The phase boundary of superconducting niobium thin films with antidot arrays fabricated with microsphere photolithography

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Abstract

The experimental investigation of the $I_c(B)$–$T_c(B)$ phase boundary of superconducting niobium films with large area quasihexagonal hole arrays is reported. The hole arrays were patterned with microsphere photolithography. We investigate the perforated niobium films by means of electrical directed current transport measurements close to the transition temperature $T_c$ in perpendicularly applied magnetic fields. We find pronounced modulations of the critical current with applied magnetic field, which we interpret as a consequence of commensurable states between the Abrikosov vortex lattice and the quasihexagonal pinning array. Furthermore, we observe Little–Parks oscillations in the critical temperature versus magnetic field.

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

1. Introduction

Nowadays superconducting thin films are used for a huge variety of superconducting microelectronic devices such as Josephson junctions, superconducting quantum interference devices (SQUIDs) and coplanar waveguide resonators. Typically, these thin films are made of type-II superconductors and are penetrated by quantized magnetic flux when operated in magnetic fields or when biased with sufficiently high currents. The investigation of these magnetic flux lines (Abrikosov vortices) and their individual and collective interactions with natural and artificial defects in the superconductor is of high interest and has been subjected to many experimental and theoretical studies for several decades now.

One reason for this sustained scientific attention is that unpinned Abrikosov vortices respond with a dissipative motion to any current flowing in their vicinity. In many cases this motion is directly related to a reduction of the performance of the microelectronic devices (increased noise, lowered quality factor, shortened coherence time). Defects, however, act as local energy minima (pinning sites) and are able to reduce or even completely suppress vortex motion and the related dissipation [1–5]. For instance, it has been demonstrated that the flux noise in SQUIDs and the dissipation in coplanar microwave resonators can be reduced by strategically positioned microholes (antidots) [6, 7].

A second and more fundamental point is associated with the fact that an ensemble of Abrikosov vortices interacting with an ensemble of defects constitutes a highly designable
model system for repulsively interacting particles in a two-dimensional potential landscape. In such systems it is possible to investigate static effects such as the formation of quasicrystals [8–12] or the controlled introduction of potential landscape disorder [13–15] as well as dynamic effects such as mode locking phenomena [16–18] and ratchet dynamics [19, 20].

Of particular interest in both research branches is the case, when the typical length scales of the defect topology, i.e. size and mutual distance, are comparable to the intrinsic length scales of the superconductor, namely the coherence length $\xi$ and the magnetic penetration depth $\lambda$, which are both temperature dependent and diverge at the critical temperature $T_c$. Well below the critical temperature, $\lambda$ and $\xi$ can usually be found in the micro- to nanometer range. The patterning of large areas of superconducting films with submicron-scaled high density arrays of defects constitutes a non-trivial challenge to standard optical lithography (limited by resolution) or electron beam lithography (limited by time).

It has been demonstrated with different approaches that taking advantage of self-assembling structures can provide a way out of the difficulties in covering large areas with tiny structures on reasonable timescales. The techniques used vary from using anodized aluminum as substrate material [21], to over depositing the superconducting film on a layer of microspheres [22], to generating structures by inverse diblock copolymer micelle formation [23]. These fabrication methods, however, are limited to certain substrate materials or they induce changes in the substrate properties and/or the properties of the superconductor.

Here we adopt another method to fabricate large area quasihexagonal arrays of submicron sized antidots which is independent of the substrate material and does not influence the superconducting material more than any standard lithography process [24, 25]. In a previous study we have demonstrated that with this fabrication technique it is possible to reduce the vortex associated losses in superconducting microwave resonators by more than one order of magnitude [26]. In this paper we analyze the properties of our microsphere patterned Nb thin films by means of transport measurements close to the transition temperature with a particular focus on signatures of commensurabilities between the antidot and vortex lattices. We also investigate the transition between the wire network and the thin film regime in our samples. The critical parameter for this transition is the coherence length $\xi(T)$, which in the network regime is larger than the width $W$ of the superconductor between the antidots and which is smaller than $W$ in the thin film regime.

2. Experimental details

We fabricated our samples by first depositing a $t = 150$ nm thick niobium film on an r-cut sapphire wafer by dc magnetron sputtering. Afterwards we cut the wafer into individual chips and carried out the lithography steps. For the fabrication of perforated samples the chips were covered with photoresist and on top of that with a monolayer of water suspended polystyrene colloids in a Langmuir–Blodgett deposition process [27]. The microspheres have a diameter of $D_s = 770 \pm 25$ nm and act as a self-assembled array of UV-light focusing microlenses, leading to a quasihexagonal hole array after the exposure, their removal and the resist development. For a perfect, hexagonal close-packed array with $D_s = 770$ nm one would obtain a corresponding hole density of $n_h \approx 1.95 \mu m^{-2}$. In reality, however, one could expect deviations from the ideal packaging due to disorder during the self-assembling.

After transferring the hole array into the Nb film via reactive ion etching (SF$_6$) we patterned cross-shaped bridge structures for electric transport characterizations into the films. For this we used standard optical shadow-mask lithography and another SF$_6$ reactive ion etching step. Figure 1(a) shows one of the bridge structures with a square center area of $200 \times 200$ $\mu$m$^2$. The antidots have an approximate diameter of $D_a = 370$ nm, which in principle can be easily varied by adjusting the lithography exposure time. In figure 1(b) an atomic force microscopy image of the niobium film with antidots is depicted, which shows a domain-like pattern of holes with dislocations and imperfections.

To quantify the amount of missing antidots, we chose twelve different sections such as the one shown in figure 1(b) and counted the antidots. By doing so we found a mean antidot density of $n_a \approx 1.55 \mu m^{-2}$ with a rather large

![Figure 1. (a) Optical image of the $800 \times 800 \mu m^2$ large cross-shaped bridge structure with a square center area of $200 \times 200 \mu m^2$ for the four-probe current voltage characterization of superconducting thin films with pinning landscapes. (b) Atomic force microscopy (AFM) image of a $30 \times 30 \mu m^2$ large section of the niobium film with the microsphere patterned quasihexagonal array of antidots; the inset shows a zoom-in. (c) Autocorrelation function $I/I_0$ of the large AFM image in (b); the arrow indicates the position and direction of the linescan shown in (d) (blue solid line) together with corresponding linescans of two different $30 \times 30 \mu m^2$ array sections (red dashed and green dotted lines); for the definition of $I$ see text.

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variance between \( n_0 \approx 1.28 \) and \( 1.78 \, \mu m^{-2} \). That means that approximately 20% of the antidots are missing as compared to the perfect lattice with \( n_0 \approx 1.95 \, \mu m^{-2} \).

Another interesting analysis of the lattice is related to the typical correlation length of the antidot array. For this purpose we assigned each single pixel of figure 1(b) the value \( k \) to the perfect lattice with \( n \approx 1 \). We also patterned and characterized bridges with 100 \( \times 100 \, \mu m^2 \) and 50 \( \times 50 \, \mu m^2 \) large center squares, but the experimental results showed no dependence on the bridge size. Note that the niobium chips were taken from the same wafer as the chips for our previous study on resonators [26].

To characterize our samples we mount them into a low-temperature setup, that provides a temperature stability of \( \Delta T < 1 \, mK \), and contact them electrically with wire bonds. We apply a magnetic field perpendicular to the film plane using a superconducting coil, and monitor current voltage characteristics (IVCs) for many values of magnetic field and temperature. After collecting all IVCs we extract the desired information as the critical current \( I_c \), the critical temperature \( T_c \) or the resistance \( R \) versus magnetic flux density \( B \) and temperature \( T \). We choose the threshold voltage \( V_c \) defining \( I_c \) and the measurement current \( I \) for the resistance \( R \) during the evaluation. To reduce the voltage noise, we take several IVCs at each value for \( B \) and \( T \) and post-process the raw data (averaging and smoothing), such that we are more sensitive to modulations of the \( I_c-T_c \) phase boundary with respect to the applied magnetic flux.

3. Results and discussion

As an overview of the whole phase boundary figure 2 depicts (a) the critical current \( I_c \) and (b) the resistance \( R \) versus magnetic flux density and temperature of one of our samples.
Figure 3. Critical current $I_c$ versus magnetic flux density $B$ of a superconducting thin film with a quasihexagonal lattice of antidots; different curves correspond to different temperatures $T$. The flux density axis is normalized to the first matching flux density $B_1 \approx 3.4$ mT.

The critical current $I_c(B)$, the critical temperature $T_c(B)$ and the resistance $R(B)$ strongly modulate with the applied flux density. There are several ‘canyons’ and ‘ridges’ indicating commensurate states between the flux line lattice and the hole array.

For a more detailed view of the positions of the maxima and minima in the 3D phase boundary, it is convenient to extract single parameter slices. Figure 3 shows several individual curves for $I_c(B)$, which correspond to vertical cuts through figure 2(a) for constant temperatures. The flux density axes in figures 2 and 3 are normalized to the first pronounced maximum in the critical current $B_1 \approx 3.4$ mT. This corresponds to a vortex density of $n_v = B_1/\Phi_0 = 1.65 \mu$m$^{-2}$, which is well below the ‘ideal’ hole density of $n_h \approx 1.95 \mu$m$^{-2}$ and slightly above the real antidot density of $n_a \approx 1.55 \mu$m$^{-2}$ determined by counting. Despite the difference between $n_v$ and $n_a$, we think that a plausible explanation for the reduced matching field as compared to the ideal one is that at $B = B_1$ the matching is rather local and predominantly determined by the hole density.

The difference between $n_v$ and $n_a$ might be explained by some vortices at interstitial positions in larger antidot-free areas and/or by antidots, which are already doubly occupied at $B = B_1$. In a previous study the effect of randomly diluting a periodic pinning array has been investigated systematically [14]. In these experiments a similar effect was observed. The first matching maximum in some samples with diluted pinning arrays was found at field values between the one corresponding to the hole density and the one corresponding to the ideal hexagonal lattice.

For higher magnetic field values we find three more matching features, cf figure 3. The first of these is found for $B \approx 2B_1 = B_2$ and most likely corresponds to two vortices per pinning site. Then there are two shoulder-like bumps for $B \approx 3.5B_1$ and $5B_1$ but none at $B = 3B_1$ and $4B_1$, which is rather unconventional and on first sight surprising.

For a perfect triangular pinning lattice, one would expect always matching features at integer multiples of the first matching field. They can correspond to a multiple occupation of the pinning sites or to composite triangular vortex lattices in antidots and at interstitial positions. In the case of interstitial, ‘caged’ vortices it might happen that one of the regular peaks is suppressed or missing [31–34], but to our knowledge it has never been reported that two consecutive ones are missing as in our case ($3B_1$ and $4B_1$) and that an additional one appears in between instead ($3.5B_1$).

We think that the peak structure of the phase boundary of our samples can be explained by a related but somewhat different mechanism. With increasing field values, the mean artificial pinning strength decreases and the vortex-vortex interaction becomes more important due to the increasing vortex density. In this situation, the vortices tend more and more to form a perfect hexagonal lattice with a homogeneous density and thus fill up the positions of missing antidots. As the vortex lattice as a whole, however, is still interacting with the pinning sites in this case, the length scale of the pinning array determines the energetically most favorable length scale of the vortex lattice. Then the matching effects might appear at integer multiples of the ‘ideal’ pinning lattice with $n_v = 1.95 \mu$m$^{-2}$. Strikingly, the values of $3.5B_1$ and $5B_1$ with $B_1 \approx 3.4$ mT correspond quite well to $3B_1^* = 4B_1^*$ with $B_1^* = n_0 \Phi_0 \approx 4$ mT. Again, a similar effect has been observed in randomly diluted pinning arrays before, where for a dilution of 20% the first matching peak corresponded more to the real antidot density and the second matching peak to the lattice constant [14].

Finally, it has been observed that the artificial introduction of disorder into a periodic pinning lattice leads to broadened matching peaks [15]. This observation also supports our interpretation of local matching for low vortex densities, as the peaks at $B = B_1$ and $B_2$ are rather sharp and the pinning lattice is locally highly ordered. When the matching becomes non-local at higher vortex densities, however, the global disorder of the pinning lattice becomes important and broadens the peaks at $B = 3.5B_1$ and $5B_1$.

Besides analyzing the phase boundary by taking horizontal slices, we can also take vertical slices for chosen currents or voltages of the 3D boundary and end up with the critical temperature versus magnetic field plots (or second critical magnetic field versus temperature, respectively) for different $T_c$ ($B_{c2}$) criteria. Figure 4 shows a plot of $T_c(B)/T_c(0) = T_c/T_{c0}$ of a perforated sample (symbols) for a resistance criterion of $R_c/R_n = 0.5$ and measured with an applied current of $I = 50 \mu$A. $R_n$ denotes the normal state resistance at $T = 10$ K. Oscillations of the critical temperature with the applied flux are clearly visible, which we associate with Little–Parks oscillations [35], as already observed in many studies on superconducting wire networks and thin films with pinning arrays [36–39].

We also plot the critical temperature versus magnetic field of a plain reference sample in figure 4(a) and calculate from these data the coherence length $\xi(0) = 16$ nm of the niobium by fitting it to the bulk expression $B_{c2} = \phi_0/[2\pi \xi(T)^2]$ with $\xi(T) = \xi(0)(1 - T/T_{c0})^{-1/2}$ [40]. As the coherence length
Reduced critical temperature $T_c/T_{c0}$ versus normalized magnetic flux density $B/B_1$ for two reasons. First, the holes have a circular shape and, second, some of them are missing. So it might even be that the patterning method has hardly influenced the properties of the niobium.

We have performed experiments close to $T_c$, but when using superconductors with a higher magnetic penetration depth, the results are also relevant at temperatures of $T = 4.2$ K or even in the mK regime. This situation occurs in very thin or dirty superconducting films and in different superconducting materials, although in some of these cases (e.g. YBa$_2$Cu$_3$O$_7$) the artificial pinning will compete with strong intrinsic pinning. In principle our patterning technique could be used with even smaller spheres [24], which would lead to commensurability effects at much lower temperatures and higher magnetic fields. In these cases, the critical current and the pinning efficiency would modulate with the applied field in a similar manner to that presented, which has to be considered for the design of possible devices.

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