

**Research area:** Solid state physics (electronic properties, magnetism, basic quantum materials)

Discipline: Material Sciences Specific Discipline: Knowledge based multifunctional materials Keywords: Skyrmions, superconducting vortices, 2D van der Waals ferromagnet Type of Scientific Proposal: Fundamental Research Industrial Relevant: No

**Instrument:** BL29 - BOREAS (Beamline for Resonan Absorption and Scattering) **End Station:** MARES (Soft X-Ray Resonant Scattering)

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Proposal Definition: New Continuation Of: --Resubmission Of: --

Proposals whose experimental reports are related to this one:

Photon energy range: 700-1000 eV Spot size: 100 x 100 Micrometers x Micrometers (FWHM) Horizontal x Vertical Total number of shifts requested: 15 Comments about the number of visits: 2

Unacceptable Dates: Preferred Dates:

# **EXPERIMENT CONTEXT**

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CELLS - ALBA Synchrotron Carrer de la Llum 2-26 08290 Cerdanyola del Vallès - Barcelona Spain http://www.cells.es



Why is SR needed to solve the proposed scientific case? We need to do the measurements at the elemental edges, which are uniquely available at the synchrotron sources.

Have you already performed synchrotron radiation experiments? Yes If yes, specify where: Indus I, Indus II, Petra III, ESRF, Elettra, ALBA

Have you already used synchrotron radiation for this project? No

# PROPOSAL FRAMEWORK

Do you have any "Plan estatal/nacional" research grants? No

Do you have any "Horizon 2020" and/or EC related research grants? No

Do you have any other research grants? No

Is the proposal in collaboration with an industrial group? No

Does this proposal have any industrial involvement or sponsorship? No

Is the proposal a significant part of PhD thesis? No

Is the main proposer an early career researcher (obtained Ph.D. within the last ten years)? Yes

List of most relevant publications or patent references of all co-proposers with data taken in ALBA https://doi.org/10.1021/acsanm.4c04068

Have you or any co-proposer published from previous submitted IDs? If yes, consult evaluated proposals to know the IDs of previous calls and confirm IDs with publications No

Can you guarantee the required resources to perform this experiment according to the experimental plan as described in the experimental plan? Yes

Abstract: Mapping the phase diagram of the superconducting vortices, so-called Abrikosov vortices, by subjecting a superconductor to chiral magnetic fields with varying topology will be a very effective engineering approach to tailor the Abrikosov vortex states, which is unexplored. We wanted to explore the Abrikosov vortex phase diagram in a type-II superconductor Bi2Sr2CaCu2O8 with a contact interface of a complex skyrmion host material Fe3GeTe2 through X-ray Fourier transform holography.

Description: See attached pdf file

Description PDF (available at the end of this document): BOREASProposal.pdf



# PROCESSES (SAMPLE OPERATIONS) PERFORMED BY USERS DURING THE EXPERIMENT AT ALBA

No such things are required.

EQUIPMENT AND PRODUCTS PROVIDED BY ALBA DURING THE EXPERIMENT

Sample environment: flat plate

Access to Alba Laboratories

Biology lab NO Chemistry lab NO Will you need the Ar-filled glove box? NO High pressure lab NO Materials Science lab NO EM sample preparation room NO ALBA laboratory support required for sample preparation:

EQUIPMENT BROUGHT BY THE USERS DURING THE EXPERIMENT AT ALBA

List of equipment brought by the users: only samples

Equipment description: No such things will be brought.

CHEMICAL PRODUCTS BROUGHT BY THE USERS DURING THE EXPERIMENT AT ALBA

List of chemical products brought by the users: No such things will be brought.

Chemical products description: No such things will be brought.



**NON-BIOLOGICAL SAMPLE 1** 

# Substance formula: Fe3GeTe2, Bi2Sr2CaCu2O8 Brief sample description: Thin flake sample Number of samples: 1 The sample you bring is: None Is there any danger associated with the proposed sample, with any preparation at ALBA or with sample equipment? No If in the two previous questions you identified risks, please detail them along with the measures you will take to handle them: No Sample state: Other **Comment:** solid Quantity: 10g Holder, container and solution for transportation: Yes Sample environment at ALBA: Flat plate Radio-active sample: Not radioactive Further safety information related to the sample itself or any preparation process that will be done at ALBA: It is safe Additional information for technical evaluation: Not applicable Q-range or 2 Theta range: Not applicable Detectors to be used: Yes Other information that may be relevant for the evaluation procedure: yes

# Proposal description (BOREASProposal.pdf)

#### Holographic Imaging of Magnetic Skyrmions Coupled Superconducting Vortices in a Fe3GeTe2/Bi2Sr2CaCu2O8 Heterostructure

1. Scientific background: Magnetic skyrmions are particle-like topological spin textures, that exist in both centrosymmetric and non-centrosymmetric systems. Both theoretically and experimentally it is widely assumed that the Bloch-type skyrmions appear in centrosymmetric systems with magnetic dipolar interaction, while Néel-type skyrmions are favored in non-centrosymmetric systems with magnetocrystalline anisotropy <sup>1,2</sup>. Both the Bloch-type and the Néel-type skyrmions have been observed in the 2D van der Waals (vdW) ferromagnet Fe<sub>3</sub>GeTe<sub>2</sub> (FGT) in single layer thin flake as well as in heterostructures<sup>2-5</sup>. Furthermore, FGT thin flakes have unveiled spin textures with a varying topology such as skyrmioniums, skyrmion bags, and skyrmion sacks<sup>6</sup>, making them distinct from the other vdW systems where spin texture is predominantly confined to the unitary topological charge, namely, skyrmions. The stability and formation of such composite skyrmion states are likely to be influenced by the details of the crystal structure and the Curie temperature ( $T_c$ ), which reaches as high as room temperature in Fe<sub>5</sub>GeTe<sub>2</sub><sup>7</sup>.

On the other hand, in type-II superconductors, the superconducting vortices, so-called Abrikosov vortices, where each vortex carrying a single quantum of magnetic flux, which enables a material to remain superconducting above the primary critical field  $H_I$  up to a second critical field  $H_{II}$ , at which point the vortex density becomes so high that the entire superconducting condensate breaks down (Fig. 1(a))<sup>8</sup>. In between these two critical fields, vortices can order in a variety of phases such as a hexagonal lattice or an amorphous solid depending on the intrinsic properties of the superconductor, the presence of defects, and the influence of edge-states<sup>9,10</sup>. Such Abrikosov vortices have been imaged with a variety of techniques including Lorentz transmission electron microscopy & magnetic force microscopy<sup>11,12</sup>, however, each suffers drawbacks in regards to resolution, acquisition time, and impact upon the vortices, and most are unable to return in-situ information on defects and edge-states. Additionally, it would be very exciting to explore the phase diagram of Abrikosov vortices by subjecting a superconductor to chiral magnetic fields with varying topology, which could be achieved through interfacing the superconductor to a material hosting unitary as well as composite skyrmion phases.

2. Motivation for the proposal: The proposed approach is to utilize the effect of a chiral magnetic field on the Abrikosov vortex states in a contact interface between a type-II superconductor and a skyrmion host material with varying topology through X-ray Fourier transform holography. Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> (BSCCO) is a type-II superconductor, where Abrikosov vortex density has been observed to increase within the two critical field values  $H_I \& H_{II}^{11,12}$ . Now, mapping a phase diagram of such Abrikosov vortices when subjected to a chiral magnetic field with varying topology will be very interesting.

In previous work, we applied X-ray holography on an exfoliated flake of FGT deposited on a previously prepared holographic mask with micron precision and the holographic sample was finalized by focussed ion beam drilling of reference holes through the stacks. The sample was cooled and exposed to magnetic fields. Repeated cooling from above  $T_C$  (~200 K) to the measurement temperature of 130 K resulted in the data shown in Fig. 1(b), which were obtained from a 300 nm thick flake of FGT, capped by 30 nm of hexaboron nitride (*h*-BN). The data were acquired by circular dichroism difference holography at the Fe  $L_3$ -edge. All topological nanodomain structures were readily erased by magnetic fields of 50 mT and subsequent hysteresis measurements yielded box-type loops thus confirming an out-of-plane easy axis. The nanodomain structures obtained at low field cooling as well as the structures close to quenching, where composite skyrmion states have previously been observed<sup>6</sup>, appeared in many different configurations indicating an almost pinning-free sample. We observed a labyrinth-domain pattern in zero field cooling, a mixed skyrmions+labyrinth domain pattern at a -8 mT field cooling, and only isolated skyrmions at a -16 mT field cooling, as shown in Fig. 1(b). It was also observed that the topological textures were stable after removing the field (at zero field condition) as well.

Subjecting BSCCO to different composite topological textures, such as stripe domains, skyrmions, skyrmioniums, skyrmion bags, skyrmion sacks, antiskyrmions, merons, and bimerons in FGT, offers a very effective approach to engineering tailored Abrikosov vortex phase diagrams. This method would allow precise control of the vortex behavior as a function of temperature and varying magnetic topology.

Besides, the Abrikosov vortices can be studied using a variety of techniques including optical Kerr microscopy, Lorentz microscopy, and scanning methods, however, each suffers drawbacks concerning resolution, acquisition time, and impact on the vortices. In this respect also, holography will overcome these limitations and image both magnetic nanodomain textures and the Abrikosov vortices in a non-destructive, non-contact, full-field fashion. Though the Abrikosov Page 5 of 7

### Proposal description (BOREASProposal.pdf)

vortices have never been imaged with soft X-ray techniques, they are conceptually very similar to topological nanodomain structures and can be imaged with X-ray holography. The expected parameters of the vortices are within proven imaging capabilities. In particular vortices in BSCCO are several hundred nanometers across, with cores of a few nanometer diameter, while in the range of 5-300 Oe applied field, the inter-vortex separation is on the order of 0.5-2  $\mu$ m<sup>11,12</sup>.

#### **3.** Experimental plan:

a) Justification of beamline requested: We plan to investigate a Fe<sub>3</sub>GeTe<sub>2</sub>(30nm)/Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub>(100nm) heterostructure, capped with 30 nm thick *h*-BN to prevent the heterostructure from oxidation. We will perform Fourier transform holography in combination with iterative phase retrieval<sup>13</sup> to image the topological spin structures and Abrikosov vortices in our sample at the different magnetic field cooling at 20-80 K temperature ranges. We need thermal cycling to generate different nanodomain structures through field cooling at different field values from above the  $T_C$  (~200 K) of FGT. The method requires circularly polarized, coherent soft X-rays tuned to the Fe & Cu L<sub>3</sub>-edges. To achieve our target resolution (30 nm), we require extremely high coherent flux, which is uniquely available at the BOREAS beamline.

b) Justification of beamtime requested: We speculate that the sample cooling from above  $T_C$  of FGT to the measurement temperature, recording holographic images at two different edges, and heating it to above  $T_C$  will take about 12 hours. Thus, measuring at least three different field cooling values will take about 36 hours. However, we need to perform the experiment at various temperatures in the range of 20-80 K and check the most interesting set of images at some temperatures and fields. We thus, estimate that to obtain desired results, we require 15 shifts (5 days) of beamtime.

c) Samples & safety-related issues: The thin flakes will be prepared at ICMAB-CSIC, Barcelona; this is already underway. The samples are stable and present no hazard. The planned experiments at the BOREAS fulfill all safety requirements.

4. Results expected & impact: X-ray holography at the BOREAS, ALBA will allow us to map the Abrikosov vortices phase diagram in superconducting Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> subjected to the chiral magnetic field with a varying topology such as stripe domain, skyrmion, skyrmioniums, skyrmion bags, skyrmion sacks, antiskyrmion, merons, bimerons, etc by making a contact interface with Fe<sub>3</sub>GeTe<sub>2</sub>. Such correlation is only accessible through synchrotron techniques (element specific) and will pave the way for future studies of the vortex phase diagram across other skyrmion/superconductor interfaces. Furthermore, the experiment will further serve as the basis for future synchrotron-radiation and XFEL-based experiments examining light-induced vortex dynamics (including melting and depinning) and the nature of light-induced superconductivity.



# 5. Figures:

Figure 1. (a) Phase diagram of a type-II superconductor. We took Fig. 1(a) from reference 8. (b) (Experimental) real space view of the spin texture of the FGT system at 130K after field cooling at different field values from above  $T_C$ results in labyrinth domain structure at zero field cooling, a mixed labyrinth+skyrmion phase at -8mT field cooling, and purely skyrmion phase at -16mT field cooling. The field of view is 2 µm in diameter. (Theoretical) real space view of spin texture for FGT in a  $N = 120^2$  system for temperature parameter (T/t) = 0.01, Hund's (Rashba) coupling parameter  $(\lambda/t) = 0.2$ , and easy-axis anisotropy parameter (Au(T)) = 0.1 after different cooling field  $(h_z)$ . The colour bars correspond to the z-component of the magnetization.

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#### 6. References:

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