How the macroscopic current correlates with the microscopic flux-line distribution in a type-II superconductor: an experimental study

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Abstract
We present a study of the real-space flux-line lattice (FLL) of pristine and neutron irradiated conventional type-II superconductors using scanning tunnelling microscopy. Our work is focused on the magnetic field range, where the critical current density shows a second peak as a result of neutron irradiation. Scanning tunnelling microscopy images, including more than 2000 flux lines, are used to evaluate various microscopic parameters describing the disorder of the FLL, such as the defect density, the nearest neighbour distances and correlation functions. These parameters are compared with the macroscopic critical current density of the samples. The results show a direct correlation of the micro- and macroscopic properties. We observe a clear transition from an ordered to a disordered lattice at the onset of the second peak. Moreover, we discuss the defects of the FLL and their accumulation to large clusters in the second peak region.

Keywords: vortex phases, vortex pinning, critical currents

(Some figures may appear in colour only in the online journal)

1. Introduction
Several interactions govern the flux-line lattice (FLL) behaviour in a superconductor. Most important are the repulsion between flux lines (FLs), favouring an ordered FLL, and the pinning energy, favouring a disordered lattice [1]. Each of these energies dominates in a distinct area of the phase diagram. By exposing a sample to neutron irradiation, we can introduce material defects which act as additional pinning centres for the FLs and thus enhance the pinning energy. Accordingly, an increase in the macroscopic critical current density ($J_c$) and in some cases the emergence of a second peak at high magnetic fields are observed [2]. As $J_c$ directly reflects the FL distribution and is often crucial for technical applications, a detailed understanding of the FLL structure and a comparison between theory and experiment are important.

Several studies on the FLL and on a single FL with various techniques such as Bitter decoration [3–7] and neutron diffraction [8–10] measurements are available in the literature. These techniques are constrained by their limited field range at which the experiments have to be carried out. Another technique is the measurement of the FLL in real space with a scanning tunnelling microscope (STM) [11]. This method was used for the study of the field and temperature dependence of the FLL [12–14] and single FLs [15, 16]. In [17] the correlation of the peak effect with the structure of the vortex lattice was investigated for Co and Mn-doped niobium diselenide (NbSe$_2$). The authors were able to measure about 100 to 200 FLs per image and found an abrupt change in the orientational order parameter and in the number of defects in the FLL at a magnetic field corresponding to the peak-effect regime, as well as a sudden change in the FLL nearest-neighbor mean-square displacement. Although there are several publications investigating the FLL, only a few groups were able to record a large number of FLs with an STM in a single image [12], and no such results exist for the second peak region, i.e. images with much more than 100
FLs. In our work we present an STM study of the FLL with a much larger number of FLs per image, which permits us to achieve a more accurate statistical interpretation on the FLL and the quantities which depend on it. The measurements were carried out at different magnetic fields on pristine (unirradiated) and neutron irradiated single crystals.

In the following sections we will first describe the experimental details (section 2) and the data evaluation (section 3) of the measurements. In section 4 we will discuss the results acquired from these evaluations and draw conclusions. Finally we will present a short summary.

2. Experiment

The investigated sample was a NbSe$_2$ single crystal. NbSe$_2$ becomes superconducting below a temperature of about 7 K; its upper critical field is about 4 T perpendicular and 12 T parallel to the Nb planes at 0 K [18]. The crystal was exposed to a fast neutron fluence of $10^{16}$ m$^{-2}$ ($E > 0.1$ MeV) in a TRIGA Mark-II research reactor [19], which creates defects capable of FL pinning.

To compare the macro- and microscopic properties, we measured the sample in a SQUID magnetometer and in an STM before and after neutron irradiation. The SQUID data of the magnetic moment was used to evaluate $J_c$ (macroscopic property, for details of the evaluation see [20]), and the STM data (figure 1) yielded the FLL and its disorder (microscopic properties). While the magnetic moment of the unirradiated sample was almost reversible except at very low fields, the irradiated sample showed an increase of $J_c$ and a second peak. The second peak started to emerge in increasing fields at about 0.7 T and had its maximum at 1.1 T.

The STM was operated in the constant current mode at 4.2 K and the bias voltage to 0.7 mV, which is well below the energy gap value of about 2 mV in NbSe$_2$. At each point, the tunnelling current ($I$) and thus the density of states, which reveals an increase of $J_c$ at positions $r_j$ and a second peak. The second peak started to emerge in increasing fields at about 0.7 T and had its maximum at 1.1 T.

3. Evaluation

3.1. Defect density

We start the evaluation by localizing the FL centres in each raw image followed by counting the number of nearest neighbours ($n_j$) of each FL $j$ by employing a Delaunay triangulation (cf figure 2). The undisturbed FLL has hexagonal symmetry, i.e. $n_j = 6$, with the field dependent nearest neighbour distance of $a_0(B) = \left(\frac{2}{\sqrt{3}} \cdot \frac{\phi_0}{B}\right)^{1/2}$, where $\phi_0 = 2.07 \times 10^{-15}$ Tm$^2$, and $B$ is the magnetic induction. In a disturbed FLL, an FL can have more or less than six neighbours, thus defining a lattice defect. To determine the defect density in the lattice, we calculate:

$$\rho_{\text{FLL}} = \frac{1}{N} \sum_{j=1}^{N} \left|6 - n_j\right|,$$

where $N$ is the number of FLs used for the evaluation. Note that $|6 - n_j|$ may be larger than one to take into account the impact of very strong disorder on the macroscopic current. However, this case is rare and occurs only at high disorder (cf figure 2; 0.9 and 1 T).

3.2. Lindemann number

An important disorder parameter, which is used in theoretical models such as the collective pinning theory [21] and other theories on the FLL (e.g. [22]), is the relative displacement $u$ of an FL from its ideal position in the hexagonal lattice. This displacement is needed for the phenomenological treatment of the order–disorder transition by applying the Lindemann criterion, which was originally used to describe melting in a metal. For instance, in [23–25] an increase of $\left<u^2\right>^{1/2}/a_0$ above a threshold value $c_\perp \sim 0.1 \sim 0.2$ ($c_\perp$ is the so-called Lindemann number) is assumed to indicate the order–disorder transition of the lattice. We characterize a similar displacement by:

$$c_{\text{FLL}} = \frac{\left<u_{\text{m}}^2\right>}{a_0},$$

where $u_{\text{m}} = \left|r_j - r_m\right| - a_0$, with $j$ and $m$ the indices of neighbouring FLs at positions $r_j$ and $r_m$. This procedure may result in a slightly smaller value of $c_\perp$.

3.3. Correlations

For more details, we analyze the bonds between neighbouring FLs. The corresponding angle $\theta_{\text{FL}}(r_\alpha)$ between the bonds and an arbitrary axis (e.g. the $x$-axis) is evaluated, so that a parameter $b_{\alpha}$ can be defined, whose absolute value describes the bond-orientational order of the lattice [26]:

$$b(r_\alpha) = \frac{1}{n_j} \sum_{n=1}^{n_j} e^{i \theta_{\text{FL}}(r_\alpha)},$$

where the sum runs over the nearest neighbours of an FL. For an ideal hexagonal lattice with an angle $\alpha$ to the $x$-axis, we get $\theta_{\text{FL}}(r_\alpha) = \alpha + m \pi/3$ and thus $b_\alpha = e^{i \alpha}$, whose absolute value is 1. Every distribution of the nearest neighbours different from an ideal FLL yields a value lower than 1, specifying the extent of the angular disorder. Using $b_\alpha$, the orientational
correlation function can be acquired by:
\[
C_b(r, \Delta r) = \sum_{j,l=1}^{N} \frac{b(r_j)b(r_l)}{n(r, \Delta r)} \Theta \left( \frac{\Delta r}{2} - |r_{jl} - r_l| \right),
\]
\[
C_g(r, \Delta r) = \sum_{j,l=1}^{N} \frac{g^g(r_j-r_l)}{n(r, \Delta r)} \Theta \left( \frac{\Delta r}{2} - |r_{jl} - r_l| \right)
\]
where \( n(r, \Delta r) \) is the number of vortices that fulfil the condition \( \Theta \left( \Delta r / 2 - |r_{jl} - r_l| \right) \), with \( \Theta \) the Heaviside step function, i.e. \( \Theta(x) = 1 \) for \( x \geq 0 \) and zero otherwise. In this work the value of \( \Delta r \) is the pixel-size of the scan image and is typically \( \leq 2 \) nm and \( r_{jl} = |r_j - r_l| \) is the distance between two FLs \( j \) and \( l \), with \( j \neq l \). \( C_b(r, \Delta r) \) is a measure of the angular misalignment of the FLs with respect to a perfect FLL. A similar correlation function that measures the translational symmetry in the FLL is the translational correlation function:
\[
C_b(r, \Delta r) = \sum_{j,l=1}^{N} \frac{b(r_j)b(r_l)}{n(r, \Delta r)} \Theta \left( \frac{\Delta r}{2} - |r_{jl} - r_l| \right)
\]
\[
C_g(r, \Delta r) = \sum_{j,l=1}^{N} \frac{g^g(r_j-r_l)}{n(r, \Delta r)} \Theta \left( \frac{\Delta r}{2} - |r_{jl} - r_l| \right)
\]
where \( A(r, \Delta r) \) is a normalization factor which is calculated numerically. It takes into account that parts of the circular area enclosed by \( r \pm \Delta r / 2 \) may lie outside the measurement area and scales the values so that \( g(r, \Delta r) \to 1 \) for \( r \gg a_0 \) in a completely disordered lattice (i.e. a liquid). In the case of an ideal lattice, \( g(r, \Delta r) \) shows sharp peaks around the first, second, etc nearest neighbour distances (i.e. \( a_0, \sqrt{3}a_0, \ldots \)), with a peak-height corresponding to the number of neighbours at that distance. With increasing disorder in the FLL these peaks broaden and merge with neighbouring peaks.
4. Results

Figure 1 shows the STM images at magnetic fields of 0.5, 0.8 and 1 T for the irradiated sample and at 1 T for the unirradiated sample. The number of FLs lies between about 1000 and 2500 per image. Even without any detailed study of the images, the transition from an ordered lattice state to a disordered state is easily seen. The similar angular orientation of the lattices at different fields in the ordered field region indicates an alignment of the FLs to boundaries in the sample, e.g. the sample boundaries.

![Figure 2: Delaunay diagrams of the irradiated NbSe₂ crystals at different magnetic fields. Each intersection indicates the position of an FL and the symbols give the number of nearest neighbours: four (○); five (□); seven (■); eight (♦); no symbol is shown for six nearest neighbours.](image-url)
4.1. Lattice defects and displacement

The FLs found in the STM images are displayed via Delaunay diagrams in figure 2, where each line indicates a nearest neighbour bond and each intersection an FL position. The symbols highlight the positions of defects and their number of nearest neighbours. In the field region where $J_c$ is high (0.9 and 1 T), the strong disorder and the large number of defects can be seen. If $J_c$ is small, a well ordered lattice is expected and confirmed by our results, although some defects are still found. Our observations show that the FLL changes from an ordered to a disordered state (order-disorder transition) between 0.8 and 0.9 T.

In the more ordered FLL of the irradiated sample, all defects emerge in pairs of five and seven nearest neighbours (cf figure 2: 0.5–0.8 T). Defects appear in pairs, with one FL having five and the other having seven nearest neighbours. In most cases, two or more of these pairs merge to defect clusters, which are supposed to be more stable than single pairs. In the second peak region, FLs with four and eight neighbours emerge (cf figure 2: 0.9 and 1 T) and the defects accumulate into larger clusters, i.e. the images show certain areas where the FLL is only slightly disordered while in others many defects are found. Note that the observation of larger clusters, as presented in figure 2, has not been reported so far and is only possible because we recorded a very large number of FLs per image. Results acquired from smaller images (the number of recorded FLs is usually below 100 in the literature) may considerably depend on the specific scan area on the sample surface. In the unirradiated sample an almost perfect long range order is found at all fields, while the
irradiated sample shows more disorder and less long range order even in the reversible regime.

In the top panel of figure 3, a comparison of the defect density \( \rho_{\text{FLL}} \) and the displacement \( s_{\text{FLL}} \) with the critical current density \( J_c \) is shown. Note that the figure has three y-axes for the different values to emphasize their correlations. \( J_c \) was evaluated using only the first branch of a SQUID magnetization measurement (increasing field), since the STM measurements also refer to this branch. The values of \( \rho_{\text{FLL}} \) can be fitted to \( J_c \) simply using a constant factor of \( 3.84 \times 10^3 \). For the displacement values, an extra ‘offset’ fit-parameter has to be introduced in addition to the constant factor to shift the values closer to \( J_c \). The ‘offset’ shift was found to be 7.81 \( \times 10^{-2} \) and the scaling factor to be 2.26 \( \times 10^9 \). We show that the correlation between \( J_c \) and the FLL defect density and \( s_{\text{FLL}} \) is well described by a linear relation, but it should be noted that only one point at the first peak (with few FLs) and two points in the second peak range contribute to the linear fit of the data points and a more complex correlation between the parameters cannot be excluded. As mentioned above, our observations indicate an order–disorder transition between 0.8 and 0.9 T. Based on this assumption, the value of \( s_{\text{FLL}} \) (our modified Lindemann number) for the order–disorder transition lies between 0.08 and 0.10.

The results suggest a direct correlation of the microscopic (\( \rho_{\text{FLL}} \) and \( s_{\text{FLL}} \)) and the macroscopic quantities (\( J_c \)). Some uncertainties of \( \rho_{\text{FLL}} \) and \( s_{\text{FLL}} \) are expected to arise from sample inhomogeneities or statistical fluctuations. For example, \( s_{\text{FLL}} \) shows an increase at 0.7 T that is not seen in \( J_c \). At low magnetic fields (i.e. 0.1 and 0.2 T) only a few 100 FLs could be detected because of the limited scan area, resulting in larger errors (see below).

In the same figure 3, but on the bottom panel, we provide an error estimation of \( \rho_{\text{FLL}} \) and \( s_{\text{FLL}} \) by dividing the scan-areas into equally sized sub-images, i.e., into 2, 4, 9 and 16 sub-images out of the original image. The sub-images are used to calculate the relative error for a smaller number of FLs per image by:

\[
\frac{\Delta \zeta}{\zeta} = \frac{1}{S} \sum_{p=1}^{S} |\zeta_p - \bar{\zeta}|, \tag{7}
\]

where \( S \) is the number of the sub-images and \( \zeta \) represents \( \rho_{\text{FLL}} \) or \( s_{\text{FLL}} \). \( \bar{\zeta} \) is calculated by equation (1) or (2) for each sub-image and \( \Delta \zeta \) is the mean value of all \( S \) sub-images. As suspected the relative error of \( \rho_{\text{FLL}} \) is found to be larger for smaller images (see upper graph of the bottom panel in figure 3). The increase of the relative error is much more pronounced in the field range where \( J_c \) is small. This is easily understood by considering the accumulation of defects into clusters as shown in figure 2. Consequently, a large number of FLs (>1000) is crucial in this field range. In contrast, the relative error in the second peak region does not change as strongly and is rather small, e.g., it is about 4% for 830 FLs at 0.9 T and similar for 600 FLs at 1 T. Thus we judge the results to be sound for \( N \geq 1000 \) at these magnetic fields.

As for \( s_{\text{FLL}} \), the relative error of this parameter exhibits a similar behaviour as \( \rho_{\text{FLL}} \); the values are much smaller and increase significantly only below about 200 FLs. Accordingly, the number of FLs per image can be lower when calculating \( s_{\text{FLL}} \) (\( \geq 200 \)) than for evaluating \( \rho_{\text{FLL}} \) (\( >1000 \)).

### 4.2. Correlations

In figure 4 the pair correlation function \( g(r, \Delta r) \) of the irradiated sample is shown at 0.5, 0.8 and 1 T and at 1 T of the unirradiated sample. In general the lattices of the irradiated sample do not show as distinct peaks as the unirradiated sample. Thus the bond length correlation of the FLL is higher in the unirradiated material. At small distances \( g(r, \Delta r) \) exhibits an oscillating behaviour, but as the distance increases, the oscillations in the irradiated sample weaken until they vanish, indicating the loss of long range order. The observed peak to peak distances in \( g(r, \Delta r) \) reflect the distances between the first, second, etc nearest neighbours of the hexagonal lattice structure. While \( g(r, \Delta r) \) of the irradiated sample hardly shows any separate peaks, aside from the oscillating maxima, the unirradiated sample has separate peaks beyond 10\( a_0 \). In a lattice with many defects (e.g. the irradiated sample at 1 T), the oscillation amplitudes decrease rapidly with distance and are no longer visible at distances greater than about 3\( a_0 \), which indicates a so-called vortex glass state [27]. But, as shown in figure 2, the vortex distribution is not completely amorphous. The decay of the oscillations is also observed at lower fields (e.g. 0.5 T) in the irradiated sample, where we expected an ordered FLL. But the oscillations fade at much larger distances (\( \simeq 10–20a_0 \)) than in the second peak region. This long-range order of the FLL for the fields 0.5–0.8 T can be described in context of a Bragg glass-like state. Moreover, \( g(r, \Delta r) \) of the unirradiated sample still
oscillates at $r/a_0 \approx 30$, showing excellent long-range order, as expected for the Bragg glass phase [22].

Figure 5 shows the translational correlation function and figure 6 the orientational correlation function of the measured lattices at the same magnetic field values as the pair correlation function. The dotted lines are exponential ($e^{-\eta r}$) and the dashed lines are algebraic ($r^{-\eta}$) decay fits to the envelopes of the functions. The highest correlations are found in the unirradiated sample, which shows a very slow (algebraic) decay for the translational correlation function, indicating a quasi long-range order and a virtually constant orientational order. These properties are indeed predicted for a two-dimensional Bragg glass [28, 29]. In the irradiated state, we find at 1 T, i.e. in the most disordered lattice, a rather fast decay of both correlation functions, as expected for a vortex glass [27].

Note, that this lattice (see figure 2) is not completely amorphous but rather polycrystalline and the fits to the envelope show an exponential decay only for small distances. In a completely amorphous lattice the exponential behavior is expected to cover the whole range of $r$. At 0.5 and 0.8 T the state of the FLL is in between the two above mentioned states. It is apparent that the translational order declines significantly at these fields, while the orientational correlation suggests more long-range orientational order. Such a behavior is actually predicted for the hexatic state of a two-dimensional system [26, 30].

Figure 7 shows the parameter defined by equation (3), which describes the bond-orientational disorder of the FLL at each FL position. This visualization technique is very sensitive to small lattice deformations. The shades indicate the values of $b_j$ ($0 \leq b_j \leq 1$), where bright shading represents an ordered, near-hexagonal lattice structure ($b_j \approx 1$) and dark shading a disordered structure ($b_j \approx 0$). The positions of the FL cores are marked by different symbols depending on the number of nearest neighbours. As expected the orientational disorder usually grows near lattice defects, i.e. where the number of nearest neighbours differs from six. Again we find a clustering of low and high disorder instead of a homogeneous disorder distribution. While the unirradiated sample shows a nearly perfect bond-orientational order, the irradiated sample displays more disorder even in areas far away from any defects (cf 0.8 T of irradiated and 1 T image of the unirradiated sample).
5. Summary

In summary, our results reveal a direct correlation of the micro- and macroscopic properties of the FLL in a type-II superconductor. In particular, we find the critical current to be proportional to the defect density of the FLL and the relative displacement of the FLs. In the second peak region a highly disordered FLL is observed, while in regions where the macroscopic current vanishes the lattice is found to be well ordered in the irradiated sample and close to perfectly ordered in the unirradiated sample. The extent of the lattice order is reflected in the nearest neighbor peaks of the pair correlation function. The peaks are seen at distances beyond $r/a_0 \approx 30$ in the unirradiated sample, reflecting a highly ordered state. In contrast to this, the peaks disappear at $r/a_0 \approx 10$–20 in the irradiated sample, for fields below the second peak regime, indicating a still fairly ordered state. The second peak regime shows strong disorder, which is seen by the disappearance of the peaks at $r/a_0 \approx 3$. The translational and orientational correlation functions suggest a Bragg glass state for the unirradiated sample at the measured fields. In the irradiated sample we observed a fast decay of these functions in the region of the second peak, as expected for a vortex glass, while the FLL state at the fields below the second peak lies between the two above mentioned states. The defect structure of the FLL consists of pairs with five and seven nearest neighbours, which often accumulate into larger clusters, whose sizes increase significantly in the second peak region. In the second peak region defects with four and eight nearest neighbours also appear. A similar clustering is found for the parameter describing the bond-orientational order of the FLL. Like the defect clusters, the bond-orientational order of the FLs shows areas where the lattice is well ordered and areas of strong disorder. To identify such effects it is essential to record a large number of FLs per image, as demonstrated by our study.

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