Improving polarized neutron imaging for visualization of the Meissner effect in superconductors

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ABSTRACT
The polarized neutron imaging technique provides a non-invasive method of characterizing localized magnetic fields inside superconductors. However, complete understanding of the magnetic field distribution has yet to be realized experimentally due to the complexity of the interaction between neutron polarization and magnetic field. In this article, we show that a well-defined and controlled magnetic field through the neutron path contributes to simplify the data analysis and makes future quantitative polarized neutron imaging possible. This is demonstrated in a set of experiments that visualize the magnetic field distribution inside and around the superconductors. The experimental results demonstrate that proper guide field setup allows the visualization of the magnetic field expulsion at the surface of the superconductor in the zero-field cooling condition, as well as the magnetic field trapped inside the superconductor under field cooling condition.

INTRODUCTION
The distribution of magnetic fields inside a superconductor is often interesting to researchers, but is difficult to be measured accurately. Conventional characterization methods such as Small Angle Neutron Scattering (SANS) or Scanning Tunneling Microscope (STM) either measure the magnetization of the whole sample without giving localized magnetization distribution information or only measure the magnetization distribution on the sample surface. Polarized Neutron Imaging (PNI), first demonstrated in 2008, provides a way of probing magnetic structures inside samples. This is achieved through recording the polarization shift of the neutron due to its Larmor precession within a magnetic field. The final neutron polarization is an integral of magnetic-field along the neutron path just as a conventional transmission radiograph is an integral of neutron attenuation along the path. The neutron’s non-destructive nature along with its deep penetration makes PNI a unique technique for studying magnetic fields inside samples. This technique has been implemented at several neutron imaging beamlines. The behavior of the neutron polarization in a magnetic field is described by the Bloch equation,

\[
\frac{dP}{dt} = -\gamma P \times B,
\]

where \( P \) is the neutron polarization, \( B \) is the external magnetic field, and \( \gamma \) is the neutron gyromagnetic ratio. The evolution of the neutron polarization defined by Eq. (1) can be visualized as a "rolling-cone" construction, as shown in Fig. 1. The opening angle of the cone, \( 2\phi \), is given by the ratio of the rate of
rotation of the \( B \) field, \( \omega \), and the Larmor frequency \( \omega_L = yB \) as

\[
\tan[\phi] = \omega/\omega_L.
\] (2)

If the field rotates slowly relative to the Larmor frequency (adiabatic case), the opening angle \( \phi \) is small, and the neutron polarization follows the magnetic field as its direction changes. While at the other extreme, when the field suddenly changes direction (extreme non-adiabatic case), the polarization simply precesses around the new field direction at the Larmor frequency. Beyond these two cases, the neutron polarization revolves around a time-varying magnetic field in a complicated manner, which can be solved based on Eq. (1) using an algorithm developed by Seeger and Daemen.\textsuperscript{10}

When utilizing PNI to probe unknown magnetic fields, it is clear from the above that the information obtained from a PNI experiment is often difficult to interpret due to the complex polarization vector motion.\textsuperscript{11} To extract useful information, it is necessary to experimentally maintain the neutron polarization in an adiabatic or extreme non-adiabatic condition outside the sample magnetic field region. Thus, the changes in neutron polarization due to the sample can be isolated and considered as a series of small regions through which the neutron passes. In each region where the magnitude of the magnetization is approximately constant, the change in direction of the neutron polarization is simply described by multiplying \( P \) by a rotation matrix defined by three Euler angles whose values can be calculated from the Bloch equation. Accordingly, the final polarization is the result of multiplying these matrices together along the neutron path. In this paper, we focus on the experimental setup used for PNI measurement of superconductors and explore the ways in which the neutron polarization can be controlled. The experimental results in the paper demonstrate that the geometry and the boundary conditions of the superconductor are essential to establishing neutron polarization control and the quantitative reconstruction of the sample magnetization.

The example superconductor we examine in this article is YBCO, a type-II superconducting material. This material exhibits a Meissner effect that, under Zero-Field-Cooling (ZFC), expels the external magnetic field and forbids a magnetic field orthogonal to its surface.\textsuperscript{12,13} In this case, as shown in Fig. 2(a), the magnetic guide fields can be applied parallel to the surfaces, and the neutron polarization can be guided adiabatically following the magnetic field. However, as shown in Fig. 2(b), neutron polarization orthogonal to the superconductor surface is not achievable through adiabatically following the magnetic field because the Meissner effect constrains the field to be parallel to the sample surface. The Meissner effect also causes the superconductor to trap the external magnetic field during Field-Cooling (FC) across its critical temperature. Two FC cases are investigated, as shown in Figs. 2(c) and 2(d). In the case of Fig. 2(c), where the trapped field is parallel to the surface through which neutrons pass, the neutron polarization experiences a non-adiabatic transition at the sample surface and simply precesses around the magnetic field inside the sample. The total precession angle, \( \phi_L \), within the sample is simply related to the line integral of the sample magnetization along the neutron trajectory as

\[
\phi_L = yBt = \frac{ym}{h} \lambda \cdot \int_{\text{path}} B(l) dl,
\] (3)
where \( m \) is the mass of neutron, \( h \) is Planck’s constant, \( \lambda \) is the neutron wavelength, and \( dl \) is the infinitesimal segment along the neutron path. For the case of Fig. 2(d) where the trapped field is orthogonal to the surface, the trapped field overlaps with the external magnetic field and thus creates a complicated surface condition.

**INSTRUMENTATION SETUP**

The PNI experiments were performed at the imaging beamline (CG-1D) at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory. The beamline provides a steady beam of single wavelength (\( \lambda \approx 2.53 \text{ Å} \)) neutrons polarized by supermirrors and an in situ Spin Exchange Optically Pumped (SEOP) polarized \(^3\text{He} \) system. A similar setup can be found in an earlier publication. The schematics and pictures of the experiment setup are shown in Fig. 3. We use a set of rotatable electromagnets [Fig. 3(b)] to provide a well-controlled magnetic field parallel to the sample surface. These electromagnets are equipped with an angular scale so that the direction of the guide field can be read off with 0.5° angular precision. A set of solenoids is used to generate the magnetic fields perpendicular to the sample surface.

Two types of commercially available YBCO are selected as our superconducting samples. The first sample is a single crystal YBCO thin film, prepared by Ceraco®. This thin film sample, with dimensions of 100 mm \( \times \) 70 mm and a thickness of 350 nm, is deposited on a 0.5 mm thick single crystal sapphire. The second sample is a single crystal YBCO sample block, which is provided by Evico® and has a dimension of 45 mm \( \times \) 45 mm \( \times \) 10 mm and its c-axis parallel to the neutron beam. Both samples have a critical temperature \( T_c \) of \( \sim 90 \text{ K} \) and are cooled using a two-stage Closed-Cycle-Refrigerator (CCR) (Sumitomo CH-204), capable of reaching a base temperature of 22 K. For the ZFC experiment, the superconducting sample is cooled while enclosed inside a mu-metal enclosure to shield against ambient fields. During the experiment, we use a rectangular shape vacuum tail to maintain the cryogenic cooling while minimizing the distance between the electromagnets and the sample to 2 cm. Both samples are placed inside the vacuum tail with their larger surfaces perpendicular to the neutron beam.

**RESULTS AND DISCUSSION**

The data collected from the experiments are the measured intensities of the “spin-up” (\( I_+ \)) and “spin-down” (\( I_- \)) states of the neutron beam, defined as the neutron spin being parallel or antiparallel to the direction of the neutron polarization analyzer (the SEOP \(^3\text{He} \) filter). From the measured neutron polarization, we calculate the neutron polarization changes due to the sample, \( P_{\text{Sample}} \), as

\[
P_{\text{Sample}} = \frac{I_+ - I_-}{I_+ + I_-}, \quad \frac{1}{A \cdot P}.
\]

where \( A \) is the \(^3\text{He} \) filter’s analyzing power and \( P \) is the supermirror’s polarizing power. The combination of \( A \cdot P \) is determined to be (0.89 ± 0.01) by measuring the neutron polarization without the sample.

We first explore the setup where the superconducting YBCO film sample is cooled in ZFC condition and then placed between two different magnetic guide fields. As shown in Fig. 4(a), two magnetic guide fields \( B_1 \) and \( B_2 \) are generated by a solenoid and an electromagnet before and after the superconducting YBCO film, respectively. The electromagnets on the back side generate a uniform magnetic field of 20 G, defined by its orientation, up to the superconductor surface. The front solenoid magnetic field diverges and becomes parallel to the superconductor plane at the surface, aligning with the radial direction pointing from the center axis of the solenoid. The field distribution along the neutron flight path around the superconductor is simulated using Magnet®, a finite element analysis-based software, as shown in Fig. 4(b). The simulation shows continuous field distribution from the solenoid and the magnets while an abrupt shift occurs at the YBCO film position. The magnetic field distribution from the solenoid at the front surface of the YBCO film is also simulated, as shown in Fig. 4(c). The simulation shows that the magnetic field is parallel to the surface of the superconductor pointing toward or away from the center of the solenoid.

The neutron is first adiabatically guided by the solenoid magnetic field so that their polarization directions align with the diverging magnetic field at the front surface of the superconductor. Crossing the superconducting thin film, neutrons undergo an extreme non-adiabatic field transition from \( B_1 \) to \( B_2 \). If we define \( \theta \) as the...
angle between \( B_1 \) and \( B_2 \), the measured neutron polarization after
the superconductor is given by

\[
P_{\text{sample}} = \cos(\theta) = \frac{\vec{r} \cdot \vec{z}}{|\vec{r}|},
\]

where \( \vec{r} \) is the position vector from the center of the solenoid and \( \vec{z} \) is
the direction of \( B_2 \). In the ideal case where all transitions in solenoid
field \( B_1 \) are adiabatic, the polarization distribution as a function
of position is calculated from Eq. (5) and demonstrated in Fig. 5(a).
The corresponding measured PNI result with the same magnetic
field configuration is shown in Fig. 5(b).

Comparison between the calculated [Fig. 5(a)] and measured
results [Fig. 5(b)] confirms that the field distribution predicted by
Eq. (5) is qualitatively correct. We further examine the correlation
between the \( B_2 \) field direction and the neutron polarization
distribution from Eq. (5) by rotating the electromagnets 45° and
135° clockwise from the vertically upward direction. The measured
results in Figs. 5(c) and 5(d) show that the imaging pattern rotation
is consistent with the rotation of the electromagnet.

To visualize the field trapped inside superconductors in the FC
condition, we first examine the case where the trapped field is parallel
to the superconductor surface. The trapped field is generated by
placing electromagnets symmetrically on each side of a single YBCO
block with the generated magnetic field vertically upward, \(^{16}\) shown
in Fig. 6(a). By controlling the current through the electromagnets,
we selectively apply 7.5 G and 15 G at the center of the sample
position. The YBCO block sample is then cooled down through \( T_c \)
within this field. After cooling, we rotate both electromagnets in the
horizontal direction, as shown in Fig. 6(b).

The neutron polarization experiences non-adiabatic transitions
at the front and back surfaces, where the direction of the magnetic
field changes by 90° (\( \pi/2 \) flip). As a result of the first flip, the neutron
polarization precesses around the trapped magnetic field inside the
sample where the amount of the precession can be calculated from
Eq. (2). The measured polarization is the cosine of the precession
angle, given by

\[
P = \cos\left( \frac{\gamma_m \lambda \cdot B_{\text{trap}} \cdot l}{h} \right),
\]

where \( l \) is the thickness of the YBCO block (1 cm) and \( B_{\text{trap}} \) is the
magnitude of the trapped magnetic field. We compare the result of
the two different trapped field magnitudes along with zero trapped
field results, as shown in Fig. 7.

All results demonstrate a shift of neutron polarization across
the sample, although the non-zero trapped field result shows devi-
ation at the sample boundary. We attribute these deviations to the
non-uniformity of the electromagnets’ guide fields at the edge of

FIG. 6. Experimental setup with a trapped field parallel to the surface the neutrons pass through: (a) relative position of the external guide fields, superconductor, and magnetic field during field trapping and (b) relative position of the external guide fields, superconductor, and magnetic field during measurement.
the sample. To determine the trapped field magnitude, we calculate the neutron polarization shift due to the sample by average across the sample area. The average polarizations are $1.00 \pm 0.01$, $0.61 \pm 0.01$, and $-0.20 \pm 0.01$ for the results shown in Figs. 7(a)–7(c), respectively. Calculated from Eq. (6), these neutron polarizations correspond to a trapped field of $0.5 \, \text{G} \pm 1.5 \, \text{G}$, $7.8 \, \text{G} \pm 0.1 \, \text{G}$, and $15.1 \, \text{G} \pm 0.1 \, \text{G}$. The applied magnetic field and measured trapped field from neutron polarization are close, although a small disparity exists due to slight misalignment between the two electromagnets and background noise.

Lastly, we explore the situation where the FC trapped field is orthogonal to the surface that the neutron passes through. This configuration is first explored by placing a small permanent magnet in front of the YBCO block while cooling through $T_c$, as shown in Fig. 8(a). The permanent magnet is placed 2 cm upstream from the YBCO block, generating a 200 G magnetic field on its surface. After cooling, the permanent magnet is removed, and electromagnets are installed to provide guide fields along the vertical [Fig. 8(b)] and horizontal [Fig. 8(c)] directions.

The trapped magnetic field extends beyond the sample boundary and superposes with the external guide field at the surface. This leads to a neutron polarization that varies in complicated manner over the superconductor surface. The complexity of the polarization at the superconductor surface is amplified by the precession due to the trapped field and when neutron leaves the superconductor. The PNI results are shown in Figs. 9(a) and 9(b) with the external magnetic fields vertical and horizontal, respectively.

To simplify the spin evolution process, we repeated our measurement with the YBCO block replaced by a YBCO thin film whose thickness (350 nm) is so small that the neutron precession within the thin film is less than 0.0008° and therefore negligible in the measurement. Thus, we eliminate the influence of the magnetic field trapped inside the sample and only investigate the surface effect of the superposition of trapped field and external guide field. The results obtained using the same setup as described in Fig. 8 are shown in Figs. 9(c) and 9(d). These results demonstrate that the polarization distribution pattern follows the rotation of the electromagnetic fields, while the center pattern remains largely unchanged. This result suggests that in the center region where the trapped field (200 G) is much stronger than the external guide field (15 G), the neutron polarization follows the field adiabatically throughout its flight path. This adiabatic behavior allows PNI to identify pinned...
flux at the surface of the superconductor. Based on this observation, it is possible to experimentally probe the magnitude of the trapped flux by varying the guide field magnitude. Unfortunately for our current setup, a controllable guide field that can reach 200 G is not yet available but will be developed in future experiments. Comparing results from the YBCO block and thin films, adiabatic neutron polarization transfer is observed in both cases near the center of the pattern. This result suggests that the neutron polarization transfer through the field trapped inside the YBCO block remains adiabatic.

CONCLUSION

Our experiments explored and demonstrated that well-controlled guide fields around the measured sample are essential to PNI measurements. This requirement is especially true for superconductors with the Meissner effect that distorts the magnetic field. For future measurement that quantitatively determines the internal magnetic field strength and distribution of a sample, instrument upgrade that enables neutron computed tomography (nCT) and sample rotation is needed. In addition, the current measurement is limited by its resolution of 350 µm, which can be further improved for investigation of flux pinning within the superconductor. From our observation, the trapped field is uniform up to our resolution limitation. Improving the resolution of PNI techniques to the size of type-II superconductor domains will provide more insight into quantum materials and is our major focus for future development.

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