In this type of measurements, an X-ray is sent towards the sample and will be diffracted by the diffracting planes. The incoming and diffracted rays define a plane called the diffraction plane. The angle that the diffracted ray makes with the intersection of the diffraction plane and the diffracting planes is called $\theta$. The geometry employed here is the Bragg or $\theta$-2$\theta$ geometry, whose name reflects the fact that the detector is situated at $2\theta$ degrees with respect to the incoming rays [see Fig. 4(a)].
In general, two types of X-ray diffraction measurements can be done: low-angle and high-angle diffractions, depending on whether $2\theta$ is smaller or greater than $8^\circ$.

In a high-angle X-ray diffraction experiment the crystalline structure is probed. If the unit cell of the material is defined by the vectors $\vec{a}_1, \vec{a}_2, \vec{a}_3$, the diffracting plane is defined in the crystalline structure by the Miller indices (hkl) as $\frac{a_1}{h}, \frac{a_2}{k}, \frac{a_3}{l}$ [see Fig. 4(b)]. Note that the diffracting planes are not necessarily parallel to the surface of the sample. When an (hkl) family of parallel planes separated by a distance $d$ is considered, Bragg’s law gives the necessary condition to obtain a coherent interference of the rays reflected on atoms of different planes: $m\lambda = 2d \sin \theta$, where $\lambda$ is the X-ray’s wavelength and $m$ is an integer. This expression is obtained from the consideration that the difference in the distance traveled by rays reflected in other planes must be an integer multiple of $\lambda$. In Fig. 5(a) a high-angle scan of a YBCO film grown on a (001) STO substrate is shown. Here, the source and the detector are moved synchronously in a manner that always keeps the latter at $2\theta$ degrees from the incoming rays. Different peaks are obtained for each order $l$ of the (00l) family of planes, which evidences a good epitaxial growth parallel to the (001) STO substrate, and the absence of parasitic phases.
Figure 5: a) Representative high-angle X-ray diffraction curve with the different peaks labeled. b) Example of a low-angle X-ray diffraction measurement used to determine the sample’s thickness.

In low-angle diffraction experiments, it is necessary to take into account two facts: i) the planes in which the diffraction occurs are the surface of the film and the film/substrate interface, which means the crystalline structure is not probed, only the chemical contrast; and ii) the refraction that rays undergo in each plane is non-negligible. The latter produces a shift in the diffraction peak transforming the previous equation into: 

\[ \sin^2 \theta = \left( \frac{m \lambda}{2 \Lambda} \right)^2 + 2 \delta, \]

where \( \Lambda \) is the film thickness, \( m \) is also an integer, and \( \delta \) is the deviation from unity of the diffractive index \( n=1-\delta \). In a low angle scan, a diffraction peak is obtained for each \( m \) that satisfies the equation above [see Fig. 5(b)]. Taking the angular positions of the \( m \) and \( m+1 \) peaks, the sample’s thickness \( \Lambda \) is readily available deriving from the previous equation:

\[ \Lambda = \frac{\lambda}{\sin^2 \theta_{m+1} - \sin^2 \theta_m} \left( \frac{m + 1/2}{\sin^2 \theta_{m+1} - \sin^2 \theta_m} \right)^{1/2} \]

The X-ray characterization was done by Rozenn Bernard in the Unité Mixte de Physique CNRS/Thales in Palaiseau, Paris.

### 2.2.2 Scanning electron microscopy

This technique was used to characterize the surface of the thin film. The equipment used was a Hitachi S4000. A scanning electron microscope (SEM) generates a very fine beam of electrons that interacts with the sample surface. The output is then
detected to obtain information about the sample topography or chemical composition. As it is shown in Fig. 6, the electrons are generated in a cold-cathode field emission electron gun and accelerated to an energy of about 20 keV. The beam is focused using a set of electromagnetic lenses and a coil is used to move the beam scanning the desired area. When the electrons reach the sample they can be elastically or inelastically scattered by the atoms or excite the latter so they emit radiation (such as X-rays) during their relaxation. Different detectors are in place to gather each type of response. The whole system sits in a column connected to a vacuum pump. In this case the SEM was used to detect secondary electrons emitted after inelastic scatterings. These electrons originate in a shallow layer of a few nanometers in the surface of the sample and are detected by a scintillator-photomultiplier detector. The latter generates an electrical signal proportional to the number of incoming electrons. The angle the surface makes with the electron beam determines the number of inelastically scattered electrons, thus, an edge in the sample will generate a higher number of secondary electrons than a flat region. In this manner, after the image reconstruction, the edge will be seen brighter than the flat region, giving the contrast to the image.

Figure 6: Schematic diagram of a scanning electron microscope.
2. EXPERIMENTAL TECHNIQUES

The type of material being analyzed plays an important role in the preparation of the sample for the SEM. If it is a non-conducting material, a metallization will be necessary to avoid charge effects that deform the image. The machine used for this was a Balzers SCD 050 sputter coater. The sample is placed on a small chamber that is pumped to around $10^{-2}$ mbar. Then a $10^{-1}$ mbar argon atmosphere is created and, by applying a 30 mA current, a plasma is generated from a Au/Pd target and deposited on the surface of the sample during 60 seconds. This way the surface of the sample will be conducting and ready to be imaged by scanning electron microscopy.

2.2.3 Atomic force microscopy

Used to characterize the surface of the sample, the atomic force microscope (AFM) consists on a tip mounted at the end of an arm called cantilever (see Fig. 7). In this case an Enviroscope AFM was used in the tapping mode, in which the cantilever is driven to a vibration with a characteristic frequency by a piezoelectric stimulated with an AC voltage. The oscillations can be measured using a laser reflected on the end of the cantilever that is detected by an array of photodiodes. When the tip approaches the surface of the sample, forces such as van der Waals’ or electrostatic interactions modify the amplitude of the oscillations according to the equation of the driven harmonic oscillator. This modification is used to feedback the piezoelectric on the sample holder to change the height so the tip remains at a constant distance from the sample. In this manner, the topography of the sample can be recorded as the tip scans the desired area. It is worth noting that depending on the material of the tip, it is possible to make the system sensitive to different kind of forces (like magnetic forces in magnetic force microscopy) or to perform resistance measurements with a conductive tip in contact mode.
The ability to obtain the topography of the films was also used to measure their thickness. Performing a photolithography with chemical etching (see 2.3.1), the deposited material can be removed in a channel down to the substrate. After that, by measuring the height of the step in the border of the channel the thickness is obtained. This is a good way to corroborate the results obtained with the low-angle X-ray diffraction.

### 2.2.4 Electrical transport measurement of the critical temperature

A simple setup was used to quickly determine the critical temperature of a sample after its deposition in the PLD setup. It basically consists on a four-point measurement using four gold-coated pins attached to a copper head at the end of a bar, progressively submerged in a liquid nitrogen Dewar. The sample is glued to a ceramic holder with gold pads and contacted using an Al wire-bonding machine. The holder is placed on the copper head, where the four golden electrical terminals fix the ceramic holder through pressure exerted by a screw (see Fig. 8). A thermocouple situated in the head measures the sample temperature. The bar is progressively submerged in the liquid nitrogen Dewar using a motor whose speed is controlled by a temperature variation feedback. A Keithley current source and nanovoltmeter are used to measure the resistance as a function of temperature. This way the critical temperature of the film is quickly determined with a precision of about half a Kelvin.
2. EXPERIMENTAL TECHNIQUES

2.3 Lithography and O$^+$ ion irradiation

Once a film with the desired structural characteristics is achieved, it is time to define the micro-bridges that will be used for electrical transport measurements. During the fabrication process multiple lithography steps are performed using different techniques.

2.3.1 Optical lithography

In optical lithography (or photolithography) the sample is covered with a chemical resist that is photosensitive. A mask is used to expose certain areas of the resist to UV light, which will change the chemical properties of these zones, making them vulnerable or resistant to a chemical agent called developer, depending on whether the resist is positive or negative. Thus, after the exposure the sample is introduced in a developer to remove the resist in the desired areas.

In each 1x1 cm$^2$ film four micro-bridges are defined. The first step for that is the creation of the gold contact pads by optical lithography. The sample is spin coated with Megaposit SPR700-1.2, a positive photoresist in which the exposed areas are the ones the developer will attack. The spinning is done at 4000 rpm during 30 seconds, so the resist has a thickness of about 1 µm. After that the resist is softbaked one minute at 90ºC. To harden the resist it is introduced in chlorobenzene 10 minutes...
after which it is rinsed with deionized water and dried up with nitrogen gas. At this point it is introduced in the mask aligner, with the mask in Fig. 9(a) that exposes the zones where the contacts will be. Note also the four alignment crosses on each corner. The sample is illuminated with 100 mJ of UV light. Finally, it is introduced in the developer MF319 15 seconds then rinsed and dried up with N₂ gas.

This has produced a resist layer with an inward step at the borders, as seen in the zoom-in of Fig. 9(a). This is important because after the sputtering of a 50 nm layer of gold on top, the inward step facilitates the acetone lifting off the resist, taking away the unwanted gold.

After the contacts have been fabricated the micro-bridges are defined. The lithography process is the same as the previous one except for the absence of the hardening step with chlorobenzene. The mask used can be seen in Fig. 9(b): four 40 µm-wide bridges with four current contacts and eleven voltage contacts in pairs separated 200 µm. It is important to perfectly align the mask with respect to the contacts, a task eased by the highly-reflective golden crosses in the corners.

Once the bridge is defined in the resist it is necessary to remove the unprotected YBCO. This is done through ion beam etching (IBE), in which Ar ions in a plasma are accelerated towards the sample. The uncovered parts of the YBCO are damaged and etched away while the resist protects the rest. In this case the IBE machine is
coupled to a mass spectrometer so the etched material can be analyzed to determine when the substrate has been reached. Careful attention must be paid to avoid over-etching and turning the substrate conducting due to Ar ion implantation.

A different etching process was used to define the channels in the calibration films to measure the thickness (see 2.2.3): chemical etching. It consists on submerging the sample in phosphoric acid $\text{H}_3\text{PO}_4$ that etches the exposed material. This lasts 15 seconds after which the sample is introduced in water to neutralize the acid. The precision in the depth and lateral dimensions of the etching in this method is smaller than with IBE.

### 2.3.2 Electron-beam lithography

Due to the magnitude of the UV light wavelength employed in photolithography, there is a limit to the miniaturization of the motives that can be lithographed. Also, to probe the physics of a system, work should be done at the same scale as the relevant lengths of the system. In this case, a high-temperature superconducting system with vortices, the relevant length scale is of a few nanometers corresponding to the coherence length $\xi$. Thus, we use electron-beam lithography to create a mask with nanometric holes through which we will irradiate the YBCO.

As its name suggests, in this lithography process an electron beam is directed towards an 800 nm-thick PMMA resist [poly(methylmethacrylate)] that covers the sample. The tool employed for this is a scanning electron microscope from Vistec, the EBPG5000+. The current used is 1 nA and the dose 10 C/m$^2$. When the electrons arrive to the resist the solubility of the latter in a chemical developer is changed, in the same way that happened in photolithography. The lithographed region in our samples is marked in Fig. 12. To avoid charge effects during the lithographic process, the resist is metallized with a 30 nm layer of aluminum. This way charges can be evacuated efficiently and the height of the sample can be better determined. The coils in the SEM column move the beam according to the instructions of a computer, where the design of the mask has been preloaded. The last step is to remove the
exposed resist. The sample is introduced in a chemical developer composed of MIBK/IPA (Methyl isobutyleketone) and isopropanol during 45 seconds.

This lithographic process was done by Christian Ulysse in the Laboratoire de Photonique et de Nanostructures in Marcoussis, France.

![Micro-bridge with the e-beam lithographed region](image)

**Figure 12:** a) Micro-bridge with the e-beam lithographed region indicated by the blue dashed square. b) Scanning electron microscopy of the bridge after e-beam lithography.

### 2.3.3 Oxygen ion irradiation

The final procedure in the sample fabrication is the O$^+$ irradiation of the YBCO through the mask created using e-beam lithography. This process is done in the ICUBE/D-ESSP laboratory in Strasbourg.

The sample is glued to a holder, which is then fixed to a carrousel so multiple irradiations can be done in one session. The carrousel sits in a chamber that is pumped to 6·10$^{-6}$ mbar. This irradiation chamber is connected to an Eaton NV200 ion accelerator, which can function with several types of ions. In this case, either a bottle of CO gas or a balloon filled with O$_2$ gas feeds the machine. The gas is ionized into a plasma accelerating electrons towards the molecules and a mass spectrometer selects the appropriate ion. Finally, an electrical field accelerates the oxygen towards the sample at an energy of 110 keV, creating a current of 2 µA. The irradiation process takes around 3 minutes, yielding a dose of 5·10$^{13}$ ions/cm$^2$. 
2. EXPERIMENTAL TECHNIQUES

2.4 Electrical characterization:

After the irradiation of the samples using oxygen ions, each micro-bridge is separated to be measured individually. For this the sample is cut in four steps using a rotatory diamond saw. Now the nanostructured YBCO micro-bridges are ready to be characterized via electrical transport measurements.

The samples first are glued to a chip (see Fig. 13) and then wired using a gold ball-bonding machine. The measurements performed are always done using four contacts, and the design of the micro-bridge ensures that different regions, irradiated and virgin, can be measured.

![Image](image.png)

*Figure 13: Photography of the probe's copper head. The different elements are labeled.*

The chip is pushed into a holder in the copper head of the probe (Fig. 13), which is introduced in the cryostat at room temperature. There is a He gas line to put the cryostat in overpressure every time it is opened. The head is made of copper and has a sample holder with 20 contacts electrically wired to the connectors that remain outside the cryostat. The latter is a liquid-He-flow cryostat in which the liquid helium is continuously transferred from a Dewar using a low-loss syphon with a motorized needle valve to control the helium flow (see Fig. 14).
The cryostat is connected to a pump to evacuate the helium and keep the flow going. To insulate the interior of the cryostat there is a thin cavity kept in vacuum at around $5 \cdot 10^{-6}$ mbar. The temperature in the cryostat is controlled by an ITC 501 from Oxford with the ability to automatize the needle valve and the heater control. Two temperature sensors feed the ITC: a cernox sensor in the head of the probe and a RhFe sensor inside the cryostat. The heater is situated next to the sensor inside the cryostat and consists on a $40 \, \Omega$ electrical resistance. The accuracy in the temperature control is around $20 \, \text{mK}$ at the temperatures used in this work. In this setup there is also a LakeShore electromagnet connected to a KEPCO Power Supply BOP 20-50GL in current mode, that allow the application of magnetic fields up to 0.5 T. Samples are measured applying an electrical current with a Keithley 6220 current source and measuring the voltage drop with a Keithley 2182A nanovoltmeter. The head in the probe is designed for the application of the field at any angle with respect to the surface, keeping always the current perpendicular to the magnetic field, in what is called the constant Lorentz force geometry. The magnetic field angle is set using a rotor with half a degree precision and its magnitude is measured with a LakeShore 450 Gaussmeter sensitive to variations of 1 Gauss. For measurements in which an AC signal was needed, a different probe was used. It possesses a RhFe sensor in the head.
2. EXPERIMENTAL TECHNIQUES

and, crucially, a coaxial cable that descends the AC signal to inject it to the sample. This signal is generated using an Agilent Technologies PNA network analyzer E8364C, which can produce signals with frequencies ranging 10 MHz – 50 GHz.

Every instrument in this setup is controlled and programmed using a computer running Labview. Specific programs have been created for each task, like resistance vs. temperature, resistance vs. field or current vs. voltage measurements, as well as for realizing measurements with both DC and AC currents simultaneously.
References:


3. Efficient artificial ordered pinning in high-$T_C$ superconductors

3.1 Introduction

3.1.1 Artificial ordered pinning in low-$T_C$ superconductors

3.1.1 Artificial ordered pinning in high-$T_C$ superconductors

3.2 Masked O$^+$ ion irradiation

3.2.1 Simulation of the O$^+$ ion damage

3.2.2 Simulation of the critical temperature $T_C$

3.3 Pinning properties of masked O$^+$ ion irradiated pinning arrays

3.4 Vortex velocity analysis in a periodic pinning array

3.4.1 Introduction to velocity analysis

3.4.2 Experimental results

3.4.3 Discussion

3.4.4 Conclusions

In this first chapter of experimental results, an in-depth description of the technique employed to create artificial ordered pinning –masked ion irradiation– is given. After it, an analysis is presented of the vortex interactions with the ordered pinning array as a function of the vortex velocity.
3.1 Introduction

Type-II superconductors are characterized for allowing the penetration of the magnetic field in the form of flux tubes called vortices. Vortex movement causes energy dissipation and lead to the loss of the zero resistance state. Defects in the superconductor, however, can inhibit vortex dissipation by pinning them down. Defects can be natively present in the material or they can be artificially introduced through various fabrication techniques. Initially random defect distributions –as those created by heavy-ion irradiation– attracted much attention, due to their capacity to greatly reduce vortex dissipation in technological applications. These included the increase of critical currents in superconducting wires and strips, for instance with the addition of nano-composites [1]. For a review see [2].

Due to the periodic nature of the vortex lattice, the use of artificial ordered defects for vortex pinning became an interesting possibility. Pinning arrays of different geometries were soon fabricated. It was found that, aside from reducing vortex dissipation, ordered pinning arrays deformed the (originally triangular) vortex lattice so it would geometrically match the pinning array. These field-matching effects were seen as sharp drops in resistance versus field curves, due to the synchronized pinning of all vortices at vortex densities equal to that of pinning sites. It was seen too that it was possible to channel vortex movement using artificial ordered defects [3,4]. Soon, the ability to engineer the energy landscape that vortices see through artificial ordered pinning opened the door to a number of interesting technological and fundamental possibilities. Vortices constitute a model for a manifold of interacting particles. Therefore, the design of the pinning potential allows the study of fundamental problems (commensurability, rectification, jamming, avalanches, etc.) that are common to a host of physical systems (colloids, atoms in optical traps, swimming bacteria, proteins in motion, electrons in Wigner crystals, skyrmion lattices, etc.). On the technological side, magnetic field effects on Josephson based devices, such as the 1/f noise caused by vortices in a superconducting quantum interference device (SQUID), could be reduced using periodic hole arrays [5]. Furthermore, the term “fluxtronics” was coined to encompass various concepts on electronic devices controlling vortex motion. For instance, superconducting diodes can be obtained
creating asymmetric defects, so it is easier for vortices to escape the pinning site in one direction than the other. This produces a ratchet effect in which a dc signal is measured when driving vortices with an ac current [6]. Another example is the signal processor fabricated with arrays of holes [7], for which the output signal depends on the phase difference between two harmonics of the driving current.

3.1.1 Artificial ordered pinning in low-$T_C$ superconductors

A number of nano-fabrication techniques allowed decades ago easily patterning low-$T_C$ materials at the required length scale to obtain effective artificial ordered defects. Initially, the thickness modulation of the superconductor [8] exploited the idea that vortex energy is proportional to its length, so vortices are pinned in thinner regions. With the goal to create individual pinning sites arranged in an ordered manner, various lithographic techniques were used to create periodic hole arrays at the sub-micron scale [9]. Also, arrays of non-superconducting magnetic inclusions [10] were used, taking advantage of the magnetic and corrugation effects to produce pinning. These techniques progressively yielded stronger and richer field-matching effects. See Fig. 1 for an example of the behavior observed with magnetic-dot rectangular arrays in Nb films by Martín et al. [11] In this system, for low applied fields, we see three clearly-defined matching minima that imply a rectangular vortex lattice. For higher applied fields, a vortex lattice reconfiguration from rectangular to square occurs, clearly marked by the change in the period and intensity of the matching effects. Later on, the focus in the artificial ordered pinning community expanded from increasing its strength to make it functional. For instance, using asymmetric pinning traps a ratchet effect which displayed multiple controlled reversals was obtained [12]. As another example, a switchable pinning array was achieved [13] via the control of magnetizations of a Co dot array: through the magnetic history, magnetizations were made to point in the same direction (turning the collective pinning on) or incoherently (turning it off). Alongside the development of artificial ordered pinning, a big body of research was obtained from investigations on how ordered pinning modifies vortex
dynamics, the vortex phase diagram and vortex phase transitions, as well as from studies on general problems common to any ensemble of repulsive particles.

![Figure 1: Magneto-resistance curve obtained in Nb with a rectangular array of magnetic dots. Each resistance minimum corresponds to a commensurate state of the vortex lattice on the pinning array. The transition from sharp to shallow minima indicates a vortex lattice reconfiguration from rectangular to square. Adapted from ref. [11]](image)

### 3.1.2 Artificial ordered pinning in high-$T_C$ superconductors

The idea of realizing studies similar to those previously done on low-$T_C$ superconductors using high-$T_C$ ones became very attractive for various reasons: aside from a higher $T_C$ they present a characteristic anisotropy, due to their layered structure, and strong thermal fluctuations. All these features provide a richer playground in which to investigate fundamental problems. Furthermore, high-$T_C$ superconductors are more attractive from the technological point of view, since they can operate at higher temperatures with its consequent energy and space savings in the cooling system.

However, the realization of efficient ordered pinning has proven more challenging in these materials, due to the combination of both technical difficulties and physical constraints. High-$T_C$ superconductors are delicate materials since their properties strongly depend on oxygen content, and they are usually damaged upon contact with water. This makes the nano-lithography techniques traditionally used for low-$T_C$
3. ARTIFICIAL ORDERED PINNING IN HIGH-\(T_C\) SUPERCONDUCTORS

materials more difficult to implement. Moreover, the much shorter relevant length scales (i.e. the coherence length for vortex-core pinning is of few nanometers), the higher thermal energy and the competition with strong intrinsic random pinning make it harder to obtain systems in which the artificial ordered pinning is the dominant mechanism.

Initially, the most successful results were obtained for a material with very weak intrinsic random pinning: \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8+\delta\). In 2005, Ooi \textit{et al.} [14] reported on the use of focused ion beam milling in BSCCO single crystals to create a submicrometric triangular array of through-holes [see inset Fig. 2(a)]. The array parameters were many times greater than the relevant physical distances in this system: the superconducting coherence length \(\xi\) (few nanometers) and the London magnetic penetration depth \(\lambda\) (few hundred nanometers). As it can be seen in Fig. 2(a), magneto-resistance measurements showed multiple dips for magnetic fields corresponding to integer multiples of the number of vortices per pinning site, even when above the melting temperature. Later on, experiments on BSCCO nanoribbons patterned also through focused ion beam milling with a square through-hole array [see inset Fig. 2(b)] were reported by Avci \textit{et al.} [15] In this case multiple and relatively deep field-matching effects were obtained, as shown in Fig. 2(b). It was found that matching effects were enhanced near the melting field and observed distinct regions with different dynamic phases for the vortex solid in the voltage-current behavior, as it had been predicted by Reichhardt \textit{et al.} [16] This patterning technique also allowed one of the first studies of vortex phases with ordered pinning in bulk high-\(T_C\) superconductors, in which Goldberg \textit{et al.} [17] identified in a patterned BSCCO crystal a first order melting transition in the bulk (that is, a sudden drastic reduction in the correlations between “pancake vortices”) while the solid phase remained in the surface.
Figure 2: a) Magneto-resistance curves at different temperatures for a BSCCO single crystal patterned with a triangular array of holes. Inset: SEM image of the sample. Adapted from ref. [14].

b) Resistance as a function of the magnetic field for various temperatures measured in a BSCCO nanoribbon patterned with a square array of holes. Inset: SEM image of the sample. Adapted from ref. [15]

The archetype high-temperature superconductor regarding technological applications is YBa$_2$Cu$_3$O$_{7-\delta}$. This is so because it presents a very strong native random pinning and, therefore, a high critical current. However, this also complicates the observation of artificial ordered pinning effects, and makes further enhancement of critical currents more challenging. Yet this system was actually one of the first among high-temperature materials in which periodic pinning was studied. Castellanos et al. [18] reported on YBCO films with a square array of holes [see inset Fig. 3(a)] fabricated through e-beam lithography and dry etching. Measurements of the critical current $J_C$ as a function of the applied magnetic field in Fig. 3(a) revealed the existence of peaks for multiple numbers of vortices per hole, analogous to the field-matching minima in magneto-resistance measurements. These were, nevertheless, shallower and less defined than those in low-T$_C$ superconductors or even BSCCO. More recently, Crassous et al. [19] reported on a reconfigurable pinning array using a ferroelectric/superconductor bilayer: BiFeO$_3$/YBCO. The regions of the BFO in which the polarization pointed towards the superconductor depressed the superconductivity locally and acted as vortex pinning sites. As shown in Fig. 3(b), the magneto-resistance measurements showed a shallow dip (black line) for the matching field that was not present (red line) after the array had been erased. This method offers the possibility to study multiple array geometries in a single sample with
relatively high matching fields in the kGauss range. The drawback of this approach is that the matching effects are relatively weak, and that it is only applicable to ultrathin (a few unit cells thick) YBCO films.

Figure 3: a) Critical current $J_c$ as a function of the applied field for an YBCO film patterned with a square array of holes. Inset: AFM image of the sample. Adapted from ref. [18]. b) Magneto-resistance curves measured before (red) and after (black) a pinning array was defined by PFM in a BFO/YBCO sample. Inset: PFM image of the pinning array. Adapted from ref. [19].

3. ARTIFICIAL ORDERED PINNING IN HIGH-TC SUPERCONDUCTORS

3.2 Masked O$^+$ ion irradiation

The technique allowing the strongest field matching effects in YBCO so far is masked ion irradiation. The concept was early explored by Kwok et al. [20], who patterned YBCO using heavy-ion irradiation to locally suppress superconductivity in a periodic distribution of regions, such as linear channels and lattices of squares. This was achieved irradiating through a system of collimators and slits that allowed the exposure of only micrometric regions of the sample. However the collective pinning properties of these structures were not demonstrated. It was only recently that Swiecicki et al. [21] reported on the use of a masked ion irradiation technique with PMMA photoresist and oxygen ions to create ordered pinning arrays in YBCO films grown by pulsed lased deposition. The effectiveness of the pinning arrays is demonstrated by the very pronounced field-matching effects observed in the magneto-resistance curve in Fig. 4, which corresponds to the square hole array mask sketched in the bottom right corner. Haag et al. [22] have recently reported on a
similar masked-irradiation approach which uses helium ions instead of oxygen, and a thin silicon stencil mask —instead of PMMA— with bigger features. These experiments have confirmed that masked ion irradiation is a powerful technique to obtain efficient pinning arrays in high-temperature superconductors.

Figure 4: a) Normalized resistance as a function of the magnetic field in a masked O⁺ ion irradiated YBCO sample. Adapted from ref. [21]. b) Illustration of the masked O⁺ ion irradiation process. The PMMA resist completely stops the ions. In the exposed areas point defects are induced in the YBCO, mainly in the form of interstitials and oxygen vacancies.

Throughout this work samples will be patterned using O⁺ ion irradiation through an e-beam lithographed PMMA mask [Fig. 4(b)].

### 3.2.1 Simulations of the O⁺ ion damage

We have used SRIM Monte Carlo simulations to model the irradiation of 50 nm YBCO films with oxygen ions at 110 keV. The O⁺ ion bombardment does not change the YBCO surface morphology [23], but creates point defects in the form of atom displacements and interstitials within the bulk of the material. The average energy transfer in collisions accompanying the ion irradiation is sufficient to displace all the atomic species in the material.

The SRIM code provides the “defects lateral distribution” (DLD) created by ions impinging a surface on one given spot. In Fig. 5(a) the simulation of the impact of 100 ions in one point was calculated using the ions energy as a parameter. The O⁺
track length into YBCO is of 150 nm, much longer that the film thickness. Thus, the ion-induced damage within the film reaches down into the STO substrate. For the experimental conditions used in the fabrication, the DLD does not vary significantly with depth. This means that an integration of the DLD along this direction may be performed.

The projected range of penetration of the O\(^+\) ions into PMMA is around 600 nm, smaller than the PMMA thickness of 800 nm. Therefore the ions are fully stopped by the mask and reach the YBCO film only through the mask holes. However, the irradiation defects appear not only underneath the exposed hole areas, but also a non-negligible amount of damage is induced outside the mask’s projected hole. This happens because ions spread out as they impinge on the YBCO film [Fig. 5(a)].

![Figure 5](image)

**Figure 5:** a) SRIM simulation of the point defects created by 110 keV oxygen ion irradiation in a YBCO/STO structure. b) Simulated local critical temperature map \(t_C\). The blue line represents the measurement temperature \(T\). Those zones in which the local \(t_C\) is smaller than \(T\) will remain in the normal state.

### 3.2.2 Simulation of the critical temperature \(T_C\)

The critical temperature reduction due to irradiation by different ions at varying energies has been shown to scale with the energy deposited into elastic collisions [24]. Therefore, the relevant parameter for the calculation of the \(T_C\) is the average number of displaced atoms, or displacements per atom (dpa) ratio. This quantity is calculated from the defects lateral distribution (DLD) previously obtained.
Although ion irradiation may in principle affect all atomic species, a comparison [24] of results from irradiation and thermal oxygen desorption experiments provides evidence that in both instances this mechanism is strongly related to oxygen vacancies in the b-axis Cu-O chains (see 2.1), which cause pair breaking effects. This was also seen by Arias et al. [25] in He\(^+\) ion irradiated YBCO.

Assuming a conduction electron spin flip scattering on isolated impurities, leading to a finite lifetime for the Cooper pairs, Abrikosov and Gorkov showed [26] that the critical temperature \(T_C\) variation can be calculated using:

\[
\ln \left( \frac{T_C}{T_0} \right) = \psi \left( \frac{1}{2} \right) - \psi \left( \frac{1}{2} + \frac{0.14}{0.015} \frac{dpa \cdot T_0}{T_C} \right)
\]

where \(\psi\) is the digamma function and \(T_0\) the virgin film critical temperature. The shape of the curve is shown in Fig. 6.

Therefore, from the simulated ion damage one can calculate the local critical temperature. In this manner, local \(T_C\) maps as that seen in Fig. 5(b) are produced. These predict that a spatial modulation of the critical temperature that mimics the mask geometry is obtained. When the system is cooled, the irradiated areas should remain in the normal state while the rest of the sample becomes superconducting. As it is shown in Fig. 5(b), the intersection of the blue line (measurement temperature \(T\)) with the simulated \(T_C\) profile determines which areas should remain in the normal state. In what follows we will see how these areas indeed remain in the normal state and behave as strong vortex pinning sites.
3.3 Pinning properties of masked O\(^+\) ion irradiated arrays

To study the pinning properties of the arrays defined via O\(^+\) ion irradiation, a square periodic array was defined on a 50 nm-thick YBCO thin film. The holes diameter in the mask is Ø\(\sim\)40 nm and the inter-hole distance is \(d = 120\) nm. A scanning electron microscope image of a mask similar to the one used is shown in the inset of Fig. 7.

If the pinning array is sufficiently strong, the vortex lattice deforms from its original triangular geometry to match the pinning array’s at certain values of the applied field. This is usually seen as minima in resistance versus applied field characteristics. A typical magneto-resistance curve, in which the field was applied perpendicular to the film plane (parallel to the YBCO c-axis), is shown in Fig. 7. The curve presents a series of minima at the matching fields \(B = \pm nB_\phi\), \(n\) being an integer or semi-integer, and \(B_\phi = \phi_0/d^2 = 0.144\) \(T\) the field at which the density of vortices equals the density of pinning sites in the square array. The strongest matching effects are seen for \(n = \pm 1, \pm 2\) and weaker ones for \(n = \pm 0.5\). This is as expected for fractional matching (semi-integer \(n\)) as vortices leave half the sites empty. Since vortices move across the array by hopping between neighboring pinning sites [27], the availability of empty sites makes stochastic depinning and lattice destabilization easier than when all sites are occupied. The latter requires a more synchronized depinning across the array.

From these magneto-resistance measurements we can conclude that the masked O\(^+\) ion irradiated regions behave as strong vortex-pinning sites.
3.4 **Vortex velocity analysis in periodic pinning arrays**

3.4.1 **Introduction to velocity analysis**

In order to characterize a pinning array and its optimal current range for magneto-transport measurements, it is necessary to study the array’s strength under different temperatures and dynamic conditions.

In Fig. 8 different magneto-resistance curves are presented for the previous sample –a YBCO film with a square array of pinning defects. Fig. 8(a) illustrates the effect of temperature at a constant current. One can see how raising the temperature makes the matching effects associated to the periodic pinning smoother and shallower. In turn Fig. 8(b) shows the behavior of magneto-resistance curves when the temperature is kept constant and the current is raised. A similar effect is seen: matching effects become smoother and shallower.
3. ARTIFICIAL ORDERED PINNING IN HIGH-TC SUPERCONDUCTORS

Figure 8: Magneto-resistance curves measured in a YBCO film with a square array of defects for a) constant current and changing temperature, and b) constant temperature and changing current. In both cases minima become shallower with increasing temperature/current. The matching field and out of matching field conditions are indicated.

The variation of the field-matching effects indicates that the periodic pinning efficiency is changing. We can use the relationship between the applied Lorentz force and the resulting vortex velocity to study under which conditions the periodic pinning is more efficient as compared to the native pinning. At the matching field condition [see Fig. 8(a)], the vortex lattice geometrically matches the periodic pinning array and the net pinning force from this source is maximized. Out of the matching condition, the effect of the periodic pinning is minimum and the majority of the vortex pinning comes from the native random pinning, which determines the background magneto-resistance level. Therefore, a measurement of the difference between the necessary Lorentz force to set vortices in motion with a certain velocity in both the matching and out-of-matching conditions will provide with a measure of the vortex pinning enhancement due to commensurability effects [28,29].

This can be done by measuring V(I) characteristics for the two different field conditions and a series of temperatures. From the injected current I it is possible to calculate the Lorentz force per unit vortex length exerted on the vortices $F_L = J\phi_0 = I\phi_0/dw$ where $J$ is the current density, $\phi_0$ is the magnetic flux quantum, $d = 50$ nm is the thickness of the film and $w = 40$ μm the width of the bridge. The measured

\[ F_L = \frac{I\phi_0}{dw} \]
voltage $V$ can be related to the vortex velocity $v$ using Lenz’s law: $\vec{E} = \vec{B} \wedge \vec{v} \Rightarrow v = V/Bl$, where $B$ is the applied magnetic field and $l = 200 \mu m$ is the distance between the voltage contacts.

As an example, Fig. 9 shows two $v(F_L)$ curves measured at the same temperature but different applied magnetic fields: at the matching field $B_\phi$ and at $B = 0.6B_\phi$, for which a local maximum is observed in the magneto-resistance – see Fig. 8(a). We note as $\Delta F_L(v)$ the difference between the Lorentz force required to achieve a given vortex velocity $v$ in those two applied fields. A graphical definition of $\Delta F_L$ is given in Fig. 9, where each vertical dashed line represents the Lorentz force at which $v = 0.0625$ m/s for each of the applied magnetic fields. The inset of Fig. 9 displays $\Delta F_L(v)$ extracted from the two curves in the main panel. From this, a study of the relation between the periodic pinning enhancement and the vortex velocity is readily available.

\[\Delta F_L(v)\]

This type of study has been done before using low-$T_C$ superconductors. Vélez et al. [28] fabricated Nb films with different ordered arrays of Ni magnetic dots, and extracted the $\Delta F_L(v)$ curves shown in Fig. 10(a) for a square, triangular and
rectangular pinning arrays, from top to bottom. The three curves show qualitatively a similar behavior. For slow vortex motion, $\Delta F_L = 0$ due to the dominance of intrinsic random pinning and the plastic motion of the vortex lattice. When the velocity is increased, the random pinning is overcome and the lattice flows in a more ordered state, commensurable with the periodic pinning array. After a maximum is achieved, $\Delta F_L$ decreases with increasing velocity. This is due to the fact that the influence of periodic pinning is gradually reduced over increasingly high drives. A similar analysis [29] was performed on a Nb film with a square array of Ni magnetic dots, with measurements at different temperatures above and below the glass transition. In Fig. 10(b) two examples are shown: the curve labeled $t_A$ is representative of $T > T_g$, and $t_B$ of $T < T_g$. For the latter, as in the previously cited work, the curves show an absolute maximum in $\Delta F_L$ [Fig. 10(c)], but here the curves shift to the left as temperature is decreased. This is represented in the inset of Fig. 10(c), where the velocity at which $\Delta F_L$ decreases 40% after the maximum is plotted as a function of temperature. We will use these analyses on Nb later for comparison with results obtained from similar studies on our high-$T_C$ superconducting YBCO samples.

Figure 10: a) $\Delta F_L(v)$ curves for a Nb film with three different Ni magnetic dot arrays: square, triangular and rectangular, from top to bottom. Adapted from Ref. [28]. b) $\Delta F_L(v)$ curves for a Nb film with a square array of Ni dots at two temperatures: $t_A = 8.185K$ above and $t_B = 8.120K$ below the glass transition temperature. c) $\Delta F_L(v)$ curves for the same sample at temperatures $T > T_{g1}$ below $T_g$. Inset: The velocity at which $\Delta F_L$ has decreased 40% from its maximum as a function of temperature. Both b) and c) were adapted from Ref. [29].
3.4.2 Experimental results

We have measured a series of V(I) curves for different magnetic fields and temperatures, and we studied the periodic pinning enhancement in the first and second matching fields. Thus, \( v(F_L) \) curves at various temperatures were measured at \( B_\phi \) and \( B = 0.6B_\phi \), and at \( B = 2B_\phi \) and \( B=1.8B_\phi \), as the matching and out of matching conditions for each case. From this \( \Delta F_L(v) \) curves are obtained as previously explained.

Figs. 11(a) and (b) show two sets of isotherms \( \Delta F_L(v) \), respectively for the first and second matching fields. The absolute values of \( \Delta F_L \) are around an order of magnitude larger for the first matching field than for the second one. The color of each curve indicates the temperature at which it was measured, bright red and bright blue corresponding respectively to 49K and 20K. From the experiments in chapter 4 we know that those temperatures range from above to below the glass transition temperature \( T_g = 34.5K \) for the first matching and \( T_g = 29.4K \) for the second matching field.

From the inspection of Fig. 11, we learn two things: i) there are three distinct temperature regimes for the dependence of \( \Delta F_L \) with velocity, and ii) the temperature evolution of \( \Delta F_L \) for a given velocity is non-monotonic. These observations are valid for both the first [Fig. 11(a)] and the second [Fig. 11(b)] matching fields.
3. ARTIFICIAL ORDERED PINNING IN HIGH-\(T_C\) SUPERCONDUCTORS

Figure 11: a) and b) \(\Delta F_L\) as a function of the vortex velocity for different temperatures ranging from \(T = 49\) K (bright red) to \(T = 20\) K (bright blue) for different applied magnetic fields. c) and d) display for the indicated applied magnetic field three curves each, representative of the temperature regimes observed.

Regarding i), the three regimes can be easily identified in the three curves displayed in Fig. 11(c) and (d) for each temperature. In the first regime at high temperatures (red), \(\Delta F_L\) grows with increasing velocity; in the second regime (green), which corresponds to temperatures that are close to \(T_g\), \(\Delta F_L\) remains nearly constant in the entire velocity range; finally, for low temperatures (blue), \(\Delta F_L\) decreases with increasing velocity for the first matching field and, for the second matching field [Fig. 11(b) and (d)], \(\Delta F_L\) becomes negative –which means that at low temperature no commensurability effects are observed at the second matching field. In some of the temperature regimes, the behavior observed here is very much in contrast with that observed in the experiments on low-\(T_C\) Nb films with periodic pinning arrays summarized above. While the high-temperature regime in which \(\Delta F_L\) increases with the vortex velocity is observed both in Nb [\(T_A\) in Fig. 10(b)] and YBCO with periodic
pinning, $\Delta F_L(v)$ is very different for both systems at temperatures below $T_g$. For Nb thin films $\Delta F_L(v)$ is small at low velocities and peaks at intermediate ones [Fig. 10(c)], while for YBCO $\Delta F_L(v)$ is maximum at the lower velocities, and steadily decreases until a moderate upturn is observed at very high velocity.

To better illustrate the temperature dependence of $\Delta F_L$, we use the data representation shown in Fig. 12. Here $\Delta F_L$ is plotted as a function of the temperature for two particular velocities, 0.025 m/s (black) and 5 m/s (red), for both the first [Fig. 12(a)] and the second [Fig. 12(b)] matching fields. In all cases, the curves display a broad maximum at intermediate temperatures, between $\sim 30$ K and $\sim 33$ K, which is close to the irreversibility line.

Figure 12: Evolution of $\Delta F_L$ as a function of temperature for two given velocities –0.025 m/s (black squares) and 5 m/s (red circles)– in a) the first matching field and b) the second matching field.

### 3.4.3 Discussion

Let us start discussing the temperature evolution displayed in Fig. 12. Both for the first and second matching fields, Fig. 12(a) and Fig. 12(b), $\Delta F_L$ is maximum around the glass transition temperature $T_g$. That is, matching effects are enhanced around the glass transition as the vortex solid phase develops and pinning becomes more efficient. This enhancement is as the one Avci et al. [15] had seen in BSCCO nanoribbons. The fast decay of $\Delta F_L$ above that temperature is explained by the reduction of periodic pinning effects due to the thermally activated flux flow in the vortex.
3. ARTIFICIAL ORDERED PINNING IN HIGH-TC SUPERCONDUCTORS

liquid phase [30]. Below the glass transition, $\Delta F_L$ decays with decreasing $T$. This can be understood by considering that, upon lowering the temperature, the background pinning produced by the intrinsic *disordered* distribution of defects grows faster than the periodic pinning. Thus, when the temperature is reduced, the pinning force available at the “out-of-matching” field grows comparatively faster than the pinning enhancement at the matching condition, making the relative pinning increase at matching weaker. This trend is further exacerbated at the second matching field [see Fig. 12 (b)], for which $\Delta F_L$ becomes negative at the lowest temperatures. This means that the commensurability effect is not strong enough to provide a more efficient pinning at the matching condition than out of it. The reason is that, at this field, the number of interstitial vortices is high (at the second matching field only half of the vortices are bound to artificial pins), which makes the intrinsic random pinning dominant.

We can now discuss $\Delta F_L(v)$ in each of the temperature regimes observed in Fig. 11. For temperatures above $T_g$, $\Delta F_L(v)$ increases with increasing vortex velocity [see red curves in Figs. 11(a) and (b)]. This can be understood by considering that (i) the low velocity limit corresponds to the linear V-I regime in which the Ohmic response of the vortex-liquid is probed (the current is not probing the pinning since the depinning mechanism is thermal activation), and (ii) that with increasing velocity the system gradually enters the nonlinear V-I regime and vortex pinning is probed [29], and so $\Delta F_L(v)$ increases monotonically. Around the glass transition temperature, $\Delta F_L(v)$ is roughly constant (green curves). This can be regarded as the transition regime into the low-temperature one, in which $\Delta F_L(v)$ decreases with increasing velocity (blue curves). This behavior, which contrasts with that observed in the Nb experiments in Fig. 10(c), could be linked to the weak correlations along the vortex line characteristic of the system investigated here. As we will demonstrate further ahead, the sample shows an enhanced anisotropy that yields a quasi-two dimensional behavior and makes vortex lines “soft” and prone to wandering and deformation – contrary to the case of low-T$_C$ superconductors, in which vortex lines can be considered three dimensional and “rigid” [29]. Early work by van der Beek *et al.* [31] on heavy-ion-irradiated BSCCO showed that in a highly anisotropic system and low driving forces, the correlation length along the vortex line decreases as the Lorentz
force is increased. Therefore, we can argue that the periodic pinning is more efficient at lower velocities because a lower Lorentz force is exerted on the vortices, yielding longer correlations along the vortex lines. This would produce stronger commensurability effects, since vortices would be less prone to wander and be affected by the disordered pinning.

Finally, the fact that $\Delta F_L(v)$ curves for high and medium temperatures display a constant value for relatively low velocities indicates that this is the current range to be favored in magneto-transport experiments so the strength of the periodic pinning array remains constant.

### 3.4.3 Conclusions:

We have investigated the temperature and velocity dependence of the periodic pinning enhancement due to commensurability effects. The comparison of the Lorentz force necessary to move vortices at a given velocity for the matching and the out-of-matching fields provided us with insights on the strength of the periodic pinning in its balance with the intrinsic random pinning. In particular we observe two characteristic features: i) at low temperatures the periodic pinning array becomes most effective for lower velocities, in contrast to previous reports in low-$T_C$ superconductors, probably due to the high anisotropy in our system; and ii) there is an optimum temperature for the strength of periodic pinning, which is around the glass transition temperature.
3. ARTIFICIAL ORDERED PINNING IN HIGH-\(T_C\) SUPERCONDUCTORS

References:


4. Analysis of the interplay between intrinsic and artificial ordered pinning on a nanostructured YBCO thin film

4.1 Introduction

4.1.1 The vortex phase diagram

4.1.2 The vortex-glass and Bose-glass scaling theories

4.2 Critical scaling analysis of V(I) isothermal measurements

4.3 Angular magneto-resistive measurements

4.3.1 Magneto-resistance at different fixed angles

4.3.2 Angular magneto-resistance at different constant fields and its simulation

4.4 Discussion

4.4.1 Critical scaling

4.4.2 Angular magneto-resistance

4.4.3 Vortex accommodation to the pinning landscape

4.5 Conclusion
In chapter 3 we have seen that the periodic pinning array introduced in our samples deforms the vortex lattice so that vortices match the array’s geometry. Nevertheless, since other native pinning sources are present, it is important to determine their influence on vortex behavior. To resolve this question, in this chapter we will investigate how the artificial ordered and native pinning sources affect the vortex phase diagram in our YBa$_2$Cu$_3$O$_{7-\delta}$ samples.

To determine the irreversibility line that separates the liquid from the glass phase we will use a critical scaling analysis on sets of V(I) characteristics. After that, we will perform measurements with tilted magnetic fields so we can ascertain the vortex pinning angular behavior. This will allow us to establish what kind of glass transition occurs in our system and determine the interplay between intrinsic and artificially ordered pinning.
4. INTERPLAY BETWEEN INTRINSIC AND ARTIFICIAL ORDERED PINNING

4.1 Introduction

4.1.1 The vortex phase diagram

In the mixed state of type-II superconductors different vortex phases can be found as a function of temperature. At low temperatures and in the presence of disorder (pinning defects), a glass phase is observed due to the absence of long range translational order between vortices. In this phase, V(I) characteristics are non-linear over the whole current range and the critical depinning current $J_C$ is well defined [see Fig. 1(a)]. The vortex movement in the glass phase is elastic as a result of infinite vortex correlations [1]. When the temperature is gradually increased, thermal vortex vibrations become greater, eventually exceeding the inter-vortex spacing [2]. This results in the loss of vortex correlations and the formation of a liquid phase. This is reflected in V(I) characteristics as the appearance of an Ohmic tail at low currents, that is, a regime in which the current is a non-perturbing probe of the vortex matter, and only thermally activated vortices move while the rest remain pinned. This plastic regime is called the thermally activated flux flow (TAFF). The Ohmic regime disappears with increasing currents: the V(I) becomes non-linear as the Lorentz force created by the current gradually depins the remaining vortices [3], leading to a resistance increase. The current threshold for which this happens gradually becomes bigger with increasing temperature.

The vortex phase diagram and the transition between the glass and liquid phases are determined by the balance between elastic energy, thermal energy and pinning energy. Of course, the presence of ordered pinning is expected to modify the vortex phase diagram. However, very few studies have been performed on the matter using high-Tc superconductors. All of them are based on the highly anisotropic Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO), and exclusively address the situation in which vortices are perpendicular to the surface. Early experiments by Ooi et al. [4] on single-crystalline BSCCO thin films with triangular arrays of through-holes, found that matching effects could be observed well above the melting line. Later Avci et al. [5] used BSCCO nano-ribbons patterned with a square through-hole array and found matching effects above and below the melting line, near which they were greatly
enhanced. Also, Goldberg et al. [6] fabricated BSCCO single crystals with a triangular array of surface holes, and observed deep changes in the vortex phase diagram that suggested the possibility of a Mott insulator phase at the matching field.

In contrast to the BSCCO used in the experiments summarized above, YBCO presents a smaller anisotropy, so that coupling between CuO$_2$ layers is greater and vortices present a more 3D-like behavior [2]. Also, YBCO films grown by pulsed laser deposition have a much stronger native random pinning, which results in a continuous second order transition [7,8], very different from the abrupt first order one seen in BSCCO [6] and untwinned YBCO single-crystals [9].

The purpose of the experiments done here is to determine how the YBCO characteristics interplay with artificial periodic pinning, and what are the effects on the vortex phase diagram.

4.1.2 The vortex-glass and Bose-glass scaling theories

The transition between different phases of the vortex matter can be quantitatively characterized. The so-called “critical isotherm” is the V(I) measured at the glass transition temperature $T_g$, and determines the point that separates the curves which verify $\lim_{I \to 0} V/I \neq 0$ (above $T_g$) and $\lim_{I \to 0} V/I = 0$ (below $T_g$). Another criterion can be used as well to define $T_g$: the temperature that separates the curves that present positive curvature in the low current limit (above $T_g$) $\lim_{I \to 0} d^2V/d^2I > 0$, and those with a negative curvature (below $T_g$) $\lim_{I \to 0} d^2V/d^2I < 0$.

A critical scaling analysis is a tool used to study the vortex phase transition and the dimensionality of the system. These are influenced by the type of pinning present. Two critical scaling models are the most frequently applied: the vortex-glass (VG) and the Bose-glass (BG) models. The first one was proposed by D. Fisher, M. Fisher and D. Huse [1], who considered random point defects as the source of pinning. The second one was introduced by D. Nelson and V. Vinokur [10], who mapped vortices along correlated disorder (columnar defects, twin boundaries, etc.) in three dimensions onto bosons in potential-energy minima in two dimensions.
Two quantities are considered when analyzing a glass transition: the correlation length in the glass $\xi_g$ and the glass relaxation timescale $\tau_g$. When approaching the glass transition temperature both quantities diverge [11]. In the case of the vortex glass the correlation length diverges as $\xi_{VG} \propto |1 - T/T_g|^{-\nu_{VG}}$ and the relaxation time as $\tau_{VG} = \xi_{VG}^{-z_{VG}}$. $\nu_{VG}$ and $z_{VG}$ are the static and dynamic critical exponents for the vortex glass. Analogously, for the Bose glass $\xi_{BG} \propto |1 - T/T_g|^{-\nu_{BG}}$ and $\tau_{BG} = \xi_{BG}^{-z_{BG}}$.

According to the two models, when the critical scaling is applied to a set of measured $V(I)$ curves, the data should collapse into two master curves [see Fig. 1(b)], depending on whether they are above or below the glass transition temperature. Adapted from Ref. [8]

For the vortex glass, this should happen when using the following scaling ansatz:

$$V \xi_{VG}^{2+z_{VG}D}/I = \chi_{\pm} (I \xi_{VG}^{-1}/T)$$

(1)
where D is the dimensionality of the vortex glass, i.e., the number of dimensions in which the correlation length $\xi_{VG}$ diverges; and $\chi_{\pm}$ are the scaling functions above (+) and below (-) $T_g$. For the Bose glass:

$$V\xi_{BG}^{-2}/l = \chi_{\pm} (I\xi_{BG}^3/T)$$

(2)

It is worth mentioning that both models are formally identical, as if one is able to find a set of parameters $D$, $T_g$, $v_{VG}$ and $z_{VG}$ that allows scaling the data according to the vortex-glass model, the scaling according to the Bose-glass model is automatically achieved [7,12] using the same $T_g$ and the critical exponents:

$$v_{BG} = (D - 1)v_{VG}/3$$

(3)

$$z_{BG} = (3z_{VG} - D + 4)/(D - 1)$$

(4)

In order for the scaling analysis and the extracted dimensionality to be physically acceptable, the critical exponents must lie within a given range of values. These are predicted by the theory [1,10], and have been consistently found in experiments. For a vortex glass, these are $1 \leq v_{VG} \leq 2$; $4 \leq z_{VG} \leq 6$ [7,8,12,13]. For a Bose glass $0.8 \leq v_{BG} \leq 1.8$; $6 \leq z_{BG} \leq 9$ [8,12,14–16].

A way to check the consistency of the dynamic critical exponent $z$ comes from the prediction that the critical V(I) isotherm (the one at $T_g$) fulfills [1]:

$$V \propto I^{\alpha +1}$$

(5)

where for the vortex glass:

$$\alpha = (z_{VG} + 2 - D)/(D - 1)$$

(6)

and for the Bose glass:

$$\alpha = (z_{BG} - 2)/3$$

(7)

Finally, it is not always possible to discriminate between both models using only the scaling analysis. However, due to the directional character of the defects considered in the Bose-glass model, an increase in the glass transition temperature is expected [17] when the field is applied parallel to them. This is in contrast with the
isotropicity of the pinning considered in the *vortex-glass* model, where a cusp in $T_g$ would be absent. This indicates that it is necessary to study the angular behavior of the system to obtain a complete analysis.

### 4.2 Critical scaling analysis of V(I) isotherm measurements

For the following investigation we have used the same sample as in the previous chapter: a 50 nm-thick YBCO thin film with a square pinning array defined using masked O$^+$ ion irradiation (holes diameter 40 nm and inter-hole distance 120 nm).

V-I characteristics were measured in several applied fields and directions, for each of which a set of isotherms was measured in a wide temperature range. Table I summarizes the magnetic fields $B$, $\theta$ is the angle between the applied field and the surface normal $\theta$ [inset of Fig. 3(a)]. In Fig. 2(a) the set corresponding to $B = 2B_\phi$ and $\theta = 0$ is shown. For the highest temperatures (bright red curves) an Ohmic regime is observed at low currents, up to a current threshold where the curves become non-linear and display a positive curvature. When decreasing the temperature, this current threshold becomes smaller. The light blue curves at low temperatures present a non-linear behavior within the entire experimental window with negative curvature. Also, they display vanishing resistance in the low current limit: $\lim_{I \to 0} V/I = 0$. This behavior is indicative of a second order continuous vortex phase transition. At high temperatures, the Ohmic regime indicates that the system is in the liquid phase and that vortices suffer a plastic motion. With decreasing temperature, the current level after which a positive curvature is observed becomes smaller. When the curvature at low currents changes from positive to negative, the system is in the glass phase. Vanishing resistance at low currents reflects the fact that infinite vortex correlations make vortices hard to depin.

We will first determine the glass transition temperature. In the inset of Fig. 2(b) the derivative $d[\log(V)]/d[\log(I)]$ of the curves in Fig. 2(a) is shown. This allows us to determine $T_g$ by discriminating the isotherms above, whose slope
increases in the low current limit, \( \lim_{I \to 0} d^2[\log(V)]/d^2[\log(I)] > 0 \), and those below, whose slope decreases in the low current limit, \( \lim_{I \to 0} d^2[\log(V)]/d^2[\log(I)] < 0 \). Note how experimentally this provides only with an upper limit estimate of the \( T_g \), since the lowest current range of the V(I) is inaccessible due to the noise floor in the setup. The horizontal lines in the inset of Fig. 2(b) indicate the temperature range between the last curve to show increasing slope at low currents and the first one to show decreasing slope. The \( T_g \) values used in the following critical scaling are always kept within this range: [28K, 30K].

Once the \( T_g \) is delimited, we attempt the collapse of the curves using the vortex-glass model [Eq. (1)]. For the Bose-glass model [Eq. (2)] it suffices to apply Eqs. (3) and (4). Successful data collapses –as the one shown in Fig. 2(b)– were obtained for every V(I) set available. In every case, a \( D = 3 \) vortex glass required unphysical \( z_{VG} \sim 10 \), and the Bose glass yielded unphysical \( z_{BG} \sim 15.6 \). Only the \( D = 2 \) vortex glass produced acceptable critical exponents. These can be found in Table I. We conclude from this that a 3D vortex glass as well as a Bose glass are to be ruled out.
4. INTERPLAY BETWEEN INTRINSIC AND ARTIFICIAL ORDERED PINNING

<table>
<thead>
<tr>
<th>$B$ (T)</th>
<th>$\theta$ (deg)</th>
<th>$B \cos(\theta)/B\phi$</th>
<th>$D$</th>
<th>$v \pm \sim 0.1$</th>
<th>$z \pm \sim 0.2$</th>
<th>$z_{\text{slope}}$</th>
<th>$T_g \pm 0.5$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.085</td>
<td>0</td>
<td>0.59</td>
<td>2</td>
<td>2</td>
<td>4.8</td>
<td>4.9–5.4</td>
<td>30.5</td>
</tr>
<tr>
<td>0.143</td>
<td>0</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>5</td>
<td>5.4–5.7</td>
<td>34.5</td>
</tr>
<tr>
<td>0.260</td>
<td>0</td>
<td>1.81</td>
<td>2</td>
<td>1.9</td>
<td>5.5</td>
<td>5.6–6.1</td>
<td>27.5</td>
</tr>
<tr>
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<td>0</td>
<td>2</td>
<td>2</td>
<td>1.8</td>
<td>5</td>
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<td>29.4</td>
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<tr>
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<td>4.8</td>
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</tr>
<tr>
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<td>–</td>
<td>2</td>
<td>1.6</td>
<td>4.5</td>
<td>5–5.8</td>
<td>30.5</td>
</tr>
</tbody>
</table>

Table 1: Scaling parameters for $V$–$I$ sets according to the vortex-glass model. $D$ is the dimensionality, $v$ and $z$ the static and dynamic critical exponents, $z_{\text{slope}}$ the dynamic critical exponent expected from the slope of the $I$–$V$ curves nearby the critical isotherm, and $T_g$ the glass transition temperature.

To verify the consistency of the dynamic critical exponent $z_{VG}$ we turn to Eq. (5). From it we know the critical isotherm’s slope should go as $\alpha + 1$, where $\alpha$ is given by Eq. (6) for the vortex glass. Using $D = 2$ we obtain an estimate for the dynamic critical exponent $z_{\text{slope}}$. The estimates for each $V(I)$ are found in Table I. In every case a good agreement is found between $z_{VG}$ and $z_{\text{slope}}$.

What we learn from the different scaling analysis performed is that: (i) the critical scaling parameters are essentially the same regardless of the magnitude and direction of the applied field –and in particular, regardless of whether it is the matching field or not; (ii) in all cases, a quasi-two-dimensional ($D = 2$) transition is observed; and (iii) as shown in the inset of Fig. 2(a), $T_g(B)$ is non-monotonic and presents enhancements at the matching fields. While (iii) is to be expected from the earlier experiments on BSCCO [4–6] cited above, (i) and (ii) are not. These imply that the dimensionality of the vortex-glass transition has been reduced from $D = 3$, usually found in virgin YBCO, to $D = 2$. 

59
4.3 Angular magneto-resistive measurements

The vortex angular behaviors for the vortex glass and the Bose glass are expected to show great differences. This can be used as a tool to solve the ambiguity that the critical scaling produces in some cases, as well as to obtain information about the pinning anisotropy and strength, and its evolution with the magnetic field.

4.3.1 Magneto-resistance at different fixed angles

We have measured magneto-resistance curves at different angles $\theta$ of the applied magnetic field, for temperatures above and below $T_g$. These were done in the constant Lorentz force geometry, and are displayed in Fig. 3(a) color-coded (only one temperature above $T_g$ represented). We see that the matching effects are displaced to higher fields as we increase the tilting angle. In Fig. 3(b) the same data are shown but this time the horizontal axis has been scaled to represent them as a function of the out of plane field component $B \cos \theta$. We readily notice that the positions of the matching effects scale perfectly, so that the matching condition becomes $B \cos \theta = nB_\phi$. This is also shown in the inset of Fig. 3(b), where the relative position of the first matching field $B_1/B_\phi$ is plotted as a function of $1/\cos \theta$. A straight line of slope one is found both for temperatures above (triangles) and below (circles) the glass transition temperature. This means that this behavior occurs regardless of whether the system is in the glass phase or not. It is worth noting that in Fig. 3(b) we do not observe a perfect collapse of all the curves, which implies that there is a finite anisotropy in the system, since the in-plane component $B \sin \theta$ contributes to the background magneto-resistance.
4. INTERPLAY BETWEEN INTRINSIC AND ARTIFICIAL ORDERED PINNING

![Image](image.png)

Figure 3: a) Resistance as a function of the magnetic field (normalized to \(B_\phi\)) for different field directions \(\theta\) indicated by the label. Measured at \(T=0.87\ T_c\) and with \(J=2.5\ \text{kA cm}^{-2}\). The first matching field observed for each angle is marked with an arrow. Inset: Scheme with the definition of \(\theta\) and \(b\) resistance as a function of the magnetic field component parallel to the c-axis \(B \cos(\theta)\), normalized to \(B_\phi\), for different field directions \(\theta=0, 30, 45, 60\) and 70 degrees (bottom to top). Measured at \(T=0.87\ T_c\) and with \(J=2.5\ \text{kA cm}^{-2}\). Inset: matching field corresponding to \(n=1\) as a function of \(1/\cos(\theta)\), for \(T=49\ \text{K}=T_c\) (triangles) and \(T=0.52\ T_c\) (circles).

4.3.2 Angular magneto-resistance at different constant fields and its simulation

To gain further insight into the angular dependence of the magneto-resistance, we performed a series of resistance measurements at constant temperature, field and current, in which we continuously varied the angle \(\theta\). Some examples are shown in Figs. 4(a)-(c) (hollow symbols) for \(T > T_g\), although, qualitatively, the same behavior is observed below \(T_g\). One can see that the curves display resistance minima for \(\theta = \pm 90^\circ\), i.e. for when the field is applied parallel to the \(ab\) plane, as it is usual in plain YBCO due to its intrinsic anisotropy [18,19]. As we had previously observed, we also find matching effects for when the condition \(B \cos \theta = nB_\phi\) is met. Figs. 4(a) and (b) display the measurements for applied fields \(B = 0.5B_\phi\) and \(B = B_\phi\), which respectively show the minima corresponding to \(n = 0.5, 1\) centered around \(\theta = 0^\circ\). Remarkably, for \(B = B_\phi\) the minimum corresponding to \(n = 1\ (\theta = 0^\circ)\) is as deep as the one due to intrinsic anisotropy (\(\theta = \pm 90^\circ\)). Note that the minimum
corresponding to $n = 0.5$ can also be observed in Fig. 4(b) around $\theta = \pm 60^\circ$. The curves in Fig. 4(c) illustrate the critical character of the matching effects: the applied field is only slightly above $B_\phi$ (2% and 10% respectively), yet the minimum corresponding to $n = 1$ is split into two that strictly satisfy $B \cos \theta = B_\phi$. A similar effect is seen in Fig. 4(d) for $n = 2$ in which the applied field was $B = 2.03B_\phi$. In the latter and Fig. 4(e) various minima appear for different orders $n$, aside from the usual minima at $\theta = \pm 90^\circ$.

A quantitative analysis of the curves in Figs. 4(a)-(e) was done using a model built to simulate the angular magneto-resistance. This model is adapted from the thermally activated flux flow expression for the resistance [2]:

$$R(B, T, \theta) = R_0 \cdot \exp \left(\frac{-U(B, T, \theta)}{K_B T}\right)$$  \hspace{1cm} (8)

where $K_B$ is Boltzmann’s constant and $U(B, T, \theta)$ is the vortex activation energy. The latter is field, temperature and angle dependent and represents the necessary energy...
for a vortex to escape the pinning force of the potential-energy well. It can be expressed as follows [2]:

$$U(B,T,\theta) = \frac{\beta}{B \cdot \varepsilon(\theta)^{a}} \left(1 - \frac{T}{T_C}\right)$$  \hspace{1cm} (9)

where $\beta$ is an energy scale, $\alpha$ determines the field dependence, and $\varepsilon(\theta) = \sqrt{\cos^2 \theta + \varepsilon^2 \sin^2 \theta}$ represents the anisotropic behavior, $\varepsilon$ being a constant. Note that $\varepsilon(\theta)$ comes from the scaling approach developed by Blatter et al. [20], which allows describing the angular dependent magneto-resistance of anisotropic superconductors in terms of an effective field $B_{eff} = B\varepsilon(\theta)$, and in the limit $\varepsilon \rightarrow 0$ (infinite anisotropy) becomes the two-dimensional Kes’ model applied usually to highly anisotropic superconductors [21]. However, as it was shown in Fig. 4(b), there is not a single parameter $\varepsilon$ that can be used to scale the magneto-resistance over the whole angular range.

To solve this, we assume that each individual matching order $n$, as well as the intrinsic anisotropy minima at $\theta = \pm 90^\circ$ originate from a different “source” of pinning with its own characteristic angular-dependent activation energy $U_i$. Accordingly, we expressed the total activation energy as the sum of the activation energies from each “source” of pinning $U = \sum_i U_i$. The notation used will be $i$ for all the “sources” of pinning, $ab$ for the intrinsic anisotropy “source”, and $n$ for each “source” corresponding to a different order of matching by the periodic pinning. This way: $i = \{ab, n\}$; $n = \{0.5, 1, 2, 3\}$.

Applying this to Eq. (9) we obtain the following expression to simulate our experimental results:

$$R = R_0 \cdot \exp \left\{ \sum_i \frac{-C_i}{[B \cdot \varepsilon(\theta - \delta_i)]^{a_i}} \right\}$$  \hspace{1cm} (10)

Since the measurements are performed at constant temperature, we have compressed all temperature dependent terms and the energy scale $\beta$ into a single parameter $C_i$. $\delta_i$ is a constant that represents the angle $90 - \delta_i$ at which the activation energy is centered or, in other words, the angle at which it is enhanced. This is determined by the position of the minima. For the intrinsic anisotropy pinning, which

63
is stronger when the field is parallel to the \(ab\) plane, \(\delta_{ab} = 0\). For each of the orders \(n\) of the matching effects caused by the periodic pinning, \(\delta_n = 90^\circ - \arccos(nB_\theta/B)\) (where \(n\) takes the values 0.5, 1, 2 and 3). \(\alpha_i = 1\) was chosen in all cases, based on previous results [19] of angular measurements in YBCO pristine thin films. In this manner, our only simulation parameters are \(R_0\) and a series of pairs \(\{C_i, \varepsilon_i\}\). Note that the latter bear the relevant physical meaning: \(C_i\) tells us about the pinning strength or pinning energy, and \(\varepsilon_i\) about the effective anisotropy of each particular source of pinning.

We used Eq. (10) to simulate the experimental data. This was done using the OriginPro software, manually searching for the best reproduction of the experimental curve. The results are shown as solid lines in Fig. 4(a)-(e) and demonstrate a remarkable agreement with the experimental data. The simulation parameters \(C_i, \varepsilon_i\) are gathered in Fig. 4(f) and (g) respectively. We see that the ones corresponding to the non-periodic pinning \(C_{ab}, \varepsilon_{ab}\) (hollow circles) are constant independently of the applied magnetic field. Moreover, the corresponding anisotropy parameter \(\gamma_{ab} = 1/\varepsilon_{ab} \approx 15 - 20\). This is to be compared with the much smaller anisotropy parameter of pristine YBCO films \(\gamma \approx 5 - 7\). The simulation parameters describing the minima associated with the periodic pinning \(C_n, \varepsilon_n\) are indeed field dependent. In particular, \(C_1\) is the largest at any field. This is as expected, given that the magneto-resistance minima for \(n = 1\) are the deepest [see Fig. 3(a)]. It is remarkable that \(C_1 \geq C_{ab}\), and specially that \(C_1 \gg C_{ab}\) at the matching field \(B = B_\phi\). That is, the periodic pinning array is the strongest source of pinning. Otherwise, \(C_n\) decreases with increasing field for all \(n\). Regarding \(\varepsilon_n\) [Fig. 4(g)], in all cases it decreases with increasing applied field, the highest value being for when each \(n\)’s matching field \(B = nB_\phi\) is applied, for which \(\varepsilon_n \approx 0.18 - 0.35\). The origin of the field-dependence of \(C_n\) and \(\varepsilon_n\) will be explained below, in the discussion section.
4. INTERPLAY BETWEEN INTRINSIC AND ARTIFICIAL ORDERED PINNING

4.4 Discussion

4.4.1 Critical scaling

The critical scaling analysis of the V(I) isothermal measurements showed that both a Bose glass and a $D = 3$ vortex glass were to be ruled out, and only a $D = 2$ vortex glass was acceptable. This quasi-two-dimensional vortex glass is not to be confused with the pure 2D vortex glass in which correlations are completely non-existent in one dimension and the glass transition temperature is zero. This pure 2D vortex glass was found by Wen et al. [22] in Tl$_2$Ba$_2$CaCu$_2$O$_{8+}$ (TBCCO) films at high magnetic fields with an anisotropy parameter $\gamma \sim 70 - 150$, or in highly-deoxygenated YBCO films by Sefrioui et al. [8] reporting $\gamma \sim 60$. Our experiments show a finite $T_g$ implying the existence of correlations along the three dimensions. Due to the fact that the dimensionality of the transition indicates in how many dimensions correlations diverge, we see correlations are infinite in two dimensions and remain finite in one (shorter than the sample’s size). Considering that we obtained $D = 2$ in every situation, even when the flux lattice matches the periodic pinning array, it is reasonable to assume that glass correlations diverge in-plane and remain finite along the $c$-axis. In conclusion, an important effect of the masked O$^+$ ion irradiation is to reduce the vortex-glass correlations along the $c$-axis. This is supported by the relatively large anisotropy $\gamma_{ab} = 15 - 20$ deduced from the angular magneto-resistance simulations, which suggest a weaker coupling between CuO$_2$ planes than in pristine YBCO [1]. In this respect –excluding concerning the quasi-two-dimensional scaling and the anisotropy $\gamma_{ab}$– the behavior observed here resembles that of intermediate-deoxygenated YBCO films also reported by Sefrioui et al. [8]. This can be understood if one considers that during the O$^+$ ion irradiation, point defects are also induced in between the mask holes. These point defects are in the form of oxygen vacancies and interstitials which would have a similar effect to that of YBCO deoxygenation. The reduced correlations along the vortex line due to weaker coupling between CuO$_2$ planes indicates that vortices in our system are unusually soft and, for example, they are prone to wander out of the artificial pinning sites to accommodate intrinsic sources of pinning.
4.4.2 Angular magneto-resistance

The angular dependence of the magneto-resistance also differs from that expected in typical Bose-glass behavior. In the latter, a resistance minimum associated with the correlated pinning appears only for when the magnetic field is applied parallel to the defects. Moreover, this angle is independent of the magnitude of the applied magnetic field. This was seen for example in heavy-ion irradiated TBCCO films [23] [Fig. 5(a)], and in heavy-ion irradiated YBCO single crystals [24] [Fig. 5(b)]. Note how the minima for both Bose-glass experiments appear at $\theta = 0$ for magnetic fields below, equal to and above the matching field $B_\phi$. Contrary to this, in the present experiments the resistance minima in $R(\theta)$ associated with the artificial pinning may appear for any direction of the applied field $0 \leq \theta < 90$, which depends on the field magnitude. Vortex localization occurs for particular values of the out-of-plane component of the magnetic field, that is, for certain in-plane vortex densities. This implies that vortex localization in artificial pinning sites is dictated by vortex-vortex interactions within the $ab$ plane –at variance with Bose-glass systems, in which the vortex line tension determines the angle of accommodation within the defects [10]. In conclusion, in the present system in-plane correlations between vortices dominate over correlations along the vortex line.

![Figure 5: Angular magneto-resistivity measurements for different applied fields and temperatures in heavy-ion irradiated a) TBCCO films (adapted from Ref. [23]) and b) YBCO single crystals (adapted from Ref. [24]). In both cases a minimum is observed when the field is applied parallel to the defects ($\theta=0$)]
4.4.3 Vortex accommodation to the pinning landscape

Vortices may accommodate to the pinning landscape in tilted magnetic fields in different ways. We consider two possibilities: (i) vortices go straight across the artificial pinning sites, as sketched in Fig. 6(a); or (ii) vortices kink to follow the artificial pinning sites, as in Figs. 6(b) and (c), consisting on stacks of correlated pancake vortices linked by Josephson strings that lie in the \(ab\)-plane. This second possibility seems more likely taking into account (1) the high anisotropy \(\gamma_{ab}\), (2) the limited \(c\)-axis correlations implied by the scaling analysis, and (3) recent experiments using scanning Hall probe microscopy in highly underdoped YBCO [25], in which flux bundles with less than one superconducting flux quantum \(\phi_0\) were identified as this type of stacked structure. Note that the Josephson strings are necessary to conserve the in-plane field component, and consequently their number must increase with the angle as sketched in Figs. 6(b) and (c). Nevertheless, for any of the two possibilities considered above, the vortex line fraction pinned within the irradiated regions decreases with increasing tilt. One expects therefore that the pinning energy due to the commensurability between the flux lattice and the periodic array decreases with increasing tilt angles. This allows for an understanding of the decrease of \(C_n\) [Fig. 4(f)] as \(B\) is increased and the matching condition \(B \cos \theta = nB_\phi\) is satisfied at higher angles [Figs. 4(d) and (e)]. Note that \(\epsilon_n\) also decreases with increasing applied magnetic field [Fig. 4(g)], which accounts for the observation that the \(n\) resistance minima in \(R(\theta)\) become much narrower as they shift towards \(\theta = \pm 90^\circ\). This could be interpreted as the matching of the flux lattice to the artificial pinning array becoming more critical as it is achieved at increasingly tilted fields. However, this conclusion should not be pushed too far, since that behavior can otherwise be understood by considering that, as the minima shift towards \(\theta = \pm 90^\circ\) in increasing magnetic fields, the out-of-plane field component \(B \cos(\theta)\) grows more rapidly \((d[B \cos(\theta)]/d\theta\) increases). Therefore, the closer to \(\theta = \pm 90^\circ\) a particular minimum sits, the faster we go “in” and “out” of the matching condition when the field is tilted, and the narrower the resistance minima.
Before concluding, some comparisons may be drawn between the present experiments and earlier ones in which different materials and pinning centers were used. *A priori,* the creation of a pinning array of ordered correlated defects should have precipitated the appearance of a *Bose glass* in our samples. However, we have obtained a different result. The key characteristics of our system are: i) the relatively high anisotropy as compared to pristine YBCO; ii) the periodic ordering of the artificial pinning sites; and iii) the presence of strong *random* pinning that coexists with the periodic one. None of these characteristics can explain the observed behavior by itself: i) a *Bose glass* has indeed been observed in more anisotropic superconductors—e.g. Tl$_2$Ba$_2$CaCu$_2$O$_{8+x}$ [23] and Bi$_2$Sr$_2$CaCu$_2$O$_{8+}$ [26]—with columnar defects; ii) a Mott insulator phase (a particular case of the *Bose glass*) has been previously observed in Bi$_2$Sr$_2$CaCu$_2$O$_{8+}$ single crystals with periodic arrays of holes [6]; and finally, iii) Horide *et al.* [12] observed a crossover from vortex glass into *Bose glass* in YBCO thin films in which strong random pinning coexists with c-
4. INTERPLAY BETWEEN INTRINSIC AND ARTIFICIAL ORDERED PINNING

axis correlated pinning induced by BaZrO$_3$ nanorods. From the above, we conclude that it is the concurrence of strong intrinsic random disorder and relatively high anisotropy that precludes the observation of Bose-glass behavior in our samples, even in the presence of a periodic pinning array. If periodic pinning sites created by irradiation could be obtained without increasing the YBCO anisotropy, a crossover from vortex to Bose-glass behavior should be observed.

4.5 Conclusions:

We have analyzed the glass transition in a YBCO thin film with a square pinning array created by masked O$^+$ irradiation. We have seen that it is strongly modified with respect to the three-dimensional vortex-glass transition usually seen in pristine YBCO. We can rule out a Bose-glass transition (usually observed with correlated disorder) and we have verified that in our system a quasi-two-dimensional vortex-glass stabilizes. We have observed that the field dependence of the glass transition temperature is non-monotonic: $T_g$ is enhanced at the matching fields. The angular magneto resistance shows pronounced minima which are solely governed by the applied field component parallel to the $c$-axis, which indicates that vortex localization does not occur for particular directions of the applied field –as in Bose glass– but for particular in-plane vortex densities. This is due to an increase in the anisotropy of the system during irradiation and the presence of strong intrinsic random pinning. All of the above allows us to conclude that vortices in our system are unusually soft and they can, for instance, wander out of the artificial pinning defects to accommodate other native pinning sources.
References:


4. INTERPLAY BETWEEN INTRINSIC AND ARTIFICIAL ORDERED PINNING


5. Thermal switching of the energy landscape geometry in nanopatterned YBCO thin films

5.1 Introduction
   5.1.1 Geometric frustration in artificial ice
   5.1.2 The vortex ice
   5.1.3 Inter-hole O$^+$ ion damage
5.2 Description of the samples
5.3 Thermal switching of the geometric frustration
   5.3.1 Magneto-resistance measurements
   5.3.2 The Josephson junction array scenario
   5.3.3 The crossover temperature $T_{cr}$
5.4 Determination of the vortex configuration at low $t$
   5.4.1 Critical current $J_C$ measurements
   5.4.2 Vortex lattice energy calculations
   5.4.3 Considerations about disorder
      5.4.3.1 Dependence on the magnetic history
      5.4.3.2 Effect of current annealing protocols
5.5 Study of thermal switching in deformed vortex-ice array geometries
   5.5.1 Description of the samples
   5.5.2 Magneto-resistance measurements
   5.5.3 Evolution with temperature
5.6 Conclusions
In the previous two chapters we have demonstrated that masked ion irradiation produces strong periodic pinning for vortices. In this chapter, a regular but inhomogeneous pinning array is analyzed: a geometrically frustrated array that stabilizes a vortex ice at low temperatures. The geometry of the array can be changed through thermal switching, turning geometric frustration reversibly on and off using temperature as a control knob.

To determine the array geometry and its evolution we will measure magneto-resistance at different temperatures. Subsequently, critical current measurements will be performed to conclude if the vortex ice distribution occurs at low temperatures. Various field and current protocols will be used as well to probe the ice for disorder. Finally, a more detailed analysis of the thermal switching mechanism will be given using deformed vortex-ice arrays.
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

5.1 Introduction

Magnetic flux quanta can be used as a model to study the problem of an ensemble of repulsive particles on a potential-energy landscape. This is a problem common to many physical systems and has been studied in multiple artificial playgrounds. However, these usually involve fixed energy landscapes, thereby impeding in situ investigations of the particles’ collective response to controlled changes of the landscape geometry. The ability to create modifiable energy landscapes can be exploited to overcome this limitation. Here, we will see how we can collectively and reversibly switch the energy landscape for every flux quanta using a single macroscopic parameter: the temperature. We will illustrate this by studying the problem of geometric frustration.

5.1.1 Geometric frustration in artificial ice

Geometric frustration is found in systems in which particles are bound by the energy landscape in such a way that it is not possible to simultaneously minimize all their pairwise interactions. This situation is found in various natural systems, the archetypes being water ice [2] and pyrochlore crystals [3–5] –in which an analogous “spin ice” is formed. As it is shown in Fig. 1, in the case of water ice oxygen and hydrogen ions organize forming a tetrahedral structure, with two energetically equivalent positions for hydrogen: inside (1) or outside (2) the tetrahedron. Since hydrogen ions repel each other, they will seek to minimize their pairwise interactions by being as far away as possible. However, the geometrical frustration in the system impedes this for all hydrogen ions: placing one outside of a tetrahedron to distance it from others in it, leaves the hydrogen ion in the inside of the adjacent tetrahedron. The best hydrogen ions can do is to distribute themselves two inside and two outside in what is known as the “ice rule” (as it is shown in Fig. 1).
In order to study frustration in a controlled environment, artificial ice [6–10] is used. So far, most of the experiments done focus on artificial “spin ice”. In one of the earliest reports, Wang et al. [6] used a 2D array of elongated permalloy islands grown by molecular beam epitaxy, that behave as nanomagnets with bi-stable single-domain magnetization. A sketch of one of the arrays used –corresponding to the square ice geometry– can be seen in Fig. 2(a). For these islands the pairwise dipolar interactions are minimized when the magnetizations are head to tail. Geometric frustration makes this possible for only a fraction of the nanomagnet pairs, while the rest are forced to adopt more energetic head-to-head or tail-to-tail configurations. The system reaches its lowest-energy state, as in water ice, when the magnetizations order according to the “ice rule” at the array vertices [as in Fig. 2(a)]: two pointing in (red), two pointing out (blue). This system has the advantage of allowing direct imaging using magnetic force microscopy (AFM sensitive to magnetic forces) or magnetic circular dichroism (XMCD), which makes it easy to snapshot the distribution of magnetizations. However, because of the relative weakness of the dipolar interactions, and because the energy barriers between the two possible states of each nanomagnet may be higher than the thermal energy [11], extended spin ice systems often show disorder – that is, a mixture of ice-rule-obeying vertices and many others having higher-energy configurations. Much effort has been put into achieving the ground state with nanomagnets, recently fulfilled by Zhang et al. [12] through thermal annealing protocols. Nevertheless, the statistics of disobeying vertices [6,9,13], the spatial
correlations between them, and the relaxation towards the ordered ground state [14–16] bear much fundamental interest.

5.1.2 The vortex ice

Flux quanta in superconductors offer an interesting playground to study those issues. Libál et al. [17] proposed using elongated double-well pinning sites [Fig. 2(b)] in which a vortex would sit at one of the two minima, depending on the interaction with nearby vortices. This creates an analogous structure to the nanomagnet where, instead of having a magnetization point one or the other way, the vortex sits in one or the other minima. In this manner, a “vortex ice” becomes a promising artificial ice system. The molecular dynamic simulations performed in this work showed that the strength of the inter-vortex interactions should drive the system into the ground state more readily than in nanomagnet arrays. Furthermore, flux quanta can be set in motion by electrical currents, which provides a new mechanism for “annealing” the ice defects, and enables dynamical studies. Experiments along this line have been reported recently by Latimer et al. [18] using holes in MoGe films, a low-$T_C$ superconductor.
To stabilize a square vortex ice, vortex pinning arrays with the geometry shown in Fig. 3(a) are used. The elongated double-well pinning sites are achieved placing two holes close together. For certain applied fields, and if the pinning energy is sufficiently high (potential-energy wells sufficiently deep), all of the magnetic flux quanta will locate in pinning sites, as the simulations by Libál et al. [17] showed. With two vortices per unit cell –yellow circles in Fig. 3(a)– the analogy with the artificial spin ice is fully achieved, since there would be one vortex per pair of holes. The geometrical frustration is evident: there is no possible arrangement in which all of the vortices are located in pinning sites and the inter-vortex distance is constant. Therefore, it is not possible to simultaneously minimize all of the pairwise interactions. As in other ice systems, the lowest-energy configuration is achieved when vortices obey the “two-in/two-out” ice rule shown in Fig. 3(a).

In this experiment we take advantage of the way the $T_C$ modulation in the system is created, and demonstrate an interesting opportunity, non-existent in other artificial ice realizations: the geometric frustration can be switched on and off via temperature changes, which provides a handle to reversibly transform a periodic square vortex lattice into vortex ice –i.e. to “freeze” and “thaw” artificial ice. This is what we call thermal switching. The array geometry can be taken from that sketched in Fig. 3(a) to that in Fig. 3(b) by simply changing the temperature. Note that in Fig. 3(b) the closest pinning sites have merged due to the disappearance of the energy barrier, and the array is no longer geometrically frustrated: vortices can arrange into a periodic square lattice in which the inter-vortex distance is constant.
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

Figure 3: Schematic diagrams of the energy landscape in masked ion irradiated YBCO at a) low temperatures, in which each pair of holes has a barrier in between, and b) high temperatures, where the barrier disappears and the holes merge forming ovals. In each case the pink lines show first neighbor distances.

5.1.3 Inter-hole O⁺ ion damage

In the masked O⁺ ion irradiation process used to define the pinning arrays, point defects are produced in the exposed areas that create a $T_C$ modulation in the material. However, it is necessary to take into account the fact that a non-negligible amount of damage is also created between pinning sites during the irradiation process. This damage depends on the distance between the holes in the mask. In Fig. 4(a) simulations of the local critical temperature are shown for three different inter-hole distances. Note how as holes are set closer the inter-hole local $T_C$ becomes lower. This is reflected in a lighter red or yellower color between the blue areas.

This fact presents interesting consequences. Let us consider a mask with two holes separated by a distance short enough so that meaningful damage is produced in between them. Then the inter-hole maximum local critical temperature $T_{1i}$ would be smaller than the film one $T_{c0}$. As presented in Fig. 4(b), if we set the temperature $T$ in
our system so that $T_i < T < T_{c0}$ the holes in the array would merge, since the inter-hole region would remain in the normal state. As we show in the following, this effect is responsible for the thermal switching observed, and can be exploited to create modifiable energy landscapes using temperature as a control knob.

![Figure 4: a) Simulated local critical temperature maps for three pairs of holes separated 90, 120 and 150 nm, from left to right. Color scale expressed in K. b) Sketch of the local critical temperature profile for the case of unequal distances between holes. S stands for superconducting state and N for normal state.](image)

5.2 Description of the samples

Vortex pinning arrays were fabricated using hole masks (diameter $\phi=70$ nm) as that shown in Fig. 5. The arrays are characterized by two distances, $L_1$ and $L_2$, which yield a characteristic field $B_{\phi} = \phi_0 / (L_1 + 2L_2 \cos 45)^2$ – the magnetic field that induces one flux quantum per array unit cell (dashed square in Fig. 5). Note that $L_1$ controls the height of the barrier between both minima in the elongated structure of Fig. 3(a), via the mechanism in Fig. 4.
Three different arrays have been created using masks with different inter-hole distances: \( L_2 = 120 \text{ nm} \) was left constant while \( L_1 \) was varied from 60 nm to 150 nm, in 30 nm increments, a rather small variation. The local \( T_C \) maps calculated for each sample are shown in Figs. 6(a)-(d). Note how, in the same way it was shown in Fig. 4(a), the arrays present different local critical temperatures in between the pinning sites depending on the distance \( L_1 \).

Table 1 presents for each sample the array parameters \( L_1 \) and \( L_2 \), the characteristic field \( B_\phi \), the onset \( (T_{\text{onset}}) \) and zero-resistance \( (T_{\text{co}}) \) critical temperatures (see Fig. 7),
and the normal-state resistivity at the transition onset $\rho_N$. Note that the denser the array, the higher $\rho_N$, as expected since in a larger fraction of the sample superconductivity is depressed. The different $T_{\text{Conset}}$ and $T_{\text{C0}}$ are understood in the following manner: $T_{\text{Conset}}$ corresponds to the local critical temperature at the center of the array unit cell [dashed square in Fig. 5], while $T_{\text{C0}}$ corresponds to the temperature at which the areas between the holes separated by the greater distance —$L_1$ or $L_2$— become superconducting, thus allowing the percolation of supercurrents across the sample. The virgin samples (prior to irradiation) presented $T_{\text{C0}} = 87K$ and $T_{\text{Conset}} = 92K$.

![Figure 7: Resistance versus temperature curve for the 120-120 sample, using $J=5\cdot10^5$ A·m$^{-2}$. The temperature at the onset of the superconducting transition $T_{\text{Conset}}$ and at the end $T_{\text{C0}}$ are indicated.](image)

<table>
<thead>
<tr>
<th>$L_1$ (nm)</th>
<th>$L_2$ (nm)</th>
<th>$B_\phi$ (T)</th>
<th>$T_{\text{Conset}}$ (K)</th>
<th>$T_{\text{C0}}$ (K)</th>
<th>$\rho_N$ ($10^6$ Ohms·m)</th>
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<td>75</td>
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</table>

Table 1: Table 1: Array parameters of each sample, characteristic field $B_\phi$, onset temperature of the superconducting transition $T_{\text{Conset}}$, zero-resistance temperature $T_{\text{C0}}$, and normal resistance at the onset $\rho_N$. 82
5.3 Thermal switching of the geometric frustration

5.3.1 Magneto-resistance measurements

In contrast to nanomagnet artificial ice, it is difficult to study our system through imaging experiments, due to the fact that imaging vortices is rather complicated, especially in high-$T_C$ superconductors. Therefore, we investigated the above arrays using magneto-transport experiments. Figs. 8(a)-(d) show the magneto-resistance data for the series of samples.

We first analyze the highest temperature measurements done at $t = T/T_{C0} \sim 1$ (orange lines) in the Ohmic regime above the glass transition temperature (see 4.1.1). For 60-120 [Fig. 8(a)] $L_f=60$ nm is shorter than the hole diameter, and therefore the array holes join by pairs, forming ovals in the mask. These form a square pinning array with oval pinning sites that is not geometrically frustrated. Therefore, this array will be useful for comparison. This sample exhibits deep minima for even values of $|n|$ (the deepest for $|n|=2$), and much shallower for odd $|n|$. The arrays in Figs. 8(b), (c) and (d) present geometric frustration and look very similar to each other. However
they behave very differently at high-$t$, as the orange curves show distinctively unequal features. Notably, sample 90-120 [Fig. 8(b)] behaves similarly to sample 60-120 [Fig. 8(a)], which as discussed above does not present geometrical frustration. For 120-120 [Fig. 8(c)], the deepest minima are for $|n|=4$. In contrast, for 150-120 [Fig. 8(d)] the most pronounced minima are for $|n|=1$, and $|n|=4$ are indeed the weakest.

When $t$ is decreased, a gradual smoothing of the curves is observed for all of the samples due to the progressive increase in the intrinsic random pinning strength. However, the temperature effects on the relative intensity of the resistance minima are very different for each sample. In Figs. 8(a) and (c) no temperature evolution is observed: respectively, the minima at $|n|=2$ and $|n|=4$ remain the most pronounced. Contrarily—and surprisingly— in Figs. 8(b) and (d) $|n|=4$ become the most intense at low $t$ (magenta/blue). Thus, despite the great differences at high $t$, the data in Figs. 8(b), (c) and (d) become very similar at low $t$. Conversely, the curves in Figs. 8(a) and (b) become very different upon cooling, in spite of their similarity at high $t$. Such a dramatic change of the field-matching effects upon cooling implies that the geometry of the vortex energy landscape strongly depends on temperature. As it is explained below, the reason is that, for two of the samples [Figs. 8(b) and (d)], the effective number of artificial pinning sites changes with temperature. Note that the different behaviors cannot be ascribed to different properties of the samples prior to irradiation (e.g. native pinning), since the arrays are defined in the same YBCO film. Also, the divergence of the superconducting coherence length close to $T_C$ cannot account for those thermal effects. In order to demonstrate this last point we estimated the coherence length in our samples, using $\xi = \xi_0 \cdot (1 - T/T_{C\text{onset}})^{-0.5}$. We employ $T_{C\text{onset}}$ and not $T_{C0}$ because $\xi$ should diverge at $T_{C\text{onset}}$ and not at $T_{C0}$. For $\xi_0$, we used the bulk YBCO $\xi_{ab} \sim 1.5$ nm. In all samples, the variation with temperature keeps $\xi$ within the range $(2, 4.8)$ nm, a change insignificant as compared to the constriction between holes $d = \{20, 50\}$ nm.

We proceed now to explain the series of minima for each of the samples. For 60-120 [Fig. 8(a)], the understanding is straightforward: the ovals form a square array of pinning sites, for which field-matching effects have been studied in the two previous
chapters. The deepest minima in Fig. 8(a) (|n|=2) correspond to one flux quantum per pinning site, |n|=4 correspond to two quanta per site, etc. The shallower dips at |n|=1 and 3 correspond, as expected, to the fractional matching (non-integer number of flux quanta per pinning site, on average). For |n|=1, for example, vortices leave half of the sites empty, and are expected to form a checkerboard pattern [10] [see sketch for n=1 in Fig. 8(a)].

We turn now to Fig. 8(b). At high t (orange curve), the resemblance with Fig. 8(a) implies that the vortex energy landscape is similar in both samples. This suggests that in 90-120 each pair of pinning sites separated by $L_1=90$ nm is effectively “merged” into a single oval-shaped pinning site [see sketch in Fig. 8(b)], yielding an array with no geometric frustration, as in 60-120. Fig. 6(b) shows that the local $T_C$ depression in between pinning sites is stronger along $L_1$ (shortest) than along $L_2$ (longest). Therefore, one expects that, at a sufficiently high $t$, the space between the closest holes (along $L_1$) becomes normal, while that across the furthest (along $L_2$) is still superconducting. Thus, at high enough $t$ the holes merge forming “ovals” across $L_1$, while a barrier between pinning sites still exists across $L_2$. This effect is different but concomitant with the thermal smoothing of the energy landscape due to vortex fluctuations [22], which also favors the merging of the closest holes by smoothing the barrier out. Thus, at high $t$ the scenario is similar to that for 60-120 [Fig. 6(a)]. This allows explaining the high $t$ data in Fig. 8(b) in terms of the square-array behavior used for Fig. 8(a).

The high $t$ (orange) curve for 150-120 [Fig. 8(d)] can be understood using the same arguments. In this case $L_2<L_1$, so the four closest holes (around the array vertices) “merge” at high $t$, producing a square array of large clustered pinning sites [see sketch in Fig. 8(d)]. This explains why the most pronounced minima occur for |n|=1 (one flux quantum per clustered site), while higher order minima gradually become shallower, as expected for square arrays. The situation is different for the sample in Fig. 8(c). Since $L_1=L_2$, the local $T_C$ and the vortex energy barriers between pinning sites are identical across both directions [see Fig. 6(c)]. Thus, a temperature-dependent landscape geometry and an evolution of the matching effects analogous to that in Figs. 8(b),(d) are not anticipated, and indeed not observed [Fig. 8(c)].
5.3.2 The Josephson junction array scenario

Alternatively, one could explain the minima in the magneto-resistance (especially at high temperatures) using a different model: the superconducting Josephson junction array [19]. According to this view, the constrictions between holes would be narrow enough to be considered Josephson junctions, that is, two superconducting regions separated by one of degraded superconductivity. In Fig. 9(a), one sees how a pinning site would be surrounded by four Josephson junctions (represented by a blue X). If a vortex is placed in the pinning site, the superconducting screening currents would flow through the Josephson junctions, provoking a phase difference $\Delta \varphi$ in the superconducting wave function across each junction. This increases the energy of the system due to the fact that it is proportional to $-\cos \Delta \varphi$ [20]. In the case that there is one vortex per pinning site [Fig. 9(b)], opposite screening currents cancel the phase differences in each junction and the system’s energy is greatly reduced. This means that when a Lorentz force is applied, vortex hopping to adjacent cells is prevented because it would create a much higher energy configuration. According to Baturina et al. [21], this description is valid for ratios of the constriction and the superconducting coherence length $d/\xi \sim 4 - 8$. In our samples, this ratio is $d/\xi \sim 7 - 10$, right in the upper limit. Nevertheless, in the Josephson junction scenario, a merging of the closest holes at high temperatures is also necessary to explain the evolution of the field-matching effects. Therefore, the conclusions to which we arrive here remain the same regardless of the description chosen, so we will continue using the previously employed one.
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

5.3.3 The crossover temperature $T_{cr}$

According to our analysis of the magneto-resistance measurements, there should be a crossover temperature below which the pinning sites in 90-120 and 150-120 should “unmerge”, so the geometry of the energy landscape switches into one similar to that in 120-120. The fact that the lowest $t$ curves in Figs. 8(b), (c) and (d) show matching effects only for $|n|=2$ and $|n|=4$, the latter being the most intense, evidences that the
energy landscapes become similar. The crossover temperature, which we note $T_{cr}$, is the temperature below which a series of $R(B)$ curves measured at decreasing temperatures show deeper minima for $|n|=4$ than for $|n|=2$. According to the “merging” sites scenario, the crossover temperature $T_{cr}$ should scale with the expected local critical temperature in between closest sites $T_{inter}^{sim}$, since the latter determines the temperature at which the space between them becomes superconducting. $T_{inter}^{sim}$ can be obtained from the simulations in Fig. 6, and its reduced magnitude $t_{inter}^{sim} = T_{inter}^{sim}/T_{c0}^{sim}$ (with $T_{c0}^{sim}$ the zero-resistance temperature expected from the simulations, see inset of Fig. 10) correlates with the reduced crossover temperature $t_{cr} = T_{cr}/T_{c0}$ ($T_{c0}$ as determined from the R(T) curves), as shown in Fig. 10.

In summary, for the samples in which $L_1 \neq L_2$ the temperature effects on the energy landscape switch off the geometric frustration, effectively yielding square arrays of big clustered pinning sites that result from the “merging” of closest pinning sites in the original array layout. At sufficiently low temperatures the original, geometrically frustrated layout is recovered. Then the magneto-resistance series of minima is similar to that of the 120-120 sample. We show in what follows that the low-temperature matching effects for 90-120 and 150-120, as well as for 120-120 at any temperature, can be understood considering the ordering of the vortices according to a square vortex ice for $|n|=2$.

5.4 Determination of the vortex configuration at low $t$

5.4.1 Critical current $J_c$ measurements

The low $t$ curves in Fig. 8(b)-(d) at $|n|=2$, for which the field and energy landscape are the adequate for obtaining vortex ice, display shallow matching effects. Since these measurements were done below the glass transition where the critical depinning current $J_c$ is defined, we can use $J_c$ vs. the applied magnetic field $B$ [Fig. 11(a)] to better quantify and compare the intensity of the matching effects between different
samples. $J_C(B)$ was obtained from isothermal E-J characteristics measured in increasing magnetic fields. A criterion $E_C=2.5 \cdot 10^{-2}$ V/m was used to determine $J_C$. At the measurement temperature $t \sim 0.8$, the three samples are below their $t_{cp}$ and below the glass transition, and showed similar zero-field critical current $J_C(0) \sim 10^9$ A·m$^{-2}$. The matching effects in $J_C(B)$ parallel those observed in the low $t$ (blue) magneto-resistance curves [Figs. 8(b)-(d)]. For the three samples [see Fig. 11(a)], the most pronounced peak corresponds to $n=4$. Sample 90-120 shows a clear peak for $n=2$, but this peak is much weaker for 120-120, and barely noticeable for 150-120. This trend is shown in Fig. 11(b), which displays the relative $J_C$ enhancement at matching—defined as the ratio between the peak height $\Delta J_C$ and the background variation of $J_C$ in the experimental window, i.e. $\Delta J_C/[J_C(0) - J_C(7B_\phi)]$. A higher relative critical current enhancement means a higher stability of the vortex configuration [23]. The bottom line from Fig. 11(b) is that i) both for $n=2$ and $n=4$, the relative $J_C$ enhancement steadily weakens as the array parameter $L_1$ increases, and ii) this happens faster for $n=2$ than for $n=4$.

Figure 11: a) Critical current $J_C$ vs. applied field $B$ for different arrays $L_1$-$L_2$ (in nm, see legend). The critical current enhancement $\Delta J_C$ as a result of the commensurability between the vortex lattice and the pinning array is indicated. b) Relative $J_C$ enhancement at matching as a function of $L_1$. 
5.4.2 Vortex lattice energy calculations

The matching effects for \( n=2 \) and \( n=4 \) cannot be explained by assuming an Abrikosov (triangular) vortex-lattice: the latter and the pinning array are incommensurate [see Figs. 13(c),(f)] in any direction. To determine this we obtained the maximum ratio of pinned vortices \( \nu_p \), running calculations in Matlab. The azimuthal orientation of the lattice with respect to the array axis is rotated every degree in the range \( 0^\circ \leq \theta \leq 90^\circ \), and the number of vortices located in pinning sites is counted for each \( \theta \). Due to the arrays’ four-fold symmetry, every possible orientation is considered in this manner.

In Fig. 12(a), as an example, the pinning ratio over the 120-120 array is plotted as a function of \( \theta \) for different vortex lattice geometries and densities around \( n=2 \). Only the square lattice for \( n=2 \) presents a sharp rise at \( \theta = 45^\circ \), while the triangular lattice is incommensurate in every orientation. The same incommensurability of the triangular lattice is obtained for other samples and vortex densities, as shown in Figs. 12(b) and (c), in which the maximum \( \nu_p \) for the three samples at \( n=2 \) and \( n=4 \) is presented considering different vortex lattice geometries.

![Figure 12: a) Ratio of pinning as a function of the orientation of the vortex lattice on the pinning array for various lattice geometries on the 120-120 sample. b) and c) Maximum pinning ratio for each vortex lattice geometry and sample for the field corresponding to b) \( n=2 \) and c) \( n=4 \).](image)

The magneto-resistance minima at integer numbers of vortices per unit cell imply that the vortex lattice deforms in order to adapt to the pinning array. For this, the pinning energy due to localization in array sites \( \Delta E_{pin} \) has to be greater than \( \Delta E_{el} \), the elastic energy increase due to the lattice deformation [23]. There are two possible matching scenarios. If the available pinning energy per site \( \epsilon_p \) is sufficient, every vortex will
locate in an artificial pinning site [Figs. 13(e),(h)]. This fully-matched distribution maximizes $\Delta E_{pin}$ at the expense of $\Delta E_{el}$. Otherwise, if $\epsilon_p$ is not high enough, a more regular distribution will be formed in order to minimize $\Delta E_{el}$, at the expense of placing only a fraction of the vortices $\nu_p$ in pinning sites and the remainders in interstitial positions. Finding the flux distribution that allows this is trivial, given the four-fold symmetry of the array. It corresponds to a square vortex-lattice [Fig. 13(d),(g)], which presents the lowest possible $\Delta E_{el}$ (excepting the triangular lattice [19]) and the highest possible $\nu_p$ (excepting the fully-matched distribution), since only one vortex per unit cell is taken out of its pinning site (for $n=2$) to obtain a square lattice. We can ascertain which of the flux distributions—fully-matched or square lattice—occurs in the present experiments via the dependence of the matching effects intensity on $L_1$. For this, we use $\Delta E_{pin} = \epsilon_p \cdot (\nu_p^m - \nu_p^{tr})$. For a triangular vortex lattice, the pinning ratio $\nu_p^{tr} \in [0.11,0.17]$, for a square one $\nu_p^m \in [0.25,0.5]$ and for the fully-matched distribution $\nu_p^m=1$ [see Figs. 12(b),(c)].

The elastic energy increase due to the deformation of the vortex lattice from its triangular (natural) geometry into any other matching configuration (square lattice or
square ice) is calculated as $\Delta E_{el} = E_{int}^{tr} - E_{int}^{m}$, where the two subtracted terms are the interaction energy between vortices for the triangular (tr) and matching (m) geometries. In the high-$\kappa=\lambda/\xi$ limit (that is, strongly type-II), $E_{int} = \phi_0^2 / 4\pi \lambda^2 \mu_0 \sum_j K_0 \left( r_{0j}/\lambda \right)$ [19,24], with $\lambda$ the temperature dependent penetration depth, $\mu_0$ the magnetic permeability of the vacuum, $r_{0j}$ the position of the $j$-th vortex, and $K_0$ the zeroth-order modified Bessel function of the second kind. In the calculations, arrays $50\lambda \times 50\lambda$ in size were used [$\lambda = (\lambda_0^2/d) \cdot (1 - t^4)^{-1/2}$ is the temperature dependent penetration depth, $\lambda_0=150$ nm and $d=50$ nm the thickness of the film]. In this manner, $\Delta E_{el}$ is calculated as a function of $L_1$.

We define $\varepsilon_p^c = \Delta E_{el} / \left( v_p^m - v_p^{tr} \right)$ as the critical pinning energy above which a given geometry is energetically favourable. The lower $\varepsilon_p^c$, the more stable the vortex configuration and the stronger the expected $J_c$ enhancement [23]. Therefore, we can compare the $L_1$ dependence of $\varepsilon_p^c$, to determine the actual vortex lattice geometry at low $t$. For a square lattice [Fig. 13(a)], $\varepsilon_p^c$ decreases with increasing $L_1$ and at a higher rate for $n=4$ than for $n=2$ (this is because the elastic energy of a regular vortex distribution decreases when the lattice density decreases). From this, one expects a strengthening of the relative $J_c$ enhancement for increasing $L_1$, which should be more pronounced for $n=4$ than for $n=2$. Experimentally, we observe just the opposite [Fig. 11(b)]: the relative $J_c$ critical current enhancement weakens for increasing $L_1$, and it does so faster for $n=2$ than for $n=4$. This is exactly what is expected from the evolution of $\varepsilon_p^c$ for the fully-matched geometry [Fig. 13(b)]. A qualitative understanding for this is that, the longer $L_1$, the larger the unit cell area, and the higher the elastic energy cost of “emptying” the interior of the unit cell in order to place the vortices on its peripheral pinning sites. In summary, the energy balance considerations and the dependence of the matching effects intensity on $L_1$ rule out the possibility of a square lattice and support a fully-matched vortex distribution—in particular, a square vortex-ice for $n=2$. 

92
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

5.4.3 Considerations about disorder

A final question is whether the vortex ice presents sizable disorder: for example, a significant fraction of i) vertices disobeying the “two-in/two-out” ice rule (see Fig. 14) as seen in some of the nanomagnet arrays, and/or ii) unit cells with irregular flux distributions due to the local presence of strong native pinning centers [25]. Note that some disorder would not prevent the observation of matching effects –as long as many cells across the array do present the square-ice matching geometry– but it would change their intensity [26]. The question is then whether the weakness of the n=2 effects relative to n=4 might reflect the existence of disorder.

![Figure 14: Examples of vortex configurations with ice-rule disobeying vertices (marked by a yellow circle).](image)

5.4.3.1 Dependence on the magnetic history

The vortex ice simulations performed by Libál et al. [17] suggested that a sizable amount of ice-rule disobeying vertices should lead to hysteretic magneto-transport and memory effects. Moreover, the presence of array cells with irregular flux distributions should also lead to this type of behavior, as it was the case in experiments [26] with quasiperiodic vortex lattices and a number of other works [27–29].

In order to look for the eventual presence of memory effects, we followed a twofold approach: i) we compared magneto-transport measurements done after field-cooling across the superconducting transition with measurements done after zero-field-cooling, and with measurements done after having cycled the magnetic field once
below $T_C$; and ii) we compared magneto-transport measurements performed before and after an “annealing” of the vortex lattice done via current cycles or similar (as suggested by Libál et al. [17] and/or following Ref. [27]). Some examples are shown below.

Figs. 15(a),(b) show magneto-resistance measurements, respectively for the 120-120 and 150-120 samples. The samples were field-cooled across the superconducting transition in an applied field $B/B_\phi = n = 2$ with no electrical current injected. Once the desired temperature was reached, a R(B) curve was measured cycling the field from $n=2$ to $n=6$, then to $n=-6$ and finally back to $n=6$. The currents used in A·m$^{-2}$ are: $J_{120-120} = 10^8, 2.75 \cdot 10^9$; $J_{150-120} = 1.5 \cdot 10^8, 1.5 \cdot 10^9$ (high $t$, low $t$). No magnetic hysteresis is observed and, in particular, the resistance value at $n=2$ is independent of the magnetic history.

![Figure 15: a), b) Magneto-resistance measurements after following the field cycling described in the text. c), d) V(I) curves measured following the protocol described in the text.](image)

Figs. 15(c),(d) show sets of V(I) curves, for the samples 120-120 and 150-120 respectively. Each set contains 4 V(I) [labeled (1) to (4)], all of them measured at
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

\[ \frac{B}{B_0} = n = 2 \]. (1) is measured right after field-cooling across \( T_C \) in a field \( n = 2 \). (2) is measured subsequently to (1). (3) is measured after cycling the field to \( n = -6 \) and then back to \( n = 2 \). (4) is measured after cycling the field to \( n = 6 \) and then back to \( n = 2 \). The V(I) are independent of the current and magnetic history.

5.4.3.2 Effect of current annealing protocols

We also performed magneto-transport measurements before and after an “annealing” of the vortex lattice. The “annealing” is intended to eliminate disorder, if present, as shown by the theoretical simulations of Libál et al. [17], which would result in a further enhancement of the critical current (or accordingly in a deeper magneto-resistance minima) at the matching condition. As we show below, we find that the matching effects were insensitive to them in all cases.

For the annealing experiments, the protocol is as follows: samples are zero-field-cooled after which a field corresponding to a density of \( n = 2 \) vortices per unit cell is applied, and a first V(I) is measured. Immediately after, an annealing is performed, followed by a second V(I) measurement. Various annealing procedures were tested. All of them are based on the injection of electrical currents that shake the vortex lattice and favor vortex ordering into the “ice rule” ground state. The annealing starts with a high switching current that induces flux flow, after which the current is gradually reduced so that vortices shake only locally around their pinning sites, to finally reach a static situation when the current is further reduced below the critical one.

The different types of annealing used are sketched in Fig. 16. Figs. 16(a),(b) correspond to procedures in which a single electrical current of decreasing amplitude is injected. The sketch in Fig. 16(d) is for the annealing in which two perpendicular currents \( I_1 \) and \( I_2 \) are injected in the cross-shaped bridge shown on the SEM image. This annealing procedure was suggested by Libál et al. [17]. By changing \( I_1 \) and \( I_2 \) a current is obtained in the center of the bridge whose direction rotates, polarity switches and intensity is decreased in each step. Finally, one last procedure consists on measuring the \( dc \) magneto-resistance \( R(B) \) while an \( ac \) excitation is simultaneously applied to the vortices, looking in particular whether this changes the
magneto-resistance minima at $|n|=2$. This is done in two ways. For the first, Fig. 16(c), a small antenna is placed close to the sample to shine radio-frequency radiation which shakes vortices. Powers up to 20 dBm and up to 1 GHz frequencies are used. For the second, a $dc$ and a sinusoidal $ac$ current are simultaneously injected using the feature offered by a Keithley 6221 current source.

![Figure 16: Schematic representations of the different types of annealing procedures performed, intended to eliminate disorder in the flux distribution: a) Rectangular and b) sinusoidal decreasing current with changing polarity; c) gigahertz radiofrequency radiation; d) rotating and polarity switching current. e) SEM image of the microbridge indicating the current injection in d).](image)

All of the methods are performed for an extensive set of conditions, including many different temperatures above and below the irreversibility line, various amplitudes of $ac$ versus $dc$ current, etc. Fig. 17 shows a few examples. In all cases, the measurements were insensitive to the annealing. In particular, V(I) curves are identical before and after annealing, and in R(B) curves we see the same relative depth of the minima (and specifically the same shallow minima at $|n|=2$) regardless of the absence or presence of RF and $ac$ excitation. Also, note the results in Fig. 17 are identical to those in Fig. 8.

We concluded from this that the relative weakness of the matching effects at $|n|=2$ is unlikely linked to the presence of disorder –it could be simply explained by considering that at this vortex density half of the pinning sites are empty and the same reasoning as for fractional matching in square arrays applies (see above). In any case,
the proper quantification and statistics of disorder in this artificial ice system demands imaging techniques [25,30–32] that would have to be adapted to visualize the static vortex distribution in our high-\( T_C \) samples.

5.5 Study of thermal switching in deformed vortex-ice array geometries

5.5.1 Description of the samples

As we have shown in our vortex-ice pinning arrays, the existence of different distances between pinning sites provokes the merging of the closest sites at sufficient high temperatures. To investigate this phenomenon more thoroughly, we have
fabricated different masks in which the original vortex-ice symmetry is deformed. This has been done in such a way that the periodicity is not lost [Figs. 18(a)-(d)].

![Images of masks used to create deformed arrays.](image)

**Figure 18:** Scanning electron microscopy images of the masks used to create the deformed arrays. In (a) the distance which is reduced between the holes $L$ is indicated as well as the scale bar for 200 nm. For each array the unit cell is marked with a white dashed square.

We have started with the 120-120 array [Fig. 18(a)], in which all distances are equal, and gradually deformed the arrays so only the holes separated by the distance marked as $L$ are brought closer together. In Fig. 18(d), one can see the charge accumulation effects in the electronic lithography process (see 2.3.2) due to the proximity of holes. In the SEM image of each array the array unit cell has been overlaid as a white dashed square. In every case, the vortex density for which there is one vortex per pinning site is $|n|=4$.

### 5.5.2 Magneto-resistance measurements

To characterize each array, we have measured magneto-resistance curves, shown in Fig. 19(a)-(d). These have been measured at a similar reduced temperature $t \sim 1$ and current density $5 \cdot 10^7$ Am$^{-2}$. From top to bottom, the curves correspond respectively to
the arrays depicted in Figs. 18(a)-(d). As expected, the resistance minima differ greatly between arrays, especially regarding the matching effects for $|n|=3$ and $|n|=4$. As the holes separated by the distance $L$ become closer, we see an evolution in the relative intensity: a strengthening of the matching effect at $|n|=3$ and a weakening of that at $|n|=4$. Also, in the top curve [Fig. 19(a)] corresponding to the symmetric array [Fig. 18(a)], there are minima present for $|n|=8$ –two vortices per pinning site. On the other hand, in the bottom curve [Fig. 19(d)] corresponding to the most deformed sample [Fig. 18(d)], there is a strong matching effect for $|n|=6$ and $|n|=8$ is not present.

All the features in the magneto-resistance curves can be explained according to the thermal switching mechanism. To the right of each curve in Fig. 19, there is a
schematic representation of the vortex distribution for $|n|=3$ and $|n|=4$. The strengthening of the matching effects at $|n|=3$ and $|n|=6$ means that increasingly, the number of effective pinning sites per unit cell is changing from four to three, that is, the merging between the closest pinning sites becomes greater. This fact is represented in the schematics with an increasingly opaque white oval over the merging sites. When the holes are closer the depression of the local $T_C$ in between the sites is greater, and thus yields a smaller barrier in between. Therefore, for the sample in Fig. 18(d), the array unit cell has just 3 pinning sites: four regular pinning halves, plus one half of the bottom right merged site, plus two quarters of the remaining merged sites. This causes stronger matching effects for $|n|=3$, and the appearance of minima at $|n|=6$—two vortices per pinning site in this case.

### 5.5.3 Evolution with temperature

According to the thermal switching mechanism, temperature should have clear effects on the energy landscape, as well as on the field-matching effects. To illustrate this, we have measured the magneto-resistance of the sample in Fig. 18(c) at decreasing temperatures. These are represented in Fig. 20. The same way that it happened in the vortex-ice arrays where $L_1 \neq L_2$, we see a smoothing of the field-matching effects, but also a different evolution in their relative intensity. Here at high $t$ [Fig. 20(a)], the minima for $|n|=3$ and $|n|=4$ present the same intensity. Nevertheless, as we lower the temperature the intensity of the minima for $|n|=4$ becomes greater than that for $|n|=3$. At the lowest temperatures [Fig. 20(d)], the only clearly visible minima is that for $|n|=4$. This evolution indicates an “unmerging” of the closest sites due to the fact that the space in between the closest holes becomes superconducting. This is reflected in the sketches to the right of the magneto-resistance curves, where the ovals gradually disappear as the temperature is lowered.
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

Figure 20: (a)-(c) Evolution with temperature of the magneto-resistance for the array shown in Fig. 18(c). The magnetic field density that corresponds to a vortex density \( n=3 \) is indicated (dashed vertical line). To the right of each curve the vortex distribution for \( n=3 \) and \( n=4 \) is shown, as well as the merging degree (opacity of ovals) of the closest holes.

Once more it has become clear that minute variations of the energy landscape geometry has dramatic effects in the vortex lattice behavior and the observed field-matching effects in the magneto-resistance curves. Moreover, we have seen how temperature constitutes a precise control knob to tune the pinning array geometry.

5.6 Conclusions

In this chapter, we have demonstrated a way to create switchable energy landscapes, which allows the study of reconfiguration and relaxation upon abrupt changes in the landscape geometry. In particular, we have obtained artificial vortex ice that can be frozen or thawed using temperature as a control knob. This is provoked by the thermal switching of the array’s geometry from a geometrically frustrated one to a periodic square array. Critical current measurements and energy considerations reveal
that the stability of the vortex ice is reduced with increasing $L_1$, and the absence of changes in the vortex response after performing annealing protocols suggest a largely ordered vortex ice in the ground state.

Thermal switching opens some new interesting perspectives not available in other ice systems. For instance, one can create different sublattices with competing order (ice vs. square lattice) whose dominance can be tuned with temperature. This provides a handle to controllably introduce defects in the vortex ice and opens the door to studies on properties such as the correlation length, confinement and dynamics of ice defects.

Beyond the artificial ice problem, the present realization opens opportunities in other areas. This work directly impacts the field of vortex control in nanostructured superconductors, a research field in which many efforts are devoted to manipulate vortices in order to obtain functionality, like fluxtronic devices. One could conceive devices in which the landscape geometry, and thus the functionality, can be tuned with temperature. Moreover, this idea can be readily extended to any physical system (e.g. ferromagnets and strongly correlated oxides) in which an ordering temperature exists (e.g. Curie temperature or Mott transition temperature) that can be changed via any form of nano-patterning.
5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY

References:


5. THERMAL SWITCHING OF THE ENERGY LANDSCAPE GEOMETRY


6. Asymmetric potential-energy wells, Bean-Livingston barriers and the ratchet effect:

6.1 Introduction

6.1.1 The ratchet effect

6.1.2 Bean-Livingston barriers

6.2 Description of the samples

6.3 Experimental results

6.3.1 Ratchet effect dependence on current

6.3.2 Ratchet effect dependence on the applied magnetic field

6.4 Discussion

6.5 Conclusions
In this chapter we will study the influence of asymmetric pinning sites on the vortex dynamics. The presence of asymmetries should yield a ratchet effect in vortex motion measurable through a rectified voltage. We will first measure the dependence of the ratchet signal on the injected current, and then on the applied magnetic field. Another sample without asymmetric pinning sites will be used for comparison and to establish the contribution of asymmetric Bean-Livingston barriers to the measured rectified voltage.
6.1 Introduction

6.1.1 The ratchet effect

A dynamical system that under stimulus exhibits a preference for motion or evolution in one direction rather than in the opposite is a very interesting goal technologically speaking. This effect is widely present in everyday life in a variety of mechanical, electrical and biological settings. The most widely recognized example is the ratchet, a device consisting on a rotatable wheel and a pawl. The wheel’s exterior is carved to display angled teeth in such a way that the pawl glides over the teeth when the wheel is rotated in one direction, but prevents the rotation in the other [see Fig. 1(a)]. In other words, the pawl sees a different potential-energy barrier depending on the direction of motion.

![Diagram of a ratchet mechanism. The wheel can only turn in one direction.](image)

![Potential-energy landscape seen by vortices in asymmetric pinning sites.](image)

The ratchet effect has acquired great importance lately due to the discovery that it plays an important role in various, seemingly unrelated, areas. For instance, the movement inside cellular cytoplasm of different proteins [1], certain financial investment strategies [2] or colloidal particle motion [3], follow the model of a biased Brownian motor [4], which can be viewed as out-of-equilibrium fluctuations in an asymmetric potential.
The idea of employing asymmetric potential-energy barriers was adapted in superconductors using asymmetric pinning sites for vortices (e.g. triangles) that would present a smaller energy barrier for vortex motion in one direction (towards the vertex) than in the opposite (towards the base). Therefore, if vortices were to be driven by an alternating force perpendicular to the anisotropy axis of the pinning sites with a temporal average $<\vec{F}> = 0$, vortices would move more slowly when the drive points to the high-energy barrier than when pointing in the opposite direction [see Fig. 1(b)]. This rectification of an alternating force could be employed [5] in multiple applications like vortex diodes or in SQUID magnetometers. For the latter, the measurement resolution is limited by the noise induced by trapped vortices in the two superconducting Josephson junctions. Introducing a pinning lattice with asymmetric sites would force vortices out of the SQUID, “cleaning” it of trapped vortices.

The ratchet effect was experimentally used with superconducting flux quanta for the first time in a low-$T_c$ superconductor: a Nb thin film grown on top of a rectangular array of triangular Ni dots [6]. The asymmetric shape of the triangles caused the measurement of a $dc$ voltage when driving vortices with a symmetric $ac$ current [Fig. 2(a)]. The measured voltage is proportional to the average vortex velocity, thus, vortices moved faster in one direction than in the opposite, or in other words, a net velocity was developed (red arrows in Fig. 2) in the direction of the lower-energy barrier. Surprisingly, when the current was decreased and only the weakly pinned interstitial vortices (in blue) were moving, these saw an inversed potential-energy landscape, as sketched in Fig. 2(b), that prompted the appearance of a net velocity in the opposite direction (blue arrows).
6. ASYMMETRIC POTENTIAL-ENERGY WELLS

Figure 2: a) Average vortex speed as a function of the applied Lorentz force in a Nb sample with Ni triangular pinning sites. b) SEM image of the triangular dots and vortex distribution overlaid. The blue triangle represents the landscape geometry for the blue vortices. Adapted from Ref. [6]

In the case of high-\(T_C\) superconductors, the ratchet effect has been more difficult to observe. This is due to the high thermal fluctuations and strong intrinsic pinning present in these systems that complicate the production of effective artificial ordered pinning sites. Wördenweber et al. [7] measured a vortex ratchet effect due to guided vortex motion using rows of antidots in YBCO oriented at a certain angle with respect to the measurement bridge. Ooi et al. [8] managed to produce in BSCCO, which presents low intrinsic pinning, a ratchet effect using triangular pinning defects (see inset of Fig. 3). As it is shown in Fig. 3, they observed multiple and rather sharp reversals of the ratchet effect when sweeping the magnetic field. These reversals occurred when reaching the matching field and their sharpness suggested that neither the pinning sites’ asymmetry nor interactions among vortices were enough to explain the effect.
6.1.2 Bean-Livingston barriers

When considering the ratchet effect, it is important to take into account other possible sources of asymmetry in the system. In a superconducting thin film, when a magnetic field is applied, vortices enter or exit the sample through the surfaces in the edges. In doing so they overcome a surface barrier called Bean-Livingston barriers [9]. These originate from basically two contributions: the attractive force that the vortex feels towards the surface and the repulsive force exerted by the magnetic field outside the surface. The combination of both produces, considering an ideal surface, a vortex line energy dependence $E$ on the distance from the surface $x$ as the one shown in Fig. 4(a). The barrier to flux entrance does not disappear until a field $H_S > H_{C1}$. In the case of flux escape, the barrier exists down to the lowest fields.
Figure 4: a) Vortex line energy $E$ dependence on the distance from the surface $x$, for $\lambda = 10\xi$ and various applied fields. Adapted from Ref. [9]. b) Longitudinal and transversal rectification voltages as a function of temperature measured in a Pb strip. Inset: sketch of the experimental configurations. Adapted from Ref. [10].

The Bean-Livingston barriers are heavily dependent on the surface’s geometry and roughness [11]. If a sample presents different surfaces at opposite edges it produces asymmetric Bean-Livingston barriers. Therefore, vortices would see an asymmetric potential energy landscape, which gives rise to a ratchet effect. This was the case, for instance, in the experiments of Pryadun et al. [10], in which a dc voltage was measured when applying an ac current in unpatterned Pb and Nb films [see Fig. 4(b)]. Despite following a fabrication process that a priori creates equal surfaces, the transport measurement bridge did not present symmetric Bean-Livingston barriers. The asymmetry strongly depended on the magnetic field, temperature and ac drive. This remarks the importance of considering the effect of surface barriers when performing ratchet-effect experiments.

The study of the ratchet effect in our samples is readily available since we have demonstrated how defects created through masked O$^+$ ion irradiation behave as strong pinning sites. In this chapter we will investigate the rectification of asymmetric pinning sites, drawing comparisons with another sample of symmetric pinning sites to monitor the contribution of the Bean-Livingston barriers to the ratchet effect.
6.2 Description of the samples

So far, in our experiments we have used irradiation masks that contained patterns made out of circular holes, a symmetric shape. For this experiment, we have designed two masks with sites shaped as the Eiffel tower. In the first one the towers form a square lattice of 200 nm spacing [see design in Fig. 5(a)]. The second one is of similar geometry except there are two symmetric blocks: half of the towers point one way and the other half the other way [see Fig. 5(b)], the inversion occurring at the middle of the bridge. A scanning electron microscopy image of one of the masks can be seen in Fig. 5(c). One readily notices that, unfortunately, the shape of the towers was lost and yielded a rounded triangle. This is due to the dose accumulation during the electron beam lithography process.

![Design of the tower masks](image)

**Figure 5:** a) and b) Design of the tower masks used for masked O⁺ ion irradiation. c) SEM image of the resulting mask.

A third sample with a square array of circular holes separated by 180 nm was used for comparison. The three samples were obtained from 50 nm-thick YBCO films and present similar magneto-resistance curves (see Fig. 6). The field-matching effects [shallower in the sample with the two blocks of towers in Fig. 5(b)] show that the triangular defects also behave as good pinning sites.
6.3 Experimental results

The characterization of the ratchet effect in our samples was done measuring the rectified voltage, that is, the difference between voltages measured injecting positive and negative currents. To avoid noise or heating effects, each measurement was corrected by the zero current voltage measured immediately after it, and a protocol was established so each measurement had the same previous current history. The
protocol is as follows: first, a positive current $J$ is applied during 100 ms and switched off for another 100 ms with no measurement taken; then the previous step is repeated taking voltage measurements $V^+$ and $V^+_{\text{zero}}$ respectively; finally, this is repeated again but applying a negative current $-J$ and obtaining $V^-$ and $V^-_{\text{zero}}$. The rectified voltage is then calculated as:

$$V_{\text{rect}} = (V^+ - V^+_{\text{zero}}) - (V^- - V^-_{\text{zero}})$$

Since at high currents the zero current intervals between measurements might not be enough to reestablish the thermal equilibrium in the sample, it is necessary to also perform the above protocol measuring first with a negative current. Then the average of the rectified voltages $\bar{V}_{\text{rect}}$ is taken.

### 6.3.1 Ratchet effect dependence on current

In Fig. 7, the average rectified voltage $\bar{V}_{\text{rect}}$ is shown as a function of the applied current for the three samples. Fig. 7(a) corresponds to the sample with triangular sites, Fig. 7(b) to the sample with the two blocks of triangles, and Fig. 7(c) to the one with circular pinning sites. The red lines represent the average rectified voltage for the positive first matching field and the black lines for the negative one. This means that in every case there is one vortex per pinning site. The green lines represent the $V(I)$ curve for the positive current and field. The measurement temperature is around $0.9T_C$ in every case. Qualitatively, similar curves were obtained for other temperatures above and below the glass transition temperature.
In the sample with triangular pinning sites [Fig. 7(a)] an increase with current of $\langle \tilde{V}_{\text{rect}} \rangle$ (with the same sign as the applied field) is seen. For the sample with two blocks of inverted triangles [Fig. 7(b)], an increase of $\langle \tilde{V}_{\text{rect}} \rangle$ and then a reversal is seen. Surprisingly, for the sample with circular pinning sites [Fig. 7(c)] a similar ratchet effect to the one in Fig. 7(a) is seen too. The onset of the observed rectified signals seem to correlate with the critical current from the $V(I)$ (green lines). In all cases, the behavior of the positive and negative-field rectified voltage is symmetric with respect to the horizontal axis. As shown in Fig. 8, when the magnetic field is inversed, vortex velocity is also reversed while the vortex-motion-generated electrical field stays
invariant. Therefore, a same sign voltage is measured even if vortices go from moving away from the barrier [Fig. 8(a)] to moving towards it [Fig. 8(b)]. This means that upon inversion of the magnetic field the rectified signal should change sign. The symmetric behavior of $\vec{V}_{\text{rect}}$ with field confirms that the measured signal corresponds to a ratchet effect. Vortices are seeing different potential-energy barriers when they move in one direction than in the opposite. The reversal of the ratchet signal seen in Fig. 7(b) indicates that, at high currents, the asymmetry of the potential-energy barriers seen by vortices is inversed.

![Figure 8: Schematic diagram of the result on vortex motion and electric field of inverting the applied magnetic field. In a) vortices move away from the barrier while in b) the opposite occurs. This causes the sign change in the rectified voltage when inverting the magnetic field.](image)

**6.3.2 Ratchet effect dependence on the applied magnetic field**

We have measured the evolution of the rectified voltage with the applied magnetic field. The same measurement protocol as the one previously described is applied for each value of the magnetic field. In Fig. 9 the average rectified voltage $\vec{V}_{\text{rect}}$ is represented by the black curve for negative field and red for positive. The green curve is the positive current $V(B)$ curve. Vertical dashed lines are used to indicate the
6. ASYMMETRIC POTENTIAL-ENERGY WELLS

position of the matching fields. Figs. 9(a) corresponds to the sample with triangular pinning sites, Fig. 9(b) to the sample with the two blocks of triangles, and Fig. 9(c) corresponds to the sample with circular pinning sites.

Figure 9: Average rectified voltage (left axis) and voltage for positive current (right axis) as a function of the applied magnetic field. a) corresponds to the sample with triangles; b) to the one with two blocks of triangles; and c) to the sample with circular pinning sites.

The average rectified voltage $\bar{V}_{rect}$ in Figs. 9(a)-(c) is antisymmetric with respect to the vertical axis. This is as expected due to the sign reversal of the ratchet signal that should occur when inversing the magnetic field. $\bar{V}_{rect}$ is of around 1% the value of the measured signal. However, several abrupt changes in $\bar{V}_{rect}$ can be seen, especially, in Fig. 9(b). The black arrows indicate the changes that occur right before the vertical dashed lines, that is, before the matching condition is met. One can see that these are repeated in every sample and they sometimes produce a reversal of the
ratchet effect at the matching field, as in Fig. 9(b). We will discuss these results below.

6.4 Discussion

The $\vec{V}_{\text{rect}}(I)$ curves in Fig. 7(c) show that a ratchet effect occurs in the sample with perfectly symmetrical circles. This suggests that the weak asymmetry of the triangular pinning sites is not responsible for the observed rectification. Nevertheless, the signal inversion seen only for the sample with two blocks of triangles might hint at a possible role of the pinning sites geometry. In any case, it is clear that asymmetric Bean-Livingston barriers are necessary to explain the ratchet signal. As previously commented in the introduction, these can arise from asymmetric edges in the measurement bridge. Even if during fabrication it was not intended to obtain asymmetric edges, these can be at the origin of the ratchet effect here obtained.

The observed dependence of $\vec{V}_{\text{rect}}$ with the magnetic field in Fig. 9 is reminiscent of the one measured by Ooi et al. [8] with triangular holes in BSCCO single crystals [Fig. 3]: rather similar abrupt changes in the ratchet signal are seen in Fig. 9, and in Fig. 9(b) a ratchet reversal occurs too. In their work, Ooi et al. considered that these might be a consequence of a “net potential acting on the whole vortex system”. The results obtained here using symmetric circular holes suggest that this effect might be a consequence of asymmetric Bean-Livingston barriers. It is difficult, however, to think of a mechanism involving surface energy barriers that would provoke such a sudden and drastic change around the matching fields. Further experiments are required to elucidate the connection between a slowing vortex lattice and the asymmetry change of Bean-Livingston barriers.
6.5 Conclusions

We have studied two YBCO samples patterned with asymmetric pinning sites, measuring the ratchet signal’s dependence on the applied current and magnetic field. From the comparison with a sample with circular pinning sites it is clear that the contribution to the ratchet signal of the asymmetric pinning sites is very low. This can be explained by taking into account the fact that the dose accumulation during the electron beam lithography of the irradiation masks deformed the original shape into rounded triangles.

The results obtained here suggest that asymmetric Bean-Livingston barriers on the sample edges are the main driver of the ratchet signal and its abrupt changes at the matching fields. It is not clear, however, how these barriers are modified when changing the vortex density in the pinning array.
References:

7. General conclusions

In this PhD thesis, we investigate artificial ordered pinning in high-temperature superconductors. The high-critical-temperature superconductor used is YBa$_2$Cu$_3$O$_{7-\delta}$, in the form of thin films grown by pulsed laser deposition. Samples have been structurally characterized using various techniques. X-ray diffraction was used to obtain information such as the crystalline structure, epitaxial growth, sample roughness or thickness. Scanning electron microscopy and atomic force microscopy allowed imaging the sample’s surface in order to ensure its smoothness and to determine the presence of outgrowths that cause strong native vortex pinning. Finally, the film’s critical temperature $T_C$ was quickly determined by gradually submerging an electrical transport probe in a nitrogen Dewar.

For magneto-transport experiments, the central technique used in this thesis, microbridges were fabricated in the films using photolithography and ion beam etching. In each bridge, the energy landscape for vortices was engineered via masked O$^+$ ion irradiation, a combination of electron-beam lithography and ion irradiation. Via e-beam lithography, a resist mask is obtained to only expose nanometric areas of the superconductor to the irradiation, so the local critical temperature is depressed in the exposed areas. These areas will remain in the normal state acting as very effective pinning sites for vortices.

First of all we perform a characterization of the artificial periodic pinning produced irradiating through a square hole array mask. Marked field-matching effects were
observed in the form of sudden drops of the resistance for vortex densities equal or multiples of the pinning sites density. We found that the strength of these commensurability effects depends on temperature and the vortex velocity in a different way as it was earlier reported for low-$T_C$ superconductors.

A study of the vortex matter on our samples and its vortex phase diagram was performed. A thermodynamic phase transition was observed between a vortex liquid and a glass. However, the presence of the artificial periodic pinning concomitant with strong native random pinning modified the usual 3-dimensional vortex-glass transition seen in pristine YBCO films, into a quasi-2D vortex-glass transition. Different angular measurements helped rule also a Bose-glass transition out and revealed an increased anisotropy of the superconductor and the unusually soft nature of the vortices.

The ability to modulate the local critical temperature in the superconductor at a nanometric scale allowed us to use vortices in a nanostructured type-II superconductor to investigate general problems common to multiple physical systems, such as artificial ice, frustration, and thermal effects. In particular, we demonstrate the ability to create pinning landscape geometries that are switchable using temperature as a control knob. We start with a geometrically frustrated pinning array that, at low temperatures, stabilizes a vortex ice without sizeable disorder and close to its ground state. Nevertheless, at high temperatures the energy landscape geometry switches and becomes a square periodic array. This allows us to stabilize and destabilize artificial ice. Beyond this example, our approach is interesting because it can be exported to other physical systems where an ordering temperature exists and can be modulated, with multiple implications in the design of functional devices.

Finally, an ion irradiation mask was designed with asymmetric pinning sites in order to obtain an energy landscape in which vortex motion would be easier in one direction than in the opposite. This constitutes a so-called vortex ratchet or diode. An asymmetric motion of vortices was observed through the measurement of a rectified voltage. The comparison with a sample with symmetrical pinning sites revealed the observed ratchet effect was due to asymmetric Bean-Livingston barriers in opposite
7. GENERAL CONCLUSIONS

edges of the micro-bridge. The dependence of the rectified voltage with the applied magnetic field also showed that a drop in the ratchet signal is produced when approaching the array’s matching field.
List of Publications


