Intermediate vortex states in nanoscale anti-dots and mesoscopic superconductors

K Hirata, A Thakur, S Ooi and T Mochiku
National Institute for Materials Science, Superconducting Materials Center, 1-2-1 Sengen, Tsukuba, 305-0047, Japan
E-mail; HIRATA.Kazuto@nims.go.jp

Abstract. We have fabricated anti-dots array in Nb and NbN, and high-$T_c$ superconductor Bi$_2$Sr$_2$CaCu$_2$O$_y$ (Bi-2212), and have measured the flow-resistance of vortices perpendicular to the array and magnetic field. We could produce a variety of vortex-matching effect; the usual and the fractional matching. In low-$T_c$ superconductors, the usual matching effect shows "dips" in the flow resistance at the matching field. However, it shows "humps" at higher matching magnetic fields to be related to the formation of giant vortices and/or interstitial vortices. In Bi-2212, the matching effect is closely related to the first order transition of vortex lattice melting in the pristine samples in the presence of the anti-dot arrays. We also have measured the vortex state in mesoscopic type-I superconductor of In. This includes the observation of topological hysteresis with a signature of occurrence in different critical fields during vortex entry and exit. We will show the existence of a metastable configuration of vortices depending on the recipes to access them, and demonstrate the manifestation of superheating and supercooling of superconducting and normal states, respectively, across the superconducting transition.

1. Introduction
Recent progress in nanosize fabrication techniques enables us to study superconductivity in nanosize superconductors. Photo-, electron beam-lithography, and Focused Ion Beam (FIB) milling are the compromising methods for fabricating nanosize superconductors from bulk samples. Small size (submicron- to nanosize) superconductors can be obtained also by electrochemical deposition. To make the size of superconductors narrower and narrower, with decreasing dimensionality, there are anticipated a competition between the superconducting coherence length and the sample size. Theoretical proposals have been made about half a century ago [1-3]. They proposed that, reducing the dimensionality, superconducting phase slip may happen by thermal fluctuations, which was confirmed by the experiment with submicron size samples [4]. Recently, Bezryadin et al [5] have obtained experimental results of quantum suppression in superconductivity, and Lau et al [6] have measured nanosize samples with changing the cross-sectional area and have obtained the results of thermally-activated phase slip and quantum phase slip. which were made by depositing MoGe compound on the carbon nanotube. To make the superconductors narrower and narrower, it may suppress the superconductivity or sometimes creates new phenomena (macroscopic quantum tunneling, for example). Changing the size of superconductors from nanosize to mesoscopic size, several interesting
phenomena have been observed. Non-integer magnetic flux carrying has been observed in Al thin film disks [7]. Formation of giant vortex state has been observed [8]. Recently, Berdiyorov et al [9] have simulated the vortex configuration in type-I superconductors. The ground state of the vortex configuration depends on the sample geometry and magnetic field. Intermediate state of flux structure has been well produced in magnetization and free energy versus magnetic field. Here we present the experimental results on In nanowires encapsulated in insulating ZnS nanotubes [10, 11], which show the supercooling and superheating effects according to the magnetic field cycles.

On the other hand, fabrication of nanosize holes (anti-dots) or of putting nanosize magnetic dots is also possible by the nanosize fabrication, which can be used to study a matching effect of vortices in a magnetic field [12-16] and electronic properties in superconductors with anti-dots array [17-19]. In most of the experiments on the vortex physics in superconducting films/crystals with an anti-dots array, the vortices were considered as classical point defects with the pinning. However, the vortices are in fact extended objects which interact with the geometry of the underlying anti-dots lattice in many-body problem. This opens up the possibility for geometry and/or pinning strength (of the anti-dot lattice) induced novel vortex phenomena such as vortex merging, symmetry induced creation of vortex-antivortex configurations, coupling, rearrangement, etc, bringing into forefront the importance of interstitial vortices among the anti-dots also. Here we present the preliminary results of our investigations on the interplay between the vortices and the geometry of the underlying nano-engineered anti-dot lattice in Nb and NbN thin films [20]. The anti-dots are fabricated in the triangular or hexagonal shape with gradually increasing area of anti-dots such that one essentially goes over from a triangular anti-dots array to a honeycomb wire network.

The effect of the anti-dots array into high-\(T_c\) superconductors (HTSCs) is also one of the interesting subjects. In high-\(T_c\) superconductors, the first-order transition of vortex lattice melting (FOT-VLM) has been found [21]. This has been observed in relatively clean samples, which have intrinsically pinning centers caused by defects (imperfections of crystals; impurities, crystalline defects, etc). Then, VLM occurs at relatively higher temperatures below \(T_c\), because, at lower temperatures, the pinning strength becomes stronger, and the VLM is mediated from the first order transition to the second order or sometimes disappears. Introducing the anti-dots array into HTSCs, it is interesting to investigate how the FOT-VLM will be changed, as the competition might be occurred between the vortex lattice and anti-dots array. We present here the experimental results on \(\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_y\) (\(\text{Bi}-2212\)) with a triangular array of anti-dots, and will show broad and steep steps at the supercooling transition depending on the magnetic field, in addition to the fractional matching effect [22].

2. Experimental

In nanowires were synthesized with ZnS nanotubes carried out in a high temperature vertical induction furnace. The details are described in Ref. 10&11. The average wire diameter was 120nm, and the length 5-20\(\mu\)m, while the average external diameter of the shell ZnS nanotube was about 200nm, which is shown in figure 1 Superconducting transition temperature \(T_c\) was 3.41K, which is close to the bulk \(T_c\). Bundle of the In nanowires in almost aligning to the magnetic field was measured to put into Quantum Design SQUID MPMS XL, and the magnetization was measured.

The anti-dot array samples of Nb and NbN films were prepared on a Si substrate [23, 24]. Initially, 100nm thick films were deposited by sputtering [20]. A probe pattern was made via photolithography followed by ion beam etching for a typical four-probe measurement with region between the voltage probes typically being 30\(\mu\)m x 30\(\mu\)m wide. The etching was performed at a typical etching rate of 0.5 nm/sec in Argon plasma [20]. Under these etching conditions, there was essentially no damage to the Nb and NbN films. In these 30\(\mu\)m x 30\(\mu\)m wide available regions, anti-dot arrays networks were patterned using the FIB milling technique utilizing an aperture of 50\(\mu\)m and a typical ion-beam current of 13pA. The transport measurements were carried out with the conventional four-probe technique using a home-made insert which goes into the Quantum Design SQUID MPMS XL. The data acquisition was made with the external device control (EDC) option to operate the Keithley 6430 sub-femtoamp remote sourcemeter and the Keithley 2182A nanovoltmeter.
An additional temperature sensor was placed very close to the sample in the insert to be read with a Cryocon 62 AC resistance bridge.

In HTSC Bi-2212, high-quality single-crystals of Bi-2212 were grown by the travelling-solvent floating zone technique [25]. Thin single-crystal films with submicron thickness were prepared through repeating cleave by adhesive tapes, which is the characteristic of Bi-2212 to easily cleaved in the $ab$-plane. After thinning the crystals sufficiently on the MgO substrate, thin Au layer (20nm) was deposited on the surface as electrodes. The films were patterned for four-terminals electric resistance measurements using a photolithography and an Ar-ion milling processes. Equilateral triangular array of anti-dots was fabricated by FIB milling using Micrion JFIB-2100 to the bridge part of films between the voltage electrodes. The superconducting transition temperature of the sample determined from zero resistance with a current of 10μA was 89.0K. In the measurements, the magnetic field was always applied perpendicular to the $ab$-plane.

3. Results and Discussion

3.1 Intermediate state of In nanowires

Figure 1 shows a TEM image of In nanowire encapsulated in ZnS nanotube. Inset of figure 1 shows the selected-area electron diffraction pattern at the point indicated by arrow, which shows that the nanotube used in the measurements is a single crystalline structure. For the magnetization measurements, the bundle of the nanotubes was used. It was confirmed that the In nanowires belong to the type-I nature, showing nearly the same magnitude of the critical magnetic field with the $M-H$ loops of the bulk In. Difference between nanowire and bulk In lies in that of critical fields with field sweep-up and sweep-down, and attributes to the phenomenon of supercooling and superheating of intermediate vortex state mentioned later.

![Figure 1](image1.png)

*Figure 1.* TEM image of the In nanowire encapsulated in ZnS nanotube. Inset shows the selected area electron diffraction pattern at the point indicated by arrow.

![Figure 2](image2.png)

*Figure 2.* A part of the magnetization loop (solid black lines) in the field sweeping at 2.5K. The arrows show the field sweep directions. Minor loops also indicated in the colour loops. The dotted arrows show the initial to the final states (open symbols) of the loops. The inset shows changes in magnetization values in the field-shaking process (see the text).

Figure 2 shows the result of magnetization measurements in $M-H$ loops, which partially indicates a $M-H$ loop and minor loops with the field-shaking process. The field-shaking process was made during the magnetization measurements with sweeping the field up and down by an amount of $\Delta H$, which are shown in the coloured loops. The inset shows a plot in changes in magnetization values with...
the field-shaking process, in which a peak can be seen as a maximum at intermediate field of about 1100e.

In the temperature-shaking process shown in figure 3, it also shows an intermediate state of vortices. The colour scale contour in figure 3 is plotted for the evolution of magnetization values from the field cooling state for various temperature-shakings, in which the colour scale from red to blue indicates the superconductivity becomes stronger. The final state in figure 3 (b) corresponds to the starting state in the panel (a). In this case, as shown in figures 3 (a) for a negative temperature-shaking and (b) for a positive, a low temperature superconducting phase is superheated and a high field normal state is suppressed, respectively. In these hysteresis observed, there are several factors to contribute; pinning centers, geometrical barriers, topologically, superheating and supercooling across the boundary between the superconducting phase and the normal state, etc. Among of them, topological hysteresis may understand the results by assuming the presence of a surface barrier during flux entry and exit. These intermediate vortex state may be related to the intermediate vortex configurations published recently [9].

Figure 3. Colour scale contour plot of the evolution of magnetization from the field cooled state following the field-cooling temperature-sweeping process with negative temperature-shaking (a) and positive (b) at 2.5K.

3.2 Vortex state with anti-dots array in Nb and NbN
As the vortex flow-resistance measurements in Nb film with regular array of anti-dots have been well made and studied [13, 26], here we present on the experimental results on NbN film, which has a relatively higher $T_c$ in metallic superconductors and has no reports to measure on the vortex matching effect to the anti-dots array in detail. Figures 4 (a) and (b) show the typical results of the flow-resistance versus normalized magnetic field $f (=H/H_m$, $H_m$ is the matching field to the array of anti-dots) measured with a current $I_{dc}=100\mu$A on the samples with changing a temperature and with a temperature $T=0.932T_c$ with changing a current $I_{dc}=1-150\mu$A, in which the scanning ion beam images are shown in the insets. In figure 4 (a), dip structures are clearly seen at integer number of the matching field $f$ up to $8^\circ$. This can be observed in the sample of the triangular anti-dots array [20]. The maximum number of vortices captured by an anti-dot with a diameter $d$ is given by the saturation number ($n_s$) such that, $n_s=\pi d/4\xi(t)$, where $\xi(t)$ is the coherence length at reduced temperature $t (=T/T_c)$ given by $\xi(t)=\xi(0)(1-t)^{1/2}$, with $\xi(0)$ being the zero temperature coherence length about 5nm. In the square
anti-dots arrays, \( n_s \) lies in the range 3.05-2.90 for \( t \) in the range of 0.925-0.923, which is far from the matching periods. So, the multi-vortex states do not seem to exist in the observations. Instead of that, it may be understood in the supermatching flux-line lattices model formed by a reorganization of vortices that enter the interstitial sites at the fields larger than \( n_s H_1 \) [15, 16]. To confirm the model, we fabricated a hexagonal lattice of anti-dots missing the center anti-dot as shown in figure 4 (b). In this case, we can observe the integer matching effect up to the saturation number 3. However, above 3\(^{rd}\) matching, the dip structure changes to enhance the flow-resistance, resulting to form the hump structure. This is understood that, up to the saturation number of vortices, they form multi-vortex state pinned at the anti-dots. However, exceeding to the number, the next vortices come to the center of hexagonal anti-dots, become interstitial vortices and mobile, and these weekly pinned vortices enhance the flow-resistance. The role of the interstitial vortices must be studied carefully from analytical thermodynamic calculations taking the interaction among the vortices and anti-dots into account.

![Figure 4](image-url)

**Figure 4.** Flow-resistance versus a normalized magnetic field \( f (=H/H_1, H_1 \) is the matching field to the array of anti-dots) obtained with a current \( I_{dc}=100 \mu A \).

The inset shows the Scanning Ion Beam image of the square anti-dots lattice \( H_1=169 \text{Oe} \) with a pitch of 350nm and a diameter of 180nm for (a), and of the hexagonal anti-dots lattice \( H_1=99.6 \text{Oe} \) with a pitch of 400nm and a diameter of 170nm for (b).

### 3.3 Vortex state with anti-dots array in Bi-2212

To introducing anti-dots array into Bi-2212 is a challenging issue, as there have been few report except into YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\). Scanning-ion beam image of the anti-dots in Bi-2212 between the voltage electrodes is shown in figure 5, fabricated by FIB. The diameter of the anti-dots is 300nm, the pitch is 1.0\( \mu \)m, and the matching field \( H_0 \) is about 23.9Oe. Figure 6 shows the results of the flow-resistance measurements. At the matching field \( H_0 \), the flow-resistance shows a dip structure as seen in low temperature superconducting film (figures 4 (a) and (b)). Furthermore, fractional matching effect can be clearly observed. Experimentally, we obtained the fractional matching at 8/9, 6/7, 3/4, 2/3, 1/2, 1/3, 1/4, 1/7, 1/9\( H_0 \). The fractional matchings of 1/3, 1/4, 1/7, 1/9\( H_0 \) can be explained well with commensurate matching of the Abrikosov vortex-lattice to anti-dots arrays. The 1/3\( H_0 \) matching is most remarkable, which corresponds to match to the second nearest neighbor anti-dots. However, 1/2\( H_0 \) matching is hard to explain at present, which may be related to the interstitial vortices as mentioned in previous subsection.

Another point of view to study vortex states in the anti-dots array in Bi-2212 is that, in Bi-2212 pristine sample without anti-dots, as the FOT of VLM has been observed (which has also been confirmed in this experiment before the fabrication), it is very interesting to see what it will be.
Figure 7. Scanning-ion beam image of the anti-dots in Bi-2212 between the voltage electrodes, fabricated by FIB. The diameter of the anti-dots is 300nm, and the pitch is 1.0μm.

Figure 8. Flow-resistance versus magnetic field measured on the sample shown in figure 1. Fractional matching can be clearly seen symmetrically against magnetic field direction.

Figure 9. TEM image of the In nano-wire encapsulated in ZnS nanotube. Inset shows the selected area electron diffraction pattern at the point indicated by arrow.

4. Summary
We have fabricated anti-dots structure into low- and high $T_c$ superconductors and have measured the flow-resistance of vortices. We could produce a variety of vortex-matching effect; the usual and the fractional matching. In low-$T_c$ superconductors, the usual matching effect shows "dips" in the flow resistance at the matching field. However, multi-vortex state explains hardly at higher matching magnetic field. We have confirmed that interstitial vortices play a role to accommodate more vortices, as it has been shown as "humps" at higher matching magnetic fields. In Bi-2212, the matching effect is closely related to the first order transition of vortex lattice melting in the pristine samples in the presence of the anti-dots arrays. We also have measured the vortex state in mesoscopic type-I superconductor of In. This includes the observation of topological hysteresis with a signature of occurrence in different critical fields during vortex entry and exit. We showed the existence of a metastable configuration of vortices depending on how to access them, and demonstrate the manifestation of superheating and supercooling of superconducting and normal states, respectively, across the superconducting transition.

References

[1] Hohenberg P C 1967 Existence of long-range order in one and two dimensions Phys. Rev. Lett. 158 383
[2] Langer JS and Ambegaokar MR 1967 Intrinsic resistive transition in narrow superconducting channels Phys. Rev. 164 498
[3] McCumber D E and Halperin B I 1970 Time scale of resistive fluctuations in thin superconducting wires Phys. Rev. B 1 1054
[4] Newbower R S, Beasley M R and Tinkham M 1972 Fluctuation effects on the superconducting transition of Ti whisker crystals Phys. Rev. B 5 864
[5] Bezryadin A, Lau C N and Tinkham M 2000 Quantum supression of superconductivity in ultrathin nanowires Nature 404 971
[6] Lau C N, Markovic N, Bockrath M, Bezryadin A and Tinkham M 2001 Quantum phase slips in superconducting nanowires Phys. Rev. Lett. 87 217003
[7] Geim A K, Dubson S V, Grigorieva L V, Novoselov K S, Peeters F M and Schweigert V A 2000 Non-quantized penetration of magnetic flux in the vortex state of superconductors Nature 407 55
[8] Kanda A, Baelus B J, Peeters F M, Kadowaki K and Ootsuka Y 2004 Experimental evidence of giant vortex states in a mesoscopic superconducting disk Phys. Rev. Lett. 93 257002
[9] Berdiyorov G R, Hernandez A D and Peeters F M 2009 Confinement effects on intermediate-state flux patterns in mesoscopic type-I superconductors Phys. Rev. Lett. 103 267002
[10] Sheet G, Gautam U K, Thakur A D, Hirata K, Bando Y and Nakayama T 2009 Clean superconducting In nanowires encapsulated within insulating ZnS nanotubes Appl. Phys. Lett. 94 053108
[11] Thakur A D, Gautam U K, Sheet G, Nakayama T, Bando Y, Goldberg D, Ooi S and Hirata K 2010 History and metastability effects in the intermediate state of mesoscopic type I superconducting Indium cond-mat.super-con arXiv:1001.3903v1
[12] Fiory A T, Hebard A F and Somekh S 1978 Critical currents associated with the interaction of commensurate flux-line sublattices in a perforated Al film Appl. Phys. Lett. 32 73
[13] Harada K, Kamimura O, Kasai H, Matsuda T, Tonomura A and MOshchalkov V V 1996 Direct observation of vortex dynamics in superconducting films with regular arrays of defects Science 274 1167
[14] Martin J I, Veliez M, Nogues J and Schuller I K 1997 Flux pinning in a superconductor by an array of sibmicroneter magnetic dots Phys. Rev. Lett. 79 1929
[15] Martin J I, Veliez M, Hoffmann A, Schuller I K and Vincent J L 1999 Artificially induced reconfiguration of vortex lattice by arrays of magnetic dots Phys. Rev. Lett. 83 1022
[16] Metlushko V, Welp U, Crabtree G W, Zhang Z, Brueck R J, Watkins B, Delong L E, Chung B
IIIc K and Hesketh P J 1999 Nonlinear flux-line dynamics in vanadium films with square lattices of submicron holes Phys. Rev. B 59 603

[17] Baturina T 2007 Nano-size antidots TiN thin film Phys. Rev. Lett. 98 257003

[18] Mironov A Y, Baturina T, Vinokur V M, Postolova S V, Kropotin P N, Baklanov M R, Nasimov D A and Latshev A V 2010 Disorder and vortex matching effects in nanoporous ultrathin TiN film Physica C to be published

[19] Baturina T, Mironov A Y, Vinokur Chchelkatchev N M, Glatz A, Nasimov D A and Latshev A V 2009 Resonant Andreev reflection in two-dimensional array of SNS junctions Physica C to be published

[20] Thakur A D, Ooi S, Chockalingam S P, Jesudasan J, Raychaudhuri P and Hirata K 2009 Vortex matching effect in engineered thin films of NbN Appl. Phys. Lett. 94 262501

[21] Zeldov E, Majer D, Koczykowski M, Geshkenbein V B, Vinokur V M and Strikman H 1995 Thermodynamic observation of first-order vortex-lattice melting transition in Bi₁₂Sr₂Ca₂Cu₂O₈ Nature 375 373

[22] Ooi S, Mochiku T and Hirata 2009 Fractional matching effect in single-crystal films of Bi₁₂Sr₂Ca₂Cu₂O₈+y with antidot lattice Physica C 469 1113

[23] Chokalingam S P, Chand M, Jesudasan J, Tripathi V and Raychaudhuri P 2008 Superconducting properties and Hall effect of epitaxial NbN thin films Phys. Rev. B 77 214503

[24] Chokalingam S P, Chand M, Kamlapure A, Jesudasan J, Mishra A, Tripathi V and Raychaudhuri P 2009 Tunneling studies in a homogeneously disordered s-wave superconductor: NbN Phys. Rev. B 79 094509

[25] Mochiku T, Hirata K and Kadowaki K 1997 Crystalline improvement of Bi₁₂Sr₂Ca₂Cu₂O₈ single crystal by TSFZ method Physica C 282-287 475

[26] Thakur A D, Ooi S and Hirata K 2009 Triangular antidot array to honeycomb wire network – Role of onterstitial vortices Physica C 469 1071