# Final Report to US Department of Energy

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"Neutron Scattering Studies of Vortex Matter in Type-II Superconductors"

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**Science Goals:**

The proposed program is an experimental study of the fundamental properties of Abrikosov vortex matter in type-II superconductors. Most superconducting materials used in applications such as MRI are type II and their transport properties are determined by the interplay between random pinning, interaction and thermal fluctuation effects in the vortex state. Given the technological importance of these materials, a fundamental understanding of the vortex matter is necessary. The vortex lines in type-II superconductors also form a useful model system for fundamental studies of a number of important issues in condensed matter physics, such as the presence of a symmetry-breaking phase transition in the presence of random pinning. Recent advances in neutron scattering facilities such as the major upgrade of the NIST cold source and the Spallation Neutron Source are providing unprecedented opportunities in addressing some of the longstanding issues in vortex physics. The core component of the proposed program is to use small angle neutron scattering and Bitter decoration experiments to provide the most stringent test of the Bragg glass theory by measuring the structure factor in both the real and reciprocal spaces. The proposed experiments include a neutron reflectometry experiment to measure the precise $Q$-dependence of the structure factor of the vortex lattice in the Bragg glass state. A second set of SANS experiments will be on a shear-strained Nb single crystal for testing a recently proposed theory of the stability of Bragg glass. The objective is to artificially create a set of parallel grain boundaries into a Nb single crystal and use SANS to measure the vortex matter diffraction pattern as a function of the changing angle between the applied magnetic field to the grain boundaries. The **intrinsic merits** of the proposed work are a new fundamental understanding of type-II superconductors on which superconducting technology is based, and a firm understanding of phases and phase transitions in condensed matter systems with random pinning. The **broader impact** of the program includes the training of future generation of neutron scientists, and further development of neutron scattering and complementary techniques for studies of superconducting materials. The graduate and undergraduate students participating in this project will learn the state-of-the-art neutron scattering techniques, acquire a wide range of materials research experiences, and participate in the frontier research of superconductivity. This should best prepare the students for future careers in academia, industry, or government.

**Results Accomplished:**

## 1. Observation of Edge Contaminated Vortex Phase in Nb:

We report the structural observation of a disordered vortex matter phase existing near the edge of a bulk type-II superconductor Nb using a novel position-sensitive neutron diffraction technique. This "edge-contaminated" vortex state was implicated in previous studies using transport techniques and magneto-optics and is postulated to play a significant role in the behavior of vortex dynamics in a wide range of type-II superconductors. In this sample, thermal annealing causes the vortex matter in the interior to reorder implying that the edge-contaminated bulk state is metastable. However, the edge vortex structure remains disordered after repeated thermal annealing indicating the spatial coexistence of a vortex glass with a Bragg glass. This observation resolves outstanding issues concerning the peak effect in type-II superconductors.

The Nb sample is an unpolished as-grown crystal of 99.99% purity. It has the shape of a cylinder with the <111〉 crystallographic direction oriented along its longitudinal axis. From our AC magnetic susceptibility measurements, we sketch the magnetic phase diagram for this sample in Fig. 1(a). We used a frequency of 1070 Hz and $Hac = 13.5$ Oe. The onset of superconductivity occurs at $T = 9.18$ K and has a width of 400 mK. The important sample parameters are: by measurement, the zerofield superconducting transition temperature $Tc = 9.0$ K, and the peak effect $Hp(T = 4.2\text{ K}) = 3850$ Oe; by extrapolation, the zero-temperature critical field values $Hc1 = 960$ Oe, $Hc2 = 6000$ Oe, and $Hc3 = 10000$ Oe. Our thermo-magnetic growth procedures are defined on the phase diagram: FC is cooling the sample in a magnetic field of 1400 Oe and ZFC is cooling the sample in zero magnetic field and then ramping the field to 1400 Oe.

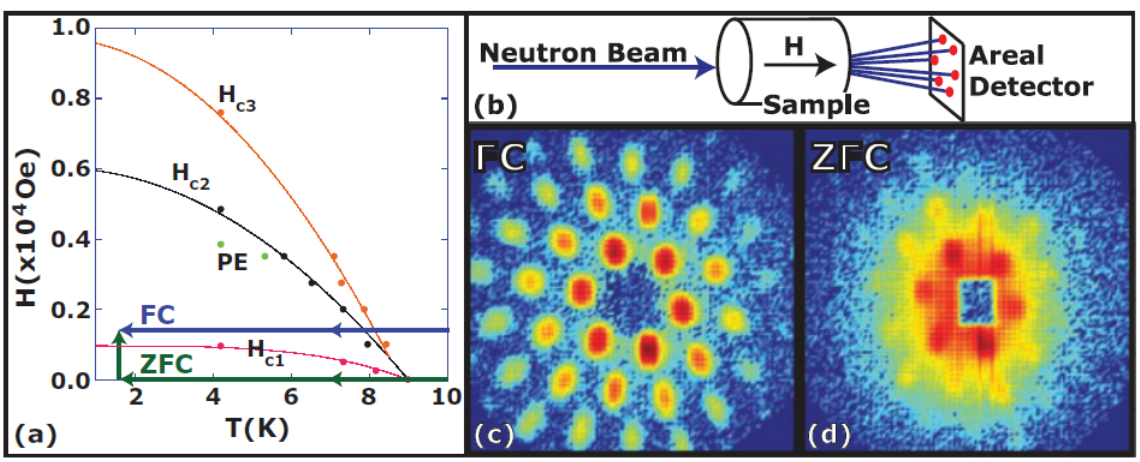


FIG. 1. (Color online) (a) The magnetic field, $H$, versus $T$ phase diagram for the Nb crystal used in this study. PE is remnant of a peak effect. The solid lines are fits to the data. The values of the critical magnetic fields are very similar to a previous Nb crystal but the peak effect line is very different. The growth procedures for FC and ZFC are sketched on the diagram. (b) The scattering geometry for conventional small-angle neutron scattering (SANS). (c,d) SANS images, summed over the rocking angle, for the FC and the ZFC states at $T = 1.5$ K and $H = 1400$ Oe.

The SANS data in Fig. 1 are measured using the V4 instrument at the BER II reactor of the Helmholtz Zentrum Berlin. The scattering geometry for the SANS images is shown in Fig. 1(b). The magnetic field, *H*, is applied parallel to the neutron beam and the cylindrical axis of the sample. The neutronwavelength is $\lambda = 12$ °A and _$\lambda/\lambda = 0.1$. Rectangular guides are used for collimation giving an angular spread of $0.146^{\circ}$. The data are summed over the entire rocking curve (the longitudinal direction of the vortex lines).

The AND/R instrument at NIST Center for Neutron Research (NCNR) allows us to explore the spatial nature of the disorder in the vortex matter. The neutron beam with a width of 0.5 (or 0.25) mm is much smaller than the sample diameter (12.1 mm).We are able to vary the section of the Nb crystal exposed to the neutron beam and measure the Bragg peak for a particular spatial location (the drawn-to-scale sketches of the topography measurements are shown in Figs. 2). However, the increased resolution is at the expense of neutron flux, and prevents us from studying the peak effect and the order-disorder transition directly.  Our scanning neutron diffraction measurements are limited to deep in the Bragg glass portion of the phase diagram. For probing different spatial sections of the vortex matter, we calibrate the center of the Nb crystal ($x = 0.0$ mm) via scans of a neutron absorber (Cd mask) located on the bottom of the sample. The Cd mask allows us to identify the location of two edges of the cylinder in the neutron beam coordinate system.

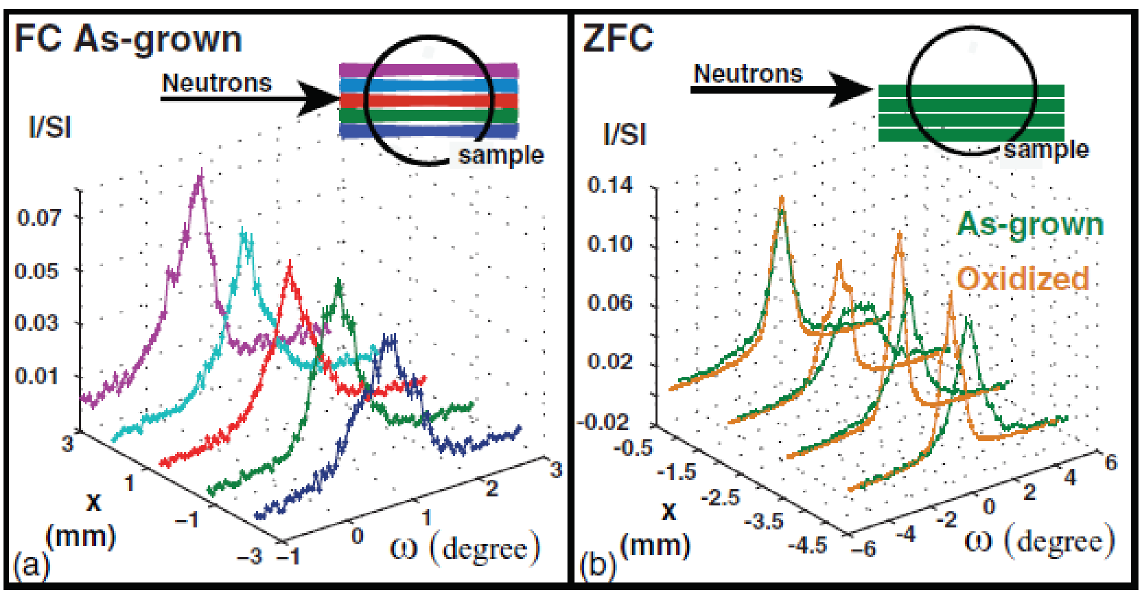


FIG. 2. (Color online) (a, top) The scattering location for the five Bragg peaks from an AFC vortex lattice. (a, main) 3D plot of the AFC Bragg peaks at 4.5 K and 1400 Oe: *I/SI* versus $\omega$ versus sample position, $x$, with a top view of the respective spatial positions in the Nb crystal. The solid lines are guides for the eyes. (b, top) The scattering location in the Nb crystal. Notice here the scattering moves from the center to one edge. (b, main) 3D plot of the ZFC Bragg peaks for the as-grown Nb sample compared to the surface oxidized sample at 4.5 K and 1400 Oe: *I/SI* versus $\omega - \omega^{\circ}$ versus $x$. The peaks are all shifted by the same $\omega^{\circ}$ for each history: The centers of the center Bragg peaks is shifted to zero. The solid lines are guides for the eyes.

In summary, using a slicing neutron diffraction approach, we have explored the vortex matter in a type-II superconductor with a disordered ZFC vortex matter and weak peak effect behavior. Our data offer structural evidence that an edge contamination mechanism is indeed present in systems with a disordered ZFC vortex state. We find resolution-limited radial Bragg peaks for all the thermal-magnetic histories but very different behavior in the azimuthal peaks. A thermal annealing procedure reveals the metastability of the ZFC vortex matter structure and a slight increase in order for the FC system. The surface oxidation of the Nb crystal shows the expected suppression of the magnetic field profile from edge contamination as well as an overall increase in order for all vortex matter histories. The presence of edge contamination in our system could explain the lack of universal behavior for the peak effect in seemingly similar Nb samples. Our results shed light on the phase diagrams for type-II superconductors and may offer a route to the growth of a true Bragg glass state.

## 2. Development of a novel 3D neutron diffraction imaging technique:

By using a thin ribbon-shaped beam, as prepared on the Advanced Neutron Diffractometer/Reflectometer (AND/R) at the NIST Center for Neutron Research (NCNR), we find that it is possible to spatially resolve distinct, locally coherent regions of the vortex lattice within the sample (similar to crystallographic grains, with a small angular mismatch between regions), transverse to the direction of the magnetic field. We made a major improvement on this method, as shown in Fig.3 below, which allows us to probe variations of the in-plane vortex-matter structure factor along the direction of the applied magnetic field. We demonstrate the effectiveness of this method in revealing the splitting of a Bragg peak along the magnetic field direction of the vortex lattice in an Nb single crystal.

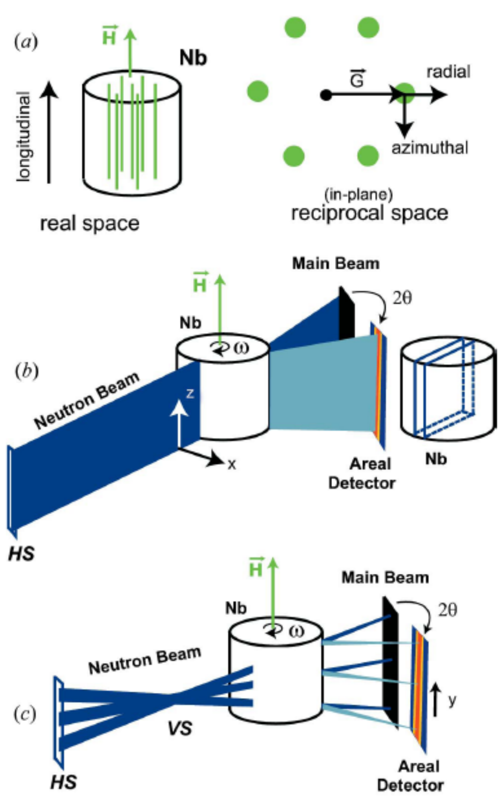


FIG.3. (a) A sketch of our bulk Nb single crystal in a magnetic field with the corresponding structure of an ideal vortex lattice in reciprocal space. (b), (c) The scattering geometry for the neutron diffractometer. The magnetic field is applied parallel to the cylindrical axis (the <111> direction of the Nb crystal). (b) The sample and magnetic field are rotated together by an angle ω with respect to the incident neutron beam. By using horizontal slits, HS, the incident neutron beam is collimated transverse to the flux lines. By shifting the sample along the x direction, different slices of the sample are probed. An areal detector is placed at the angle 2Θ, which is determined by the applied magnetic field. The slice in the Nb crystal shows the part of the sample exposed to neutrons. (c) By adding vertical collimation, VS, after the horizontal slit in the neutron path in conjunction with an areal detector, we can physically distinguish the scattered neutrons from different vertical sections of the sample.

### 3. Observation of screw dislocations in vortex lattice:

The major achievement of our project is that we developed a neutron-di_raction imaging technique which allows us to separate the scattering intensities from various sample locations. We found that by using two sets of perpendicular collimation, it is possible to register the vertical position of the scattering source in the sample along with the in-plane (rocking) scattering intensities. This technique parallels dark-field transmission electron microscopy (TEM) and scanning electron microscopy (SEM) in the sense that we are only detecting a particular Bragg reflection and using a raster technique.

We demonstrate the effectiveness of our method by measuring the vortex matter structure in a type-II superconductor. We observe that the azimuthal Bragg peak from the VL in a Nb crystal splits into two (or more) along the magnetic field direction. The result is shown in Fig.3 below. A simple model, shown in Fig.4, of a three-dimensional (3D) VL is constructed to illustrate that the origin of this phenomenon is the presence of screw dislocations.

Our experiment is carried out on a surface oxidized Nb crystal with the magnetic field applied parallel to the <111> crystallographic direction. An azimuthal scan is performed by placing the position sensitive detector (PSD) at the Bragg angle and rotating the sample and the magnetic field around the field direction. In this measurement, the magnitude of the scattering vector q always equals the reciprocal lattice vector G while the azimuthal angle varies.

Fig. 3 shows the scattering results from three different thermal magnetic histories at the same temperature and magnetic field. The logarithm of the scattering intensity is shown as a color plot depending on the azimuthal angle, and the vertical sample location, z (z = 0 mm corresponds to the bottom of the sample). The scattering intensity from the field cooled (FC) VL displays a distinct Y shape (Fig. 3a).

If the vortex lines are straight throughout the entire sample, the scattering result would be one vertical line. The Y shaped Bragg peak indicates that the vortex lines are not spatially uniform. Near the top of the sample, there are two distinct Bragg peaks. As z is decreased, the centers of the peaks converge. This structure persists in the high-field cooled (HFC) VL (Fig. 3b).

Since the FC and HFC VLs are grown through the same nucleation process, it is not surprising that qualitatively similar structures occur. The Bragg peak from the zero-field cooled (ZFC) VL (Fig. 3c) is disordered with respect to the other states due to the presence of edge contamination on the sample surface. Nevertheless, the scattering intensity still contains the suggestion of a Y shape. The quantitative differences between these three states demonstrate that the VLs are metastable.

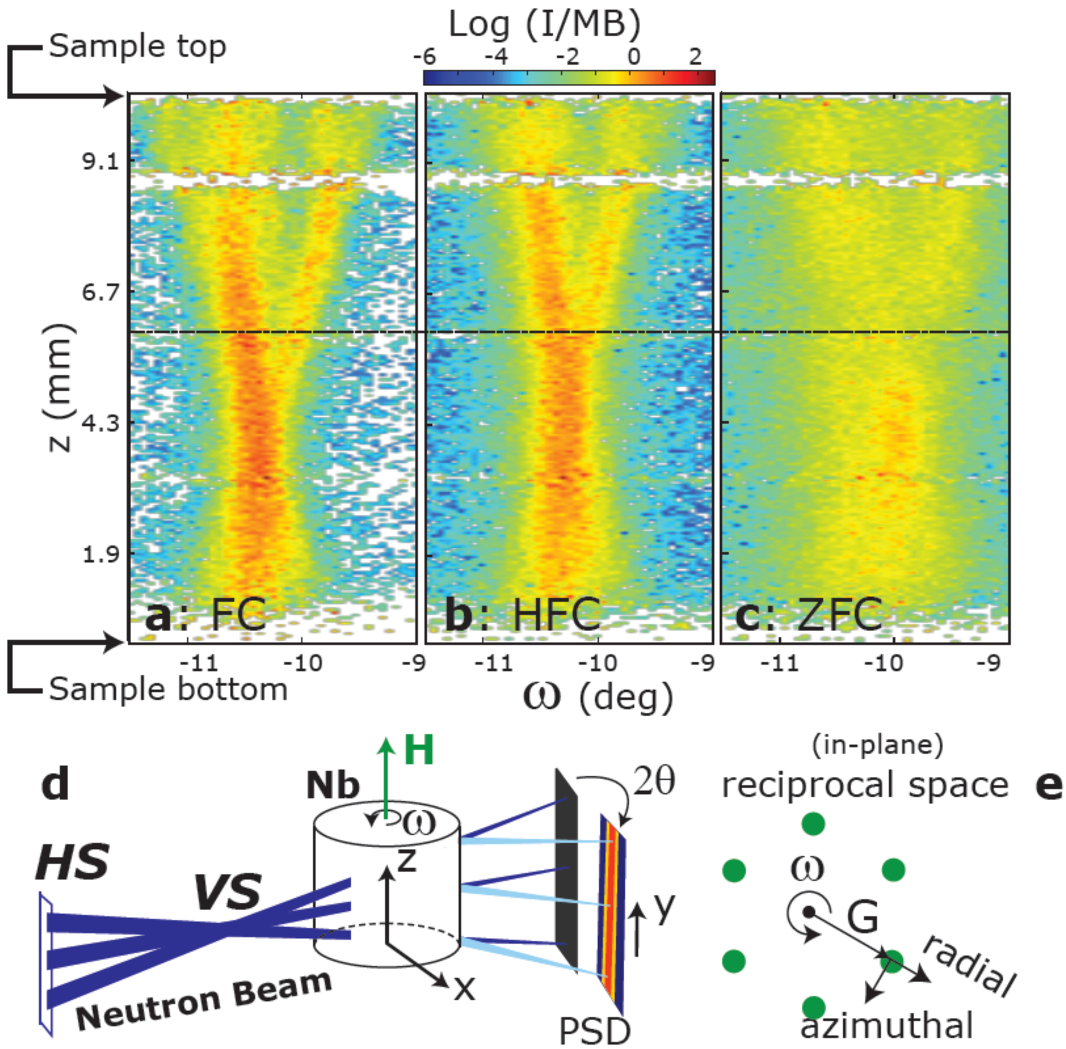


FIG.3. a-c, The neutron scattering intensities from vortex lattice at H = 0.14 T and T = 4.00 K on two dimensional (2D) color plots as functions of the azimuthal angle, and the vertical position in the sample, z. The scattering intensity (I) has been divided by the main neutron beam (MB) intensity and log (I=MB) is plotted on a linear scale. a, The field cooled (FC) VL is formed by cooling the sample through the transition temperature in the presence of the magnetic field (H = 0.14 T). b, The high-field cooled (HFC) VL is formed by cooling the sample at H = 0.40 T and then decreasing the magnetic field to H = 0.14 T. c, The zero-field cooled (ZFC) VL is formed by cooling the sample in zero magnetic field and then ramping up the magnetic field to H = 0.14 T. d, Schematic of the neutron diffraction setup: Two horizontal slits (only one is shown, HS) confine the neutrons to a ribbon-shaped beam. Three vertical slits (only the center one is shown, VS) provide collimation along the cylindrical axis (z direction). e, A reciprocal space sketch of an in-plane VL. The azimuthal direction is defined as perpendicular to the reciprocal lattice vector G while the radial direction is parallel. An azimuthal scan probes the angular orientation of the VL by rotating the sample and the magnetic field around the field direction.

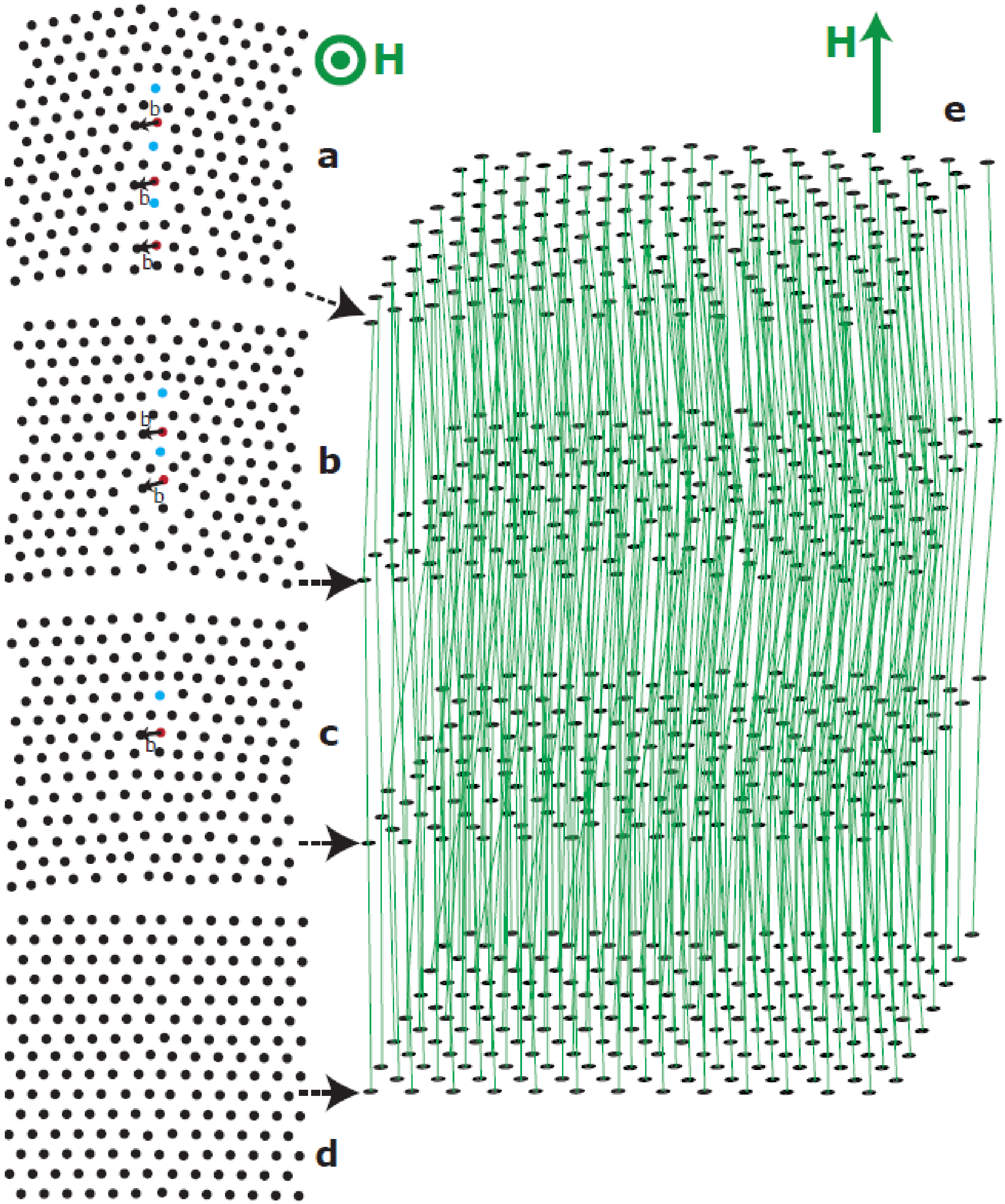


FIG. 4. A diagram of the defects in the VL. a-d, The in-plane VL structures at different vertical locations in the sample. a corresponds to the plane on the sample top and d corresponds to the one in the sample center. The number of vortices is the same in each plane. A pair of disclinations with coordination number of seven (the blue vortex) and five (the red vortex) form a dislocation with its Burgers vector, b, labeled. The array of dislocations separates the VL into two domains with a difference in angular orientation. The number of the Burgers vector decreases as the vertical location is lowered, resulting a diminishing of the angular orientation difference between the two domains. e, An extrapolated 3D plot of the VL based on the in-plane structures in a-d.

**Educational Activities**

This grant provided educational opportunities to our students and postdoctoral associates. Two Ph.D. degrees were earned, by Helen Hanson and Xi Wang, both defended their Ph.D. theses in June 2011. Immediately upon their thesis defenses, both Hanson and Wang were offered professional jobs. Hanson accepted a permanent job with Intel Corporation and Wang accepted a postdoc job at Harvard.

Postdoc Dr. Ivo Dimitrov was able to move onto a position at Brookhaven National Lab. Dr. Jing Shi of Wuhan University was able to spend a sabbatical leave at Brown to learn about neutron physics. Since his return to Wuhan, he has been involved in the neutron condensed matter physics in China.

**Publications:**

[1] Helen A. Hanson, Xi Wang,I.K. Dimitrov, J. Shi, X.S. Ling, B.B. Maranville, C.F. Majkrzak M. Laver, U. Keiderling, M. Russina, "Structural evidence for an edge-contaminated vortex phase in a Nb crystal using neutron diffraction" Phys. Rev. B 84, 014506 (2011).

[2] Xi Wang, Helen A. Hanson, Xinsheng Sean Ling, Charles F. Majkrzak, Brian B. Maranville, "3D Spatially Resolved Neutron Diffraction from a Disordered Vortex Lattice" (arXiv:1102.4776), J. App. Cryst. 44, 414 (2011).

[3] Xi Wang, H. A. Hanson, B. B. Maranville, T. Gnaupel-Herold, C. F. Majkrzak, and X. S. Ling, “Observation of Screw Dislocations in a Vortex Lattice by Neutron Diffraction”, manuscript in revision.