Imaging pinning and expulsion of individual superconducting vortices in amorphous MoSi thin films

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(Received 16 July 2019; published 5 September 2019)

We use a scanning nanometer-scale superconducting quantum interference device (SQUID) to image individual vortices in amorphous MoSi thin films. Spatially resolved measurements of the magnetic field generated by both vortices and Meissner screening satisfy the Pearl model for vortices in thin films and yield values for the Pearl length and bulk penetration depth at 4.2 K. Flux pinning is observed and quantified through measurements of vortex motion driven by both applied currents and thermal activation. The effects of pinning are also observed in metastable vortex configurations, which form as the applied magnetic field is reduced and magnetic flux is expelled from the film. Understanding and controlling vortex dynamics in amorphous thin films is crucial for optimizing devices such as superconducting nanowire single photon detectors (SNSPDs), the most efficient of which are made from MoSi, WSi, and MoGe.

DOI: 10.1103/PhysRevB.100.104504

I. INTRODUCTION

The dynamics of quantized vortices play a crucial role in determining the electronic properties of devices made from type-II superconductors. The dissipative motion of vortices, driven by flowing electrical current, destroys the material’s ability to carry current with zero resistance. By controlling the pinning of vortices, they can be immobilized, thereby restoring the superconducting state. Optimized pinning [1–9] and other methods of arresting vortex motion [10] have been used to extend the coherent dissipation-free superconducting state to high critical current densities, fields, and temperatures. The microwave response of superconductors is also affected by the high-frequency dynamics of vortices [11]. Vortex trapping contributes to loss in microwave resonators and conditions the performance of the circuits used as qubits in superconducting quantum computers [12]. Evidence has also emerged that superconducting vortices play a significant role in superconducting nanowire single photon detectors (SNSPDs) [13], whose high speed, detection efficiency, and low dark count rates make them attractive for a wide variety of applications. In particular, vortices are likely involved in both the mechanism used for the detection of photons and in the generation of dark counts.

Here, we image individual superconducting vortices in thin-film MoSi using a scanning nanometer-scale superconducting quantum interference device (SQUID). Spatial maps of the magnetic field above patterned MoSi wires are collected as a function of applied field and reveal the presence of individual vortices and their pinning sites. Measured field profiles fit well to the Pearl model and yield the bulk London penetration depth $\lambda_L = 510 \pm 10$ nm at 4.2 K, which agrees with previous transport measurements of similar films [14]. Pinning sites of various strengths strongly affect the configuration of vortices. In particular, upon the reduction of the applied field, they hinder the expulsion of magnetic field from the film, resulting in long-lasting metastable vortex configurations. The results suggest that improved control of the density and strength of these pinning sites could be important for the optimization of devices based on amorphous MoSi films.

II. EXPERIMENTAL SETUP

We investigate a 65 ± 5-nm-thick film of amorphous Mo$_{0.76}$Si$_{0.24}$ which is patterned into 9-μm-wide strips with short sections of narrower width, as shown in Fig. 1(b). Similar MoSi nanowires have recently been used as SNSPDs with some of the highest reported detection efficiencies [15]. The composition and thickness are chosen in order to maximize the critical temperature $T_c$ while obtaining an amorphous film with low defect densities and weak pinning effects [14], as desired in most applications.

We use a scanning SQUID-on-tip (SOT) sensor [16,17] to map the component of the magnetic field perpendicular to the MoSi film as a function of position and applied field at 4.2 K. The SOT used here has an effective diameter of 310 nm, as extracted from measurements of the critical current $I_{SO}$ as a function of a uniform magnetic field applied perpendicular to the SQUID loop $B_a = B_y \hat{z}$ [18]. The MoSi sample is mounted below the SOT sensor in a plane parallel to the SQUID loop and perpendicular to $B_a$. Since the SOT’s current response is proportional to the magnetic flux threading through it, it provides a measure of the $z$-component of the local magnetic field $B_z$ integrated over the loop at its apex. By scanning the sample below the SOT at constant tip-sample spacing $z$, we map the local magnetic field $B_z(x, y, z)$. The sub-μm resolution is limited by the spacing and, ultimately, the SOT diameter.

III. VORTEX IMAGING

Before imaging $B_z(x, y)$ generated by the film, we nucleate the vortices by cooling the sample through its measured $T_c = 7.2$ K down to 4.2 K in an externally applied field $B_a$. This field-cooling process causes flux penetrating the film above $T_c$ to concentrate in the form of quantized vortices which are trapped within the sample as it cools. The trapping potential
is the result of the geometrical barrier produced by extended Meissner screening currents flowing near the sample edges [19]. Figure 1(c) is a typical wide-field image of $B_z(x, y)$ at a fixed tip-sample spacing of $z = 705$ nm, showing the Meissner screening (dark regions) and the presence of a few trapped vortices (bright spots).

In Fig. 2(a), more detailed maps of $B_z(x, y)$ taken after field cooling at $B_a = 0.5$ mT show a few isolated vortices. Line cuts across the MoSi wire through the center of a single vortex are shown for different $z$ in Fig. 2(b), and the dependence of the field as a function of $z$ directly above the vortex is shown in Fig. 2(c). These data are well fit by a model considering both the Meissner screening of the film [20] and the field produced by a perpendicular vortex in a film such that its thickness $t \ll \lambda_L$ (thin-film limit) [21], as first described by Pearl [22] and Carneiro and Brandt [23] (see Appendix D). Fits to these two effects measured at different $z$ allow us to independently determine the Pearl length $\Lambda = 2\lambda_L^2/t$, and thus $\lambda_L$, as well as the real tip-sample spacing $z$ for each measurement. At 4.2 K, we find $\Lambda = 8.1 \pm 0.9$ µm and a bulk $\lambda_L = 510 \pm 10$ nm, which agrees with measurements by Kubo of films with similar concentration and thickness [14].

### IV. VORTEX TRAPPING AND EXPULSION

By field cooling with different values of $B_a$, we initialize different vortex densities in the MoSi film, as shown in Fig. 3.
In the central region of the widest part of the wire, the measured vortex density depends linearly on $B_a$ with slope given by $\Phi_0^{-1}$, where $\Phi_0 = h/(2e)$ is the magnetic flux quantum, $h$ is Planck’s constant, and $e$ is the elementary charge. In fact, the density should go as $(B_a - B_K)/\Phi_0$, where $B_K$ is a critical induction below which no vortex trapping occurs, but because $B_K \ll B_a$ in our experiments, the measured results match theoretical predictions [24] as well as previous measurements in similar Nb [25], yttrium barium copper oxide (YBCO) [24], and Pb [26] wires. This behavior corresponds to the vortex density expected when the total flux through the film above $T_c$ nucleates into vortices and is trapped within the film. However, when the entire area of the MoSi wire is considered, including the edges, the measured vortex density is significantly less than the full above-$T_c$ flux density. The images of $B_z(x,y)$ in Fig. 3(a) show vortex-free regions near the sample edges, which shrink with increasing $B_a$, as also observed in Pb films by Embon et al. [27]. This nonuniform vortex density, which is concentrated in the center of the wire, reflects the reduced effective width compared to the wire width, in which vortices can be trapped by the Meissner screening currents. In thin-film wires whose width $w \gg t$, the screening current density decreases slowly from the edges, pushing vortices into the central part of the wire [19,27–29]. As a result, flux threading the sample near its edges above $T_c$ is subsequently expelled upon cooldown.

In order to investigate the potential trapping the vortices, after field cooling we image the vortex configuration upon the reduction of $B_a$. The left panel of Fig. 4(a) shows an image of $B_z(x,y)$ after field cooling at $B_a = 2.8$ mT to 4.2 K. The panels on the right show the same region immediately after ramping the applied field down to lower $B_a$ at constant temperature. In the case shown in the top half of Fig. 4(a), the field is ramped down $B_a = 0.5$ mT in 30 s and the $10 \times 10^{-\mu m^2}$ image is taken line-by-line in a total time of 260 s with $x$ as the fast axis. Towards the end of the image, after 200 s, we observe a sudden change of the vortex configuration. The system goes from an “inflated” state, in which vortices appear in the previously unoccupied edge region, to a state similar to that observed after field cooling, containing fewer trapped vortices and vortex-free edges. Upon further repeated imaging, the second configuration is always observed, except for small changes due to thermally activated vortex hopping, which will be discussed in the next section. In the bottom half of Fig. 4(a), the field is reduced in three steps to $B_a = 0.2$ mT, with similar discontinuities marking a transition between “inflated” and equilibrium states showing up in the images taken at $B_a = 1.9$ and 0.8 mT. The panels of Fig. 4(b), showing a $8.25 \times 8.25-\mu m^2$ view of $B_z(x,y)$ near the wire edge, display similar behavior 190 s after $B_a$ is reduced from 1.5 to 1.0 mT and 320 s after it is subsequently reduced to 0.5 mT. In particular, we note that in the inflated states observed at 1.0...
FIG. 4. “Inflated” metastable vortex configurations. (a) Left image shows a map of $B_z(x, y)$  705 nm over a MoSi wire segment after field cooling at $B_a = 2.8$ mT. The image on the top right shows a measurement of the same area immediately after a decrease of $B_a$ to 0.5 mT in 30 s. The $10 \times 10\, \mu m^2$ images are taken line-by-line in a total time of 260 s, with $x$ as the fast axis (71 lines at 3.7 s/line). The white line represents the moment at which we observe a discontinuous change in the vortex configuration. The bottom-right images are taken at three fields as $B_a$ is reduced from 2.8 to 0.2 mT. (b) $8.25 \times 8.25\, \mu m^2$ images of $B_z(x, y)$ near the wire edge, each taken in a total time of 245 s (51 lines at 4.8 s/line). (b) The leftmost image shows the configuration after field cooling at $B_a = 1.5$ mT. Other images show subsequent configurations after decreasing $B_a$ to 1 mT and then to 0.5 mT. The plot displays $B_z$ as a function of time at the position indicated by the gray circle, showing both the reduction in $B_a$ and the signatures of vortex expulsion, which are highlighted in the zoomed-in sections. Note that the flux measured at this position (gray circle) and the spatial maps do not correspond to simultaneous measurements; they correspond to measurements carried out under the same initialization conditions. Therefore, the gray lines connecting the 2D maps to various points along the plot are meant to indicate similar events in different experimental runs, not two measurements of the same event.

and 0.5 mT, the vortices in the edge region appear pinned to the same set of pinning sites. Such vortex expulsion behavior is observed repeatedly for similar intervals of $B_a$ and over similar time scales at constant temperature.

To confirm that transitions from the inflated state to the final state are not induced by interactions with our scanning tip [30], we carry out the same measurements with the SOT at a fixed position, just outside the edge of the sample, as marked by the gray circles in Fig. 4(b). Once again, $B_a$ is reduced at a constant temperature and the resulting field $B_z$ next to the wire is plotted as a function of time in the main part of Fig. 4(b). After the initial reduction due to the ramping down of $B_a$, $B_z$ is constant until around 150 s, when it increases in a sudden step. This increase corresponds to the expulsion of the extra vortices present in the inflated state. Vortex expulsion with similar time scales is observed upon repeated experiments and for different fields. Since this behavior corresponds to what was observed in the scanning experiments, we rule out interactions with the tip as the trigger for the observed flux expulsion. Although strong interactions were previously observed between vortices in YBCO and a magnetic force microscopy tip [30], the much weaker stray fields produced by our SOT tip produce a negligible perturbation to each vortex, calculated to be less than 10 fN following the procedure described by Embon et al. [26].

The observation of a metastable inflated state, which lasts on the order of a hundred of seconds before flux expulsion and relaxation to the equilibrium configuration, points to the presence of pinning sites and thermally activated vortex hopping. The presence of vortices near the edges in the metastable state cannot be accounted for by the trapping potential provided by Meissner currents: this potential only traps vortices in the middle of the wire, with its barrier occupying the region near the edges. The vortices near the edges can be immobilized...
FIG. 5. Thermally activated vortex hopping and pinning strength. (a) Maps of \(B_z(x, y)\) at \(z = 705\) nm over a MoSi wire segment after field cooling with \(B_a = 0.8\) mT. Each \(10 \times 10\) \(\mu\)m\(^2\) image is of the same region and is taken one after the other line-by-line in \(260\) s with \(x\) as the fast axis (71 lines at 3.7 s/line). Circles and arrows highlight vortices hopping between neighboring pinning sites. (b) Left image is a map of \(B_z(x, y)\) under the same conditions but in the additional presence of an ac applied current along \(\hat{y}\), producing a \(F_{ac}\) along \(\hat{y}\). The right image shows \(B_{z,ac}(x, y)\), which is measured simultaneously. The highlighted areas show vortices which oscillate as a result of \(F_{ac}\). Scan area \(10 \times 10\) \(\mu\)m\(^2\).

only by pinning sites. These potential wells, however, are not deep enough to completely freeze the vortex motion at 4.2 K and, due to thermally activated vortex hopping, the edge vortices are eventually expelled as the sample relaxes to its equilibrium configuration.

V. PINNING SITES

The presence of thermally activated vortex hopping at 4.2 K is evident from repeated imaging of \(B_z(x, y)\) at equilibrium. As shown in Fig. 5(a), certain vortices are seen to jump between neighboring pinning sites within a single image or between successive images. The former effect is evident in the discontinuities between one \(x\) line and the next, while the latter can be seen when a vortex changes position between images. These jumps occur on a time scale ranging from a few to a few hundred seconds. Given this time scale and the similar time scale of the metastable configuration held together by pinning sites at the sample edges, we estimate the depth of these pinning potentials \(E_{pin}\) using the Boltzmann formula for a thermally activated hopping rate: 
\[
\nu_{\text{hop}} = \nu_0 e^{-\Delta E_{\text{hop}} / k_B T}
\]
where \(k_B\) is the Boltzmann constant, \(T\) is the temperature, and \(\nu_0\) is the attempt frequency. Solving for \(E_{pin}\), we find an energy between 4 and 12 meV, whose uncertainty results from the imprecisely known ratio of \(\nu_{\text{hop}} / \nu_0\) (\(\nu_{\text{hop}}\) ranges from a few mHz to 1 Hz, while \(\nu_0\) is assumed to lie between \(10^5\) and \(10^{12}\) Hz [31,32]). These weak pinning sites are therefore not strong enough to freeze vortex motion at 4.2 K and thus provide a path through which the system can eventually find its equilibrium configuration. As \(B_a\) is increased, we note that pinning sites displaying thermally activated hopping occur more frequently from 25% of pinning sites at \(B_a = 0.6\) mT to up to 85% at \(B_a = 1.5\) mT (see Appendix E). As the vortex density increases, stronger vortex-vortex interactions, which have a long-range \(1/r\) force in thin films, destabilize an increasing number of vortex pinning sites.

Although many pinning sites exhibit thermally activated vortex hopping, some sites do not, pointing to deeper trapping potentials. We further investigate these sites by driving vortex motion using an ac current \(I_{ac}\) applied along the \(y\) direction (long direction of the wire). This current results in an ac Lorentz force \(F_{ac} = \frac{\mu_0}{2\pi} \Phi_0\) on each vortex along the \(x\) direction. \(F_{ac}\) drives oscillations of the vortices around their equilibrium positions. The resulting motion can be measured using the scanning SOT as either a blurred vortex image in \(B_z(x, y)\), as shown in the left panel of Fig. 5(b), or, more clearly, as a dipolar signal in the magnetic field produced by the sample at the frequency of the ac current drive \(B_{z,ac}(x, y)\), as shown in the right panel [26]. The amplitude of this signal reflects the size of the oscillation and the gradient of the static field \(B_z(x, y)\) produced by the vortex.

Vortices at different pinning sites respond differently to \(F_{ac}\), indicating the varying strength of the pinning potentials throughout the sample. At \(B_a = 0.9\) mT, weak pinning sites (30%), showing thermally activated hopping, are induced to jump from site to site by \(F_{ac}\) with amplitudes in the 100-fN range. Strong pinning sites (50%) show no measurable response to this \(F_{ac}\). Intermediate pinning potentials (20%) show oscillatory responses to the \(F_{ac}\). From the strength of \(F_{ac}\), we extract effective pinning spring constants of around 0.6 \(\mu\)N/m. Compared to similar measurements in Pb films, these potentials are 100 times shallower (less curvature),
indicating their larger spatial extent and weaker strength. By increasing Lorentz force up to a maximum value $F_{\text{ac,max}}$ at which these vortices escape from their pinning sites, we can integrate and estimate the depth of the potential well, typically obtaining $E_{\text{pin}} \sim 60 \text{ meV}$.

Together, our results point to a range of $E_{\text{pin}}$ from a few meV up to at least tens of meV, excluding the pinning sites that do not respond to thermal or Lorentz perturbations. As expected, these values are a fraction of the estimated condensation energy of the vortex core in this sample of $\frac{H_v^2}{\Phi_0^2} \xi^2 \pi \gamma^2 = 110 \text{ meV}$, where $\xi = 7 \text{ nm}$ is the coherence length derived from a measurement of the upper critical field $H_c = \frac{\Phi_0}{2\pi \xi^2} = 6.8 \text{ T}$ [14], $H_F = \frac{\Phi_0}{\sqrt{8\pi^2 \xi^2}} = 66 \text{ mT}$ is the thermodynamic critical field, and $\mu_0$ is the permeability of free space.

If we exclude the weak thermally activated pinning sites, we also find that the vortices are always driven in phase with $F_{\text{ac}}$ and along the direction of the drive ($x$ direction). This behavior points to isotropic conservative trapping potentials, which are sparsely distributed, i.e., large pinning centers that do not overlap. These large and well-separated pinning centers correlate some of the strongest pinning centers, which do not respond to either thermal or Lorentz perturbations, with the observations made by Embon et al. in similar experiments on Pb thin films [26]. There, vortices displayed anisotropic responses and spring softening upon driving with Lorentz forces, because pinning centers were close enough to form complex, anisotropic potentials for vortices. Those data were well-described by a computational model that included small clusters of pinning defects separated on the order of a superconducting coherence length. Our results also differ from recent scanning SQUID susceptibility measurements in the layered superconductor FeSe, which revealed weak and highly anisotropic pinning potentials, constraining vortex motion predominantly along twin boundaries [33].

The vortex pinning potentials observed in our MoSi films are likely the result of either crystalline precipitates within the amorphous alloy, structural defects, or local variations in the thickness and elemental composition. Given the presence of visible inclusions and roughness of varying sizes observed by scanning electron microscopy (SEM) of our sample [18], structural defects and thickness variations are the most probable causes. In fact, by comparing $B_c(x, y)$ images of vortices with SEM of the same part of the sample, we correlate some of the strongest pinning centers, which do not respond to either thermal or Lorentz perturbations, with pockmarks, perhaps representing perforations, on the surface of the film. The films may also have small inhomogeneities in Si concentration, which would result in local changes of $\lambda_L$ [14] and thus potential pinning sites. The presence of such inhomogeneities in thickness or composition could explain some slight differences in the maximum observed fields produced by different vortices. Precipitates are typically a limiting factor in much thicker films, which require long sputtering times for deposition [14].

VI. CONCLUSIONS

Our scanning SOT experiments, because of the sensor’s combination of high spatial resolution and high magnetic field sensitivity, reveal images of individual superconducting Pearl vortices in amorphous MoSi thin films. In addition to providing a measure of the penetration depth of the film, we directly observe their thermally activated hopping at 4.2 K. Since the vortices are not completely frozen onto their pinning sites, we are able to estimate the depth of the weakest pinning potentials and observe metastable vortex configurations present for tens of seconds before the system reaches equilibrium.

The dynamic nature of the vortex configurations in MoSi at 4.2 K may have implications for SNSPDs and other devices fabricated from such films. The thermally activated vortex hopping observed here may be a source of residual dark counts in SNSPDs. Our experiments also make plain the necessity of further reducing the density of unintentional pinning sites by improving sample quality. In order to aid in the optimization of MoSi-based superconducting devices, future scanning SQUID studies should aim to study different MoSi films grown under different conditions.

ACKNOWLEDGMENTS

We thank Prof. Christian Schönberger, Prof. Richard Warburton, and Daniel Sacker for guidance in the fabrication of the MoSi samples. We also thank the machine shop of the Department of Physics and Dr. Monica Schönberger and the Nano Imaging Lab for support with part design and AFM, respectively. We acknowledge the support of the Canton Aargau; the ERC Starting Grant NWScan (Grant No. 334767); and the Swiss National Science Foundation via Grants No. 200020-159893 and No. 200020-178863, the Sinergia network Nanoskyrmionics (Grant No. CRSII5-171003), and the NCCR Quantum Science and Technology (QSIT).

APPENDIX A: MOSI SAMPLE FABRICATION

The 65 ± 5-nm-thick Mo$_{0.75}$Si$_{0.24}$ film is deposited onto an SiO$_2$ substrate by cosputtering with a DC and rf bias on Mo and Si targets, respectively. The thickness is determined using atomic force microscopy (AFM) and SEM, while the concentrations of Mo and Si are measured by x-ray photoelectron spectroscopy (XPS) [18]. The film is patterned into a series of meandering wires over a $500 \times 500\mu\text{m}^2$ area [18] by a combination of electron-beam lithography and reactive ion etching.

APPENDIX B: SOT SENSOR FABRICATION

The SOT is fabricated by evaporating Pb on the apex of a pulled quartz capillary according to a self-aligned method pioneered by Finkler et al. [16] and perfected by Vasyukov et al. [17]. The evaporation is carried out in a custom-made evaporator with a base pressure of $2 \times 10^{-8} \text{ mbar}$ and a rotatable sample holder cooled by liquid He. In accordance with Halbertal et al. [34], an additional Au shunt was deposited close to the tip apex prior to the Pb evaporation for protection of the SOTs against electrostatic discharge. SOTs were characterized in a test setup prior to their use in the scanning probe microscope [18].

APPENDIX C: SCANNING SOT EXPERIMENTS

The SOT and the MoSi sample are mounted in a custom-built scanning probe microscope operating under vacuum.
FIG. 6. Dependence of thermally activated vortex hopping on applied field. (a–c) Time series of images of \( B_z(x, y) \) taken at three different field-cooling fields \( B_a \). Dashed circles in (a) and (b) highlight vortices which hop in subsequent frames. Given the large proportion of vortices hopping in (c), we do not highlight them there.

APPENDIX D: MODEL FOR MAGNETIC FIELD ABOVE THIN-FILM WIRE

The model used to fit the spatial dependence of \( B_z \) above the superconducting MoSi wire considers the wire’s expulsion of flux due to the Meissner effect and the field produced by a superconducting vortex in the Pearl limit. By summing the resulting field profiles, we model the 1D profile shown in Figs. 2(b) and 2(c), which cut through the center of the field produced by a single vortex in the wire.

The model for the field produced by Meissner screening follows Brisbos et al. [20]:

\[
B_{z, \text{Meissner}}(x, z) = 1 + \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{(x-x')d'x'}{[(x-x')^2 + z^2]^{3/2} + \frac{2\lambda L}{\pi t}}.
\]

The model for the field produced by the superconducting vortex is based on Pearl’s model, because the wire thickness is much less than the penetration depth \( t \ll \lambda_L \) [22,23]:

\[
B_{z, \text{vortex}}(x, z) = \Phi_0 \int_{-\infty}^{\infty} \frac{d^2k}{4\pi^2} \frac{e^{ikx}}{1 + \frac{4\lambda^2}{\pi^2} f(k, z)},
\]

where \( f(k, z) = c_1 e^{-\rho z} \) above the film, i.e., for \( z > 0 \), where 0 is the surface of the film. \( c_1 \) depends on the film thickness \( t \):

\[
c_1(k) = \frac{[(k + \rho)e^{\rho t} + (k - \rho)e^{-\rho t} - 2\lambda L^2]}{c_2}
\]

\[
c_2(k) = (k + \rho)^2 e^{\rho t} - (k - \rho)^2 e^{-\rho t}
\]

\[
\rho = \sqrt{k^2 + \left(\frac{2\lambda L^2}{t}\right)^2}.
\]
The measured field is then fit with the function $B_z(x, z) = B_{z, \text{Meissner}}(x, z) + B_{z, \text{vortex}}(x, z)$, where the London penetration depth $\lambda_L$ and probe-sample distance $z$ are used as fit parameters. The value of $\lambda_L$ is found to be $510 \pm 10 \text{ nm}$ (making the Pearl penetration depth $\Lambda \approx 8 \mu \text{m}$), and the $z$ values are shown in Fig. 2(b) for five different probe-sample distances.

### APPENDIX E: EFFECT OF VORTEX DENSITY ON THERMALLY ACTIVATED HOPPING

As the field-cooling field $B_x$ is increased, the proportion of pinning sites showing thermally activated vortex hopping increases. The increasing vortex density results in stronger vortex-vortex interactions, which destabilize more and more vortex pinning sites. This effect can be observed in Fig. 6, which shows three sequences of images taken one after the other for three different field-cooling fields $B_x = 0.6, 0.8$, and $1.0 \text{ mT}$. Each frame requires $153 \text{ s}$ to measure, and successive images are taken immediately after the previous image finishes [18]. As shown in Fig. 7, the proportion of pinning sites showing thermally activated vortex hopping, $R = n_t / n_0$, where $n_0$ is the density of vortices showing thermally activated hopping and $n_t$ is the total vortex density, increases with $B_x$ until nearly all vortices show thermally activated behavior.

![FIG. 7. Proportion vortices showing thermally activated hopping as a function of $B_x$ in measurements like those shown in Fig. 6.](image)


