Observation of magnetic flux pinning in a thin Nb film with a square lattice of nickel dots

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Abstract

The pinning of magnetic flux by a square lattice of ferromagnetic Ni dots embedded in a superconducting Nb film was investigated. In the case of demagnetized Ni dots, both AC susceptibility and DC magnetization data exhibit clear anomalies at the first and second matching fields, \( H_{f1} = 13 \) Oe and \( H_{s2} = 2H_f \), respectively. The second matching anomaly is clearly observed in both AC and DC data only when a remanent magnetization of Ni dots is induced along the external magnetic field, which is oriented perpendicular to the Nb film plane. The observed behavior supports theoretical predictions of a substantial contribution to the flux pinning energy by anisotropic interactions between the superconducting condensate of Nb and the Ni dot magnetization.

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1. Introduction

The enhancement of vortex pinning is crucial for the success of many technological applications of superconductors. One of the novel approaches to enhance pinning in a controlled way is to pattern superconducting films with ordered arrays of cylindrical holes called “antidots” [1,2]. A perfect lattice of antidots is known to induce clearly defined “matching” anomalies in the magnetization and electrical resistivity at applied fields for which the equilibrium number of Abrikosov vortices (“flux lines”, FL) in the mixed state coincides with an integral multiple \( n \) of the number of antidots. The antidot diameter \( D \) and the coherence length \( \xi(T) = \xi_c(1 - T/T_c)^{-1/2} \) determine the maximum or “saturation number” \( n_s \) of flux quanta that may be trapped by each antidot [3]:

\[
n_s = D/4\xi(T).
\]

The value of \( n_s \), and therefore the overall pinning strength of the superconducting film, can be controlled by varying the temperature \( T \) or magnetic field \( H \) [4]. Additional flux penetrating the film will appear as FL repelled into the interstitial region between the antidots, resulting in the coexistence of two subsystems: (i) multi-quantum fluxoids are localized at the antidot sites, and (ii) weakly pinned,
single-quantum vortices or “interstitial flux lines” (IFL) reside in the interstices and dominate the dissipative behavior of the film. Recent experiments [5] and computer simulations [6,7] have revealed that the IFL exhibit interesting dynamics and “supermatching” flux line lattice (FLL) structures for \( n > n_c \). Therefore, opportunities exist to explore novel film patterning techniques for the quantitative control of FLL pinning and electromagnetic dissipation in superconducting devices and materials.

Another promising approach for the control of FLL pinning is to incorporate regular arrays of ferromagnetic particles into superconducting thin films such that magnetic field or temperature may be varied to adjust the magnetization of the particles and vary the overall pinning properties and dissipative behavior of a film. Otani et al. [8] demonstrated that a square array of ferromagnetic particles deposited on top of a thin Nb film results in an oscillatory field dependence of the film magnetoresponse when the particles are magnetized in-plane, whereas the oscillations entirely disappeared when the array was demagnetized. This result indicates the importance of controlling the orientation of the magnetization of the particles in order to modulate the FLL pinning. Different behavior has been observed [9] for Nb films completely covering a lattice of relatively thin ferromagnetic Ni dots (i.e., right-circular cylinders). Even though matching anomalies in the film magnetoresponse were found, no effect of the magnetization of dots on pinning properties was registered, and they found no indication of pinning periodic in applied field in superconducting films with arrays of nonmagnetic dots. These results were interpreted as evidence of spatial modulation of the superconducting order parameter by the action of a ferromagnetic proximity effect. Other preliminary results indicate that the existence and strength of matching anomalies in the case of nonmagnetic dots depends upon the thickness of the superconducting film and/or its overlap with the dot lattice [10–12].

In contrast to previous studies, the effects of lattices of Ni dots that completely penetrate a superconducting Nb film were investigated in magnetoresponse measurements [12,13]. The aspect ratio (height to diameter) of the dots was substantially higher than in previous investigations, which favors the retention of some Ni dot magnetization perpendicular to the Nb film plane. Indeed, a large asymmetry in critical current was observed when the Ni dipoles were aligned or anti-aligned with respect to an externally applied magnetic field, which suggested that an anisotropic dipole–vortex interaction [14] contributes to the net pinning force exerted by the dots.

All previous investigations [8–13] of superconducting films patterned with arrays of magnetic dots reported matching anomalies in the film magnetoresponse. To exclude any possible electrode contact effects and minimize other finite-current effects on the results, we performed AC susceptibility and DC magnetization measurements on Nb films with square Ni dot lattices synthesized during previously reported transport studies [12,13]. DC magnetization data were also acquired to directly characterize the Ni dot magnetization at temperatures slightly above \( T_c \), where the relatively large magnetic response of the superconducting state of Nb is absent.

2. Experimental procedure

Our sample materials consist of a Si substrate supporting a square lattice of cylindrical Ni dots (120 nm diameter, 110 nm tall) surrounded by a Nb film of 95 nm thickness. The Ni dots thus completely perforate the film and have a discontiguous Nb cap on their top surface [12]. The overall dimensions of the film were 1 mm × 1 mm. The Ni lattice was patterned via electron-beam lithography using polymethylmethacrylate (PMMA) photoresist. Ni was then deposited by electron-beam evaporation, followed by lift-off of the PMMA in acetone, leaving behind bare Ni dots on the Si substrate. No effort was made to remove any oxide barrier on the Ni dots prior to Nb deposition. The transition temperature \( T_f = 8.8 \) K in zero magnetic field for all samples, with and without Ni arrays, which implies no macroscopic reduction of \( T_c \) due to magnetic or other proximity effects is active. From \( H_{c2} \) measurements [12,13], the mean free path in the dirty limit is \( l \approx 4.5 \) nm. A more detailed description of sample fabrication is given elsewhere [12,13].

DC magnetization and AC susceptibility measurements were performed using a Quantum Design MPMS5 SQUID magnetometer with the external
magnetic field aligned perpendicular to the plane of the film. Demagnetization of Ni dots was accomplished by oscillating the direction of the applied DC field with a gradual decrease of amplitude, starting at $H = +1.0$ T, while the sample was held at room temperature. The superconducting magnet then was reset (quenched to eliminate any trapped magnetic flux) while the sample was held out of the magnetometer. After demagnetization the sample was cooled in zero field and isothermal field dependencies of the DC magnetization and AC susceptibility were taken at several temperatures. Where needed, the Ni dots were magnetized at 10 K by applying external magnetic fields up to 3 T, followed by a reset of the magnet.

3. Experimental results

DC magnetization data for both a Nb film with demagnetized Ni dots, and an unperforated Nb film, both held at $T = 8.7$ K, are shown in Fig. 1a. The patterned film exhibits well defined matching anomalies at $H_1 \approx 13$ Oe, $H_2 \approx 26$ Oe, and a very weak but detectable anomaly at $H_3 \approx 39$ Oe. It is important to note that the matching anomalies are symmetrically placed (within ±1 Oe) with respect to zero field, which implies that demagnetized Ni dots pin flux in essentially the same manner as antidots [15]. Further, an overall increase in the magnetization hysteresis width, compared with the plain Nb sample, is evident. Since both the patterned and plain Nb samples have similar geometries and sizes, the substantial increase in the hysteresis width exhibited by the patterned sample is indicative of an enhancement of the critical current density $j_c$. However, the enhancement of $j_c$ is observed only at low fields, whereas at fields above $H_c$ the enhancement of $j_c$ vanishes, suggesting that magnetic dots provide little improvement in flux pinning in this field regime.

We find that matching anomalies are well defined only at temperatures close to $T_c$, similar to the results of other investigations [1,2,4,16,5,8–15,17] of superconducting films perforated by antidot or magnetic dot lattices. Fig. 1b shows that the matching effects smear out and eventually disappear at temperatures below 8.3 K. However, the plain Nb sample exhibits a relatively low hysteresis width, showing that the Ni dots still contribute to the overall pinning at lower temperature. Further, decreases in temperature lead to the appearance of magnetic instabilities of the patterned film in the form of abrupt magnetization jumps. The first instabilities are observed at $T = 7.9$ K (see Fig. 2) and they apparently persist at all temperatures investigated between 7.9 and 2 K. It is interesting that at 7.9 K instabilities are observed only on descending branches of the DC magnetization curve, while the ascending branches are smooth. A more complete study of the instability effects will be published elsewhere [18].

DC magnetization data taken at temperatures just below $T_c$ are shown in Fig. 3a for the patterned sample shortly after the Ni dots were magnetized at
Fig. 2. DC magnetic moment vs. applied magnetic field for a Nb film with demagnetized Ni dots held at a fixed temperature of 7.9 K. Note the abrupt jumps in magnetic moment observed for decreasing field amplitudes.

10 K. These results differ from the behavior of magnetization data for the demagnetized sample (compare with Fig. 1a) in that the magnetization curves are not symmetric with respect to zero field. When the sample was polarized in positive magnetic field (Fig. 3a), two clear matching anomalies were observed at \( H_1 \approx 13 \) Oe and \( H_2 \approx 26 \) Oe, as found for the demagnetized sample (see Fig. 1a). On the other hand, only the first matching anomaly was observed, and the second matching anomaly (expected at \( H_{-2} \approx -28 \) Oe) was not evident in the negative field range. The mirror image of this behavior was observed when the sample was polarized in negative applied field, as shown in Fig. 3b. Note that there is subtle evidence for a third matching anomaly near 39 Oe in the demagnetized case, and in the negative field range for the reverse-magnetized sample data of Fig. 3b.

The field dependence of the AC magnetic moment \( m_{AC} \) at \( T = 8.78 \) K (only 0.02 K below \( T_c \)) is shown in Fig. 4 for both demagnetized and magnetized samples, as well as for the plain Nb film. At this temperature the pinning is very weak and we employed the AC technique, since it is a more sensitive method compared with DC magnetization. The demagnetized sample exhibits matching anomalies at positive fields near \( H_1 \) and \( H_2 \), as shown in Fig. 4a. Note that the second matching anomaly is very weak in the real part, and it is best resolved in the imaginary part of the AC data. The signature at \( H_2 \) in the real part (current response) of the moment for the magnetized sample data of Fig. 4b is more apparent in the positive field range. A very weak third matching anomaly is also present in the imaginary (dissipative) part in the positive field range. Similar to the DC magnetization data, only the first matching anomaly at \( H_1 \approx 13 \) Oe is observed when the magnetizing field was anti-aligned with the DC sweep field. It is evident that the matching anomalies are much better defined in the case of a sample magnetized in the same direction as the field sweep, and the anomalies are suppressed for fields swept in the opposite direction, whereas the strength of the anomalies of the demagnetized sample is somewhere in between the two magnetized cases. The slight asymmetry (\( \approx 1 \) Oe) in the placement of the matching anomalies is due to trapped flux in the superconducting magnet. These data document the effect of the Ni dot magnetization on flux pinning, and are a major result of this work.

Fig. 3. (a) DC magnetic moment vs. applied magnetic field for a Nb film with magnetized Ni dots held at a fixed temperature of 8.7 K. At least two matching anomalies are clearly observed when the positive applied field is aligned along the remanent magnetization of the Ni dots. (b) Only the first matching anomaly is easily observed at positive applied fields for the case of the remanent magnetization anti-aligned with the applied field. Observed matching fields are indicated by dashed vertical lines.
In order to directly characterize the magnetic states of the Ni dots, we performed magnetization measurements of our samples at the temperature of 10 K, which is slightly above $T_c$, and thus no effect of superconductivity is present. To our knowledge, this is the first direct measurement of the transverse (with respect to the Nb film plane) magnetization of a regular array of submicron, ferromagnetic dots embedded in such a film. After an initial ferromagnetic response at the lowest applied DC fields, the sample becomes diamagnetic, indicating the ferromagnetic saturation of Ni dots and a background diamagnetic response of the substrate (see Fig. 5a). We obtained an estimate of the magnetization curve for the Ni dots by subtracting the extrapolated linear diamagnetic component of the signal, and the result is shown in Fig. 5b, which exhibits typical ferromagnetic behavior with a saturation field of about 1 T. The remanent moment of the dots after the field was returned to zero from the maximal value of 3 T is about 10% of the saturation moment, as shown in Fig. 5c. Thus we conclude that after applying a magnetizing field of the order of tesla and removing it at $T = 10$ K, Ni dots conserve approximately 10% of their saturation magnetization in the direction of the magnetizing field, and this is responsible for the observed magnetic pinning effect evident in the data of Fig. 4.

![Fig. 4](image)

![Fig. 5](image)
Two small, symmetrically placed, sharp decreases of magnetization are evident at fields of about 0.7 and −0.7 T. The magnetization curves appear noisy at fields higher than about 1.5 T, especially in the case of a virgin magnetization curve, and noticeable scatter of the data is found above magnetization values near half of the saturation value. The origin of these anomalies is presently unclear.

Previous investigations [8–13] of magnetic flux pinning in superconducting films patterned with regular arrays of magnetic particles employed only electrical transport measurements, which raises a question of whether or not the static pinning interaction between flux and the particles, or dynamic interactions between moving vortices and the particles, are responsible for the matching effects. Indeed, matching anomalies in magnetoresistance were found to exist only for intermediate transport currents, while the anomalies tend to disappear both at higher and lower currents, leading to a suggestion that random background pinning plays a dominant role at low velocities, masking the pinning of the ordered magnetic dot lattice [9].

It is therefore important to study the matching anomalies in the Bean critical state, which is a metastable, static configuration of pinned FL in the presence of a uniform distribution of critical current flowing in a persistent mode throughout the sample. Thus, complementary to previous studies [8,9,13] in which matching anomalies were observed only for moving vortices, our results (see Figs. 1, 3 and 4) show clearly that Ni dots induce magnetic pinning and matching anomalies in the establishment of a metastable critical state. Matching anomalies are expected for a square lattice of dots at field values given by

\[ H_m = n \Phi_0 / d^2 \]  
(2)

where \( H_m \) is the matching field, \( \Phi_0 \) is flux quantum, and \( d \) is the lattice spacing between the dots. The samples studied have a Ni dot density \( 1/d^2 = 1.2 \mu m^{-2} \), and by using Eq. (2), we calculate \( H_m = n \times (14 \text{ Oe}) \), which is in good agreement with our experimental data. An application of experimental parameters [9,13,15] to Eq. (1) yields estimates of \( n = 1 \) to 3, depending upon the temperature \( T < T_c \), which is consistent with the data shown in Figs. 1, 3 and 4; note that these estimates do not include magnetic pinning [14] or proximity effects of the Ni dots, but only the attractive effect of a normal hole in the Nb film at the dot sites.

We now return to our observation that the establishment of a remanent magnetization of the Ni dots by applying an external magnetic field \( H = 3 \text{ T} \) at \( T = 10 \text{ K} \) leads to substantial enhancement of the second matching anomaly in an applied DC field aligned parallel with the polarizing field, and complete suppression of second matching anomaly when the applied DC field is anti-aligned with the remanent polarization vector. These results are consistent with previous observations [12,13] of matching anomalies in transport critical current for films prepared in a manner similar to the present study. Since the magnetization of the Ni does not change the measured values of any matching anomalies, the overall effect of the dots is not to simply bias the average internal field within the Nb film. However, the asymmetric nature of the magnetization curves for magnetized dots suggests the microscopic vortex configuration is determined by an anisotropic interaction between the FL and the local magnetic field generated by the magnetized dots. We consider the interaction energy \( E = - \mu \cdot H \) between a magnetic dipole \( \mu \) and an external magnetic field \( H \). Since the self-field of a FL increases with decreasing distance from the FL center, \( E \) is minimized by the FL moving closer to a stationary magnetic moment which is aligned parallel to the vortex field. Therefore, the force is attractive for the aligned Ni dot case, and there is an energy penalty for the FL to be near an antiparallel moment, and there is a repulsive interaction between FL and anti-aligned Ni dots. An early Gibbs free energy argument [19] showed that the force between a magnetic moment embedded in a superconductor and a FL is attractive. However, additional interactions must be considered to gain an accurate picture of the magnetic pinning effects of magnetic dots.

Numerical solutions [14] of the Ginzburg–Landau equations have been obtained for the field and current distribution around a magnetic moment aligned perpendicular to the plane of a superconductor. The pinning force between a FL and an embedded magnetic dipole consists of at least three contributions: (1) the interaction between the magnetic field generated by the FL and the magnetic moment, (2) the
interaction between the FL and the superconducting screening currents created in response to the magnetic dipole, and (3) the interaction of the FL core with an area of reduced superconducting condensation energy due to the absence of superconducting material within a dot and the proximity effect of the Ni in an annulus surrounding the dot. The first two interactions depend on the relative orientation of the FL and the dipole, and should lead to the enhancement of the second matching anomaly when the applied field and the magnetic moment are aligned, and a suppression of this anomaly in the anti-aligned case.

By analyzing matching anomalies in the transport critical current [13] it was conjectured that the first and second matching anomalies in the Nb film with embedded Ni dots correspond to the one fluxoid-per-dot and two-fluxoid-per-dot configurations, respectively. This expectation is supported by previous observations [16] of only weak “supermatching” anomalies observed at applied fields above the saturation value $H_s$ at which additional FL can only enter the interstitial region between ordered antidots supporting multi-quantum fluxoids. Because of the local electromagnetic energy ($=-\mathbf{B} \cdot \mathbf{H}$) and the repulsive nature of the long-range part of the interaction between an IFL and the current distribution around a doubly quantized Ni dot, the double-fluxoid state should be less stable than the single-fluxoid configuration [13]. Indeed, our data (see Fig. 3) strongly suggest that an anisotropic dipole–FL interaction energy can significantly enhance the stability of a double-fluxoid state in the field-aligned case, and strongly destabilizes it in the anti-aligned case (note that a similar observation could be made about a very weak third matching anomaly in Fig. 3b). Thus the overall stability of the FL configuration in the film can be very effectively controlled by altering the alignment of the dots, and this opens the possibility of designing patterned superconducting films with magnetically switchable pinning for device applications.

The weak, third matching anomaly observed in DC magnetization data for the demagnetized sample may correspond to a “supermatching” FLL phase consisting of a doubly quantized fluid on the Ni dot and one IFL [5,15,16]. However, drawing correspondences between observed matching anomalies and simple IFL topologies assumes that the average magnetic field is homogeneous across the sample. Alternatively, a multi-terrace structure for the critical state of films patterned with lattices of antidots has been predicted [20] as a compromise between the linear decrease of field strength (Bean profile) with distance in a strong-pinning thin film with no patterning, and the energy gain provided by a single-terrace, well ordered FLL realized at matching fields close to $T_c$, where pinning of the IFL is weak [16,21]. The overwhelming majority of published evidence for matching anomalies is representative of this high temperature regime.

However, if the film is in the critical state at temperatures well below $T_c$, the external field is shielded by metastable supercurrents that are accompanied by an inhomogeneous distribution of magnetic field, and numerical simulations [22] suggest that the flux profile in the critical state can be quite complicated. In this case only small domains of a sample film should satisfy a particular matching condition, and the overall magnetization of the film may, or may not exhibit well defined matching anomalies. We observe the disappearance of matching anomalies as the temperature goes down (see Fig. 1b), and a previous study [23] reported a similar washing out of matching anomalies at lower temperatures for regular arrays of antidots. There is presently no reliable experimental data documenting the existence of multi-terrace structures in superconductors with lattices of artificial defects.

The multi-terrace structure is characterized by narrow regions of extremely high current boundary supercurrent interlaced with areas of perfectly matched FL, and the magnetization curve of a patterned superconductor should exhibit periodic jumps corresponding to the creation of new terraces having different flux densities in the modified critical state. The sharp jumps in magnetization curve observed in our study at lower temperature (see Fig. 2) may indicate a competition between the commensurability effect of the periodic Ni dot lattice and a formation of a more conventional Bean-like field profile within the film. Even though we cannot identify field periodicity of the magnetization jumps at these temperatures, they may indicate the local formation of terrace-like structures, whose size may be limited by strong extrinsic pinning by nonperiodic...
defects and reduced interactions (i.e., shorter penetration depth) between FL at temperatures well below $T_c$. This hypothesis, however, does not rule out random thermomagnetic instabilities as a possible mechanism for the magnetization jumps, such as recently observed in unpatterned Nb films [24]. These questions will be examined in more detail in a separate publication [18].

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