Direct observation of magnetic flux and interstitial vortices in perforated mesoscopic squares of superconducting films

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Abstract. We report magnetic visualizations of fluxoid states in perforated mesoscopic squares of superconducting films with a scanning superconducting quantum interference device microscope. The observed magnetic images clearly reveal how the magnetic flux is distributed and trapped in a hole at different magnetic fields. The magnetization determined from the magnetic images exhibits a tilted sawtooth behavior with the magnetic field, indicating the multiple flux quanta trapped in the hole. Further increase of the magnetic field creates an interstitial vortex (vortices) between the hole and the sample edge. The penetration field $H_p$ of the interstitial vortex depends on the sample size and follows a simple relation $\mu_0 H_p \approx \Phi_0/w^2$ with the spacing $w$ between the hole and the sample edge.

1. Introduction
A mesoscopic superconducting ring provides an opportunity to investigate the fundamental property of fluxoid states in laterally modulated, perforated superconducting films. The fabrication of a regular array of antidots (holes) in superconducting films leads to unique anomalies in magnetic field dependences of superconducting transition temperature, critical current and magnetization at characteristic fields where the density of a magnetic flux lattice is harmonically related to the density of holes [1, 2, 3]. These are the hallmark features for the fluxoid quantization with magnetic flux quanta in holes and analogous to the Little-Parks effect in a superconducting cylinder (ring) [4]. The multiplicity of flux quanta is, however, limited by the radius $R$ of the hole with respect to the coherence length $\xi$ and characterized by the saturation number $N_s(\approx R/2\xi)$, above which vortices appear within interstices of the holes. In this case, additional anomalies occur due to the formation of supermatching flux-line lattices [5, 6, 7, 8].

When the array of holes is sparse, the situation is different. Irrespective of the saturation of magnetic flux quanta in holes, it is energetically more favorable to create (interstitial) vortices between holes than the multiple flux-quanta vortices in holes, provided that the density $n_c$ of holes is less than $\xi/R^3$ with the superconducting coherence length $\xi$ [9]. Moreover, as the
superconducting film becomes thinner, the self-energy and mutual interaction of vortices depend on their positions with respect to the size and shape of the film [10, 11]. As Brandt argued [12], in a thin film ring with inner $a$ and outer radii $b$, a vortex can sit in the middle of the annular region, rather than to enter the hole when applied magnetic field exceeds a size-dependent penetration field $\mu_0 H_p \approx \Phi_0/w^2$ with the annular width $w = b - a$. More exact analytical results for the penetration field have been given to the limited case of a very narrow ring with $w \ll a$ [13, 14] and thin narrow slabs [15].

The understanding of the size-dependent penetration of interstitial vortices is important for practical applications like the reduction of $1/f$ noise in superconducting quantum interference devices (SQUIDs) [16, 17], while experimental evidences for the penetration field were limited to thin slabs [18, 19]. In this study, we report direct observation of fluxoid states in perforated superconducting squares films with a scanning SQUID microscope. We observed nonmonotonic evolution of magnetic flux accumulated in the hole with the applied magnetic field. Further increase of the magnetic field results in the penetration of the interstitial vortex (vortices) in the annular region. We found that the penetration field $H_p$ of the vortex is size dependent and follows a simple relation $H_p \approx \Phi_0/w^2$ with the spacing $w$ between the hole and the sample edge.

| Table 1. Dimensions of perforated squares of amorphous Mo$_x$Ge$_{1-x}$ thin films |
|-----------------------------------|
| Sample  | Thickness  | Side length  | $R$  |
|         | ($\mu$m)  | ($\mu$m)    | ($\mu$m) |
| MoGe-A  | 0.22       | 26          | 3.5    |
| MoGe-B  | 0.42       | 55          | 4      |

2. Experimental
The samples we used mainly in this study were amorphous Mo$_x$Ge$_{1-x}$ (MoGe) films with $x \approx 75$ %. The parameters for these films are as follows. The superconducting transition temperature $T_c$ is $\approx 6.7$ K, normal resistivity $\rho_0$ (10 K) is $\approx 1.7$ $\mu\Omega$m, the slope $S$ of the second critical field $H_{c2}$ near $T_c$ is $\approx 2.2$ T/K. Using the dirty limit expressions, we estimated the zero-temperature magnetic penetration depth $\lambda(0) \approx 0.53$ $\mu$m and the zero-temperature coherence length $\xi(0) \approx 4.6$ nm [20]. We patterned the films into hundreds of small squares with different sizes and also fabricated holes in samples by ultraviolet lithographic and chemical etching techniques. Dimensions of two samples (A and B) we focus in this study are summarized in Table 1.

Magnetic images were taken with a scanning SQUID microscope (SQM-2000, SHI Nanotechnology). The microscope had a dc SQUID sensor chip integrating Nb-based Josephson junctions and an inductively coupled, pick-up Nb coil with 10 $\mu$m in diameter. For the safe operation of the microscope, we tilted the sensor chip with respect to the sample surface by $\sim 10$ degrees and kept the distance ($\sim 5$ $\mu$m) between the pick coil and the sample surface during the measurements. The lateral resolution of the microscope was $\sim 4$ $\mu$m, which allowed us to image a (Pearl) vortex ($\sim 10$ $\mu$m in diameter) in MoGe thin superconducting films with reasonable lateral resolutions [21]. The samples and the sensor were surrounded by $\mu$-metal. The ambient magnetic field around the sample space was typically $\approx 2$ $\mu$T. All the magnetic images presented in this study were taken on flux states at $T \approx 3.7$-3.9 K prepared by the field-cooled procedure. The details of the measurements are described in Refs. 21 and 22.
Figure 1. Magnetic images of fluxoid states observed in MoGe-A sample. (a) Micrograph of an identical sample to the one the magnetic images were taken. The sample size is 26 × 26 μm² with 0.22 μm in thickness. The radius of a centered hole is 3.5 μm. Images were taken at different applied magnetic fields of (b) 0, (c) 4, (d) 8, (e) 12, (f) 16, (g) 20, (h) 24, (i) 28, (j) 32, (k) 36 μT, respectively. All images are the same in size of 42 × 42 μm² (21 × 21 pixels). The color bar represents the magnitude of magnetic flux Φ_pixel over the area of 2 × 2 μm² divided by the flux quantum Φ₀ (the dark color corresponds to the low value of Φ_pixel). For each image the hole center is marked with a red cross. (l) Profiles of magnetic images (a)-(j) across the hole center as illustrated in the inset. For clarity, the field profiles are shifted vertically (Some of them are also shifted horizontally). The ambient magnetic field around the sample stage is ≈ 2 μT.

3. Results and discussions
Figures 1b-1k show magnetic images of fluxoid states of the MoGe-A sample at different applied magnetic fields. The sample size is 26 × 26 μm² with a centered circular hole (R = 3.5 μm). The color bar indicates the magnitude of magnetic flux Φ_pixel over the area of 2 × 2 μm² divided...
by the flux quantum $\Phi_0$ (the dark color corresponds to the low value of $\Phi_{\text{pixel}}$). For each image the hole center is marked with a red cross. One can see how the magnetic flux in the hole varies with the applied magnetic field. This behavior is also seen in a plot of magnetic profiles across the hole center shown in Fig. 11. At 4 $\mu$T, the “dome” of magnetic flux suddenly appears in the hole. As the magnetic field is elevated, the flux dome shrinks with respect to the field outside the sample, but it again becomes large at 16 $\mu$T. This nonmonotonic sequence continues up to 32 $\mu$T, above which an interstitial vortex(vortices) appears between the hole and the sample edge (Fig. 1k). We note that the image size of a vortex in the thin film is $\sim$ 10 $\mu$m in diameter (which corresponds to $\sim$ 5 pixels of the image), while the narrowest (widest) spacing between the hole and the sample edge is 9.5 $\mu$m (15 $\mu$m). Thus, the penetration of the interstitial vortex between the hole and the sample edge can be recognized in magnetic images by the high intensity of magnetic flux filled in the annular region of the sample. Therefore, we can distinguish the penetration of interstitial vortices in the annular region and the flux accumulation in the centered hole.

Figure 2. Magnetization curve of MoGe-A determined from magnetic images in Fig. 1. A solid line is a guide to the eye. The ambient magnetic field perpendicular to the sample is $\approx$ 2 $\mu$T as determined from the magnetization curve ($N = 0$) at $M = 0$.

Figure 2 shows the magnetization curve determined from magnetic images by following the procedure given in Ref. 22. As observed, the magnetization exhibits a tilted sawtooth behavior with the applied magnetic field up to 32 $\mu$T. Each magnetization jump indicates the entry of the quantized magnetic flux in the sample. Since the magnetic flux is accumulated in the hole without an interstitial vortex(vortices), we believe that the hole has the multiple flux quanta. The corresponding fluxoid number $N$ is indicated in Fig. 2.

With further increasing the applied magnetic field, the interstitial vortex (vortices) is created between the hole and the sample edge, while the fluxoid number in the hole remains constant. At this moment, we do not understand yet how the interstitial vortices should distribute in the annular region and also how the magnetization curve behaves. In the following, we focus on the first penetration of the interstitial vortex and determine the penetration field $H_p$ as the lowest magnetic field where the interstitial vortex appears in magnetic images. From the magnetic images taken in the two samples with different sizes we find that the penetration field $H_p$ is size dependent, i.e., $\mu_0 H_p = 36$ $\mu$T and 5 $\mu$T for the smaller (MoGe-A) and larger samples (MoGe-
B), respectively. The number of flux quanta, the fluxoid number \( N = 3(2) \), accumulated in the hole for MoGe-A(-B) is much smaller than the corresponding saturation number \( N_s \approx R/2\xi \approx 350 \) (400). Thus, the argument of the saturation in the hole is not relevant to the appearance of the interstitial vortex observed in this study.

Let us interpret the present results according to the Brandt model [12]. In Fig. 3 the results of \( H_p \) are plotted against the spacing \( w \) between the hole and the sample edge. Since there is no unique width for square loops, as illustrated in the inset, we take the widest spacing \( w_1 \) along the diagonal of the square (of which results are plotted by open symbols) and the narrowest one \( w_2 \) (which does with filled symbols). Here, the ambient magnetic field (perpendicular component determined from the magnetization curve in Fig. 2) is taken into account. Good accordance is seen between the results and theoretical curves representing relations \( \mu_0 H_p = C \Phi_0/w^2 \) with \( C = 3.9 \) and 1.8 for \( w_1 \) and \( w_2 \), respectively. In the Brandt model the coefficient \( C \) is of the order of unity. Thus, it is not possible to conclude which spacing should be relevant. A further experiment on superconducting rings with uniform annular width would clarify this issue.

![Figure 3](image)

**Figure 3.** Size dependent penetration field. Inset illustrates how we define the spacing \( w \) between the hole and the film edge. Open and filled symbols correspond to widest \( w_1 \) and narrowest spacings \( w_2 \), respectively. Square (circle) symbols indicate the results for amorphous MoGe square films (a polycrystalline Nb square film). Solid and dotted lines represent the Brandt relation with different coefficients [12].

Finally we comment on results made on a polycrystalline Nb square sample (\( T_c \approx 9.2 \) K) with a centered hole, of which size is nearly identical to the MoGe-B sample (\( R \gg \xi \)). We observed up to two flux quanta \( N = 2(< N_s \sim 100) \) accumulated in the hole and subsequent penetration of interstitial vortices in the annular region. As plotted in Fig. 3 with circle symbols, the results of the penetration field are in good accordance with the relation obtained in amorphous MoGe films. This agreement implies that the mechanism for the interstitial penetration is governed by the sample size, rather than the material parameters.

In summary, we have presented magnetic images of fluxoid states of perforated superconducting square films with the scanning SQUID microscope. We observed the accumulation of magnetic flux in the hole with increasing the applied magnetic field. The magnetization determined from the magnetic images exhibits the tilted sawtooth behavior with the applied magnetic field. These findings indicate the field evolution of multiple flux quanta
through the hole. With further increasing field, we observed the interstitial vortex (vortices) between the hole and the sample edge. The corresponding penetration field $H_p$ for the interstitial vortex is size dependent and follows the Brandt relation $\mu_0 H_p \approx \Phi_0 / w^2$ for both amorphous MoGe and polycrystalline Nb films.

Acknowledgments
N. K acknowledges K. Makise for providing us Nb films. This work was supported by JSPS KAKENHI (Grant Numbers 26287075 and 17K05537), the Inter-university Cooperative Research Program of the Institute for Materials Research, Tohoku University (Proposal No. 16K0004 and 17K0051), and NIMS Nanofabrication Platform in Nanotechnology Platform Project sponsored by the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan.

References
[1] Bezryadin A, Ovchinnikov Y N and Pannetier B 1996  Phys. Rev. B 53 8553
[2] Moshchalkov V V, Baert M, Metlushko V V, Rosseel E, Van Bael M J, Temst K, Bruynseraede Y and Jonckheere R 1998  Phys. Rev. B 57 3615
[3] Baert M, Metlushko V V, Jonckheere R, Moshchalkov V V and Bruynseraede Y 1995  Phys. Rev. Lett. 74 3269
[4] Little W A and Parks R D 1962  Phys. Rev. lett. 9 9
[5] Metlushko V, Welp U, Crabtree G W, Osgood R, Bader S D, DeLong L E, Zhang Z, Brueck S R J, Ilic B, Chung K and Hesketh P J 1999  Phys. Rev. B 60 R12585
[6] Field S B, James S S, Barentine J, Metlushko V, Crabtree G, Shtrikman H, Ilic B and Brueck S R J 2002  Phys. Rev. Lett. 88 067003
[7] Silhanek A V, Van Look L, Jonckheere R, Zhu B Y, Raedts S and Moshchalkov V V 2005  Phys. Rev. B 72 014507
[8] Ooi S, Mochiku T and Hirata K 2011  Physica C 471 804
[9] Buzdin A I 1993  Phys. Rev. B 47 11416
[10] Kogan V G 1994  Phys. Rev. B 49 15874
[11] Brandt E H 2005  Phys. Rev. B 72 024529
[12] Brandt E H and Clem J R 2004  Phys. Rev. B 69 184509
[13] Brojeny A A B and Clem J R 2003  Phys. Rev. B 68 174514
[14] Kogan V G, Clem J R and Mints R G 2004  Phys. Rev. B 69 064516
[15] Maksimova G M, 1998  Phys. Solid State 40 1607
[16] Dantsker E, Tanaka S and Clarke J 1997  Appl. Phys. Lett. 70 2037
[17] Wördenerweber R and Selders P, 2002  Physica C 366 135
[18] Stan G, Field S B and Martinis J M 2004  Phys. Rev. Lett. 92 097003
[19] Kokanović I, Helzel A, Babić D, Sürgers C and Strunk C 2008  Phys. Rev. B 77 172504
[20] Kes P H and Tsuei C C 1983  Phys. Rev. B 28 5126
[21] Kokubo N, Okayasu S, Kanda A and Shinozaki B 2010  Phys. Rev. B 82 014501
[22] Kokubo N, Okayasu S and Kadowaki K 2017  Multi-Vortex States in Mesoscopic Superconductors The Oxford Handbook of Small Superconductors ed Narlikar A V (United Kindam, Oxford University Press) chapter 3 pp 81-107