Observation of magnetic-field penetration via dendritic growth in superconducting niobium films

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Using a high-resolution magneto-optical technique, we have studied the penetration and expulsion of magnetic flux in superconducting niobium thin films. We have found that over a wide region of the phase diagram, the picture of uniform flux fronts as described by a critical-state model breaks down and that the penetration of the field takes place through the growth of magnetic dendrites. Magnetic decoration studies with single vortex resolution show that the dendrites have a complex structure and consist of walls of both high and low magnetic field and are filled with a state of matter which structurally appears to be a vortex fluid. Some of the dendrites are seen to grow by nucleating regions with hundreds of antivortices in the film.

It is well known that flux penetration into a type-II superconductor can occur in a catastrophic fashion. The energy dissipation caused by such vortex motion can even drive the sample into the normal state. These flux jumps[1] have received much attention in the past, due to their relevance to applications, since stability is of crucial importance in the operation of many devices. However, in general they have not been studied using techniques with high spatial resolution which could allow one to examine in detail the nontrivial patterns that can be formed. On the other hand, studies of patterns[2] in the magnetic flux distribution in superconductors which have been done have generally been restricted to equilibrium configurations such as those observed in the intermediate state, while out of equilibrium distributions are usually considered only in terms of a Bean[3] or other kind[4] of a critical state model. In general, the analysis of data from bulk measurement techniques such as magnetization and susceptibility experiments often makes assumptions about the structure of the magnetic response which can be unwarranted in the face of careful, spatially resolved studies.[5–8]

In this paper, we report on a detailed study of out of equilibrium patterns of flux penetration into thin, superconducting niobium films. We find that the universally accepted description in terms of a critical-state model breaks down over a wide region of the phase diagram and that rather than the penetration of flux by uniform flux fronts, as described by a Bean model,[3] the field penetrates through the sudden nucleation of magnetic dendrites whose structure and growth appear similar to that seen for dielectric breakdown in insulators. The dendrites are found to have a complex morphology which ranges from quasi-one-dimensional structures at $T/T_c \sim 0.35$ with little branching to highly branched, "sea-weed-like" structures when approaching $T/T_c \sim 0.65$. Above $T/T_c \sim 0.65$ the penetration is via relatively smooth flux fronts whose gross pattern can be described by a critical-state type of model, however, a careful examination of the images show evidence of coarsening of the fronts as they propagate. The dendrites themselves are wondrously complex structures with walls of alternating high and low magnetic field regions: decoration studies show them to be filled with a high-density vortex fluid. Even less expected than the dendrites themselves is the fact that the walls of the dendrites can have large regions containing several hundred antivortices. To our knowledge, the observation that a type-II superconductor can respond to an applied magnetic field by nucleating a large number of vortices opposite in sign to the applied field is unprecedented. The dendrites appear to grow very quickly in comparison to our temporal resolution of several milliseconds, strengthening the analogy of the phenomenon we observe to the dielectric breakdown of insulators. We find that the nucleation of these large instabilities can be suppressed by raising the temperature of the sample.

The majority of the experimental results to be presented here were obtained using a high-resolution magneto-optical technique. The technique works by placing a magneto-optical thin film in close proximity to the superconductor to be studied and using polarized light to measure the local magnetic field in the film which mirrors the magnetic response of the superconductor. The maximum spatial resolution afforded by the technique is given by a convolution of the distance between the superconductor and the film, the film thickness and the wavelength of the light we use to prove the film. In the present experiments, to optimize this, we have followed the lead of Schuster *et al.*[8] and evaporated a thin film of EuSe directly onto the sample. Our cryogenic system is a conventional optical cryostat with both liquid helium and nitrogen jackets. The sample is mounted on an oxygen-free high-conductivity copper cold finger with appropriate thermometry and temperature control mounted close to the sample. The magneto-optical film is imaged using a conventional polarized light microscope, which is mounted horizontally with positioning stages for scanning the experiment. The cryostat was carefully designed to allow the sample to remain isothermal and aligned independent of the level of the cryogens in the apparatus allowing for good long-term stability. The entire system was mounted on a high-quality optical table to minimize the influence of vibrations at even the highest magnifications. The images were directly digitized using a computer system and a cooled charge-coupled device (CCD) chip[9] with 12 bit dynamic range gray scale and excellent linearity and low dark current figures. Our final spatial resolution is ~ 0.5 μm. We have also used Bitter decoration imaging using

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the same apparatus as described previously[10] to allow us to resolve individual vortices within the dendrite structure itself. This technique is complementary to the magneto-optical technique as it has a higher spatial resolution but obviously cannot look at individual nucleation processes.

The samples studied in this experiment were pure niobium thin films grown by dc magnetron sputtering in Argon from 8-inch diameter targets with the substrates positioned 230 mm above the target.[11] The system was cryopumped to a base pressure of 5×10^{-9} Torr and the evaporations were done with an Argon partial pressure of 3.5 mTorr with a niobium deposition rate of 1.7 nm/s. These were high-quality films used to produce excellent, strain-free Nb-Al_2O_3-Nb junctions with $T_c(0)=9.1$ K and a critical current of $\sim2\times10^6$ A/cm^2 at 4.2 K. We have carried out observations on films grown on three different kinds of substrates, glass microscope cover slips, single crystal quartz plates and oxidized silicon wafers. The films grown on the silicon substrates are known to be strain-free. We find the same features for a number of films grown on all three substrates giving us confidence that we are probing an intrinsic phenomenon rather than an extrinsic effect due to sample inhomogeneities.[12]

Shown in Fig. 1 is the main result of this paper. In Fig. 1(a) is shown the remnant magnetic field distribution for a sample which was cooled from above T_c in the presence of a field of 135 Oe. At 7.42 K the temperature was stabilized and the field was subsequently removed. In the photo, the darker regions represent more intense local fields while the lighter regions correspond to lower fields. The observed distribution is qualitatively what would be expected from conventional critical-state models. The vortices leave the sample from the edges giving a "rooflike" field distribution whose slope is given by the critical current density J_c. There are some inhomogeneities in the distribution due to the intrinsically random nature of the pinning but in general, the gross features agree with one's expectation. A more careful study of the flux front does show evidence of a coarsening of the front as it propagates (this is visible in the photo). A detailed study of this will be published elsewhere.

Figure 1(b) shows the response of the same film at 5.97 K. The contrast with Fig. 1(a) is evident. The pattern is much more complex with a critical-state component near the edges of the sample but also with large dendritic structures with significant branching. These structures are not at all predicted in a critical-state model and suggest that there exists a magnetic instability as the field goes to zero. Shown in Fig. 1(c) is the result of a measurement at 3.30 K where the character of the dendrites has changed to more one-dimensional structures. The evolution of these patterns with temperature is continuous with it being rapid near 5.8 K and still evolving, albeit slowly, at our lowest temperature of 3.3 K.

The observed dendritic structures have a number of remarkable properties. They always nucleate very rapidly regardless of the rate that the external field is changed. We were not able to measure the nucleation time but an upper limit is several milliseconds which is the frame refresh time of our CCD. We believe that the actual time is much faster.[6] Once the nucleation takes place, the whole structure is frozen in place and does not evolve upon further changes in the applied field. When the applied field is changed sufficiently, a new dendrite will appear similar in character to the others but different in detail. It is possible to cover the sample with a dense, superimposed tangle of dendrites. The new dendrites do not lie directly on the pattern of previous ones copying their shape but have complicated shapes of their own. This explicitly shows that the phenomena is not due to a macroscopic pattern of weak links in the sample. As further evidence for this, we find that identical experimental conditions produce patterns which are similar in their overall features but

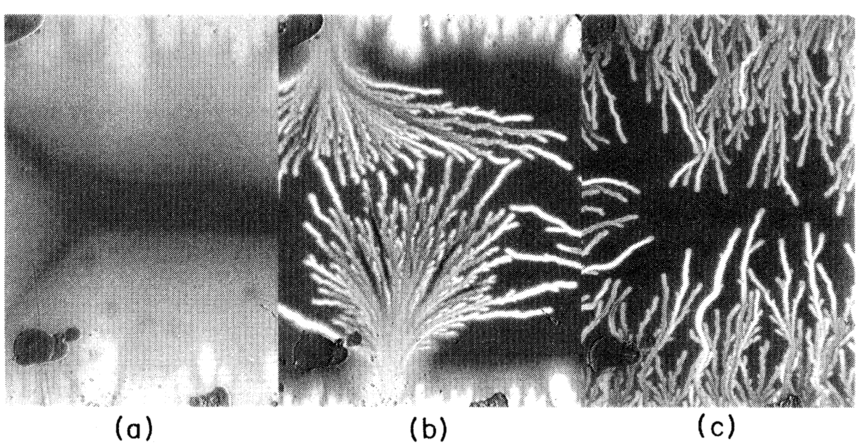

FIG. 1. Shown is the magnetic response of a thin niobium film as measured using the magneto-optical technique. Film thickness is 0.5 μm, and its lateral dimensions are ≈3(height)$\times8$(width) mm^2. The field of view spans approximately 25% of the area of the sample, between the top and bottom edges, and two vertical lines ≈2 mm apart. The field of view lies closer to the left edge, as is clear from the "Y" shaped profile in part (a). In (a) is shown the response at 7.42 K, in (b) is shown the response at 5.97 K and in (c) is shown the response at 3.30 K. As the temperature is decreased the penetration of the magnetic flux changes from a critical-state-like response to one in which the penetration proceeds through the growth of magnetic dendrites.

different in detail showing that it is not an extrinsic effect. Patterns obtained on increasing or decreasing the field show some differences in the amount of branching but the dendrites occur for both types of field histories. The structures that we see can be compared to those in Refs. 13 and 14. The experiment by DeSorbo and Newhouse[13] is similar to ours, but with a spatial resolution which is less by roughly two orders of magnitude. Aside from this limitation, there is a strong similarity between the two sets of data, which lead us to believe that their samples were probably in the type-II regime, due to the shortening of the mean free path in thin evaporated films. The same argument holds for the decoration pictures taken by Dolan.[14] Both these studies were done at a fixed temperature, so they failed to detect the interesting evolution that we observe. They also failed to detect antivortices.

We have also done experiments in which we have slowly ramped the field at rates of a few hundredths of an Oe/sec which allows us to individually monitor each nucleation process. By subtracting subsequent frames just before and after a nucleation we have observed that the nucleation does not necessarily occur at the edge of the sample however each branch stops at the middle of the sample—they never seem to carry on to the other side. This rules out simple surface barrier effects as the origin of the dendrites. The stopping of the dendrites at the middle of the sample shows that demagnetization is the driving force. A simple statistical analysis seems to indicate that the number of events $N\Phi$ with a given amount of flux Φ is roughly independent of Φ up to a certain cutoff.

The dendrites themselves have a very complex structure, an example of which is shown in Fig. 2. The branches have walls of high and low field regions and are filled with a relatively uniform, high field region. This further suggests a dynamic origin for the dendrites. For some of the dendrites, there are even regions in which sizable numbers of antivortices are nucleated. This can be seen quite clearly in Fig. 2 where the regions composed of antivortices are colored in red. Each of these patches contain several hundred antivortices. The observation of an inverse local field in a superconductor in response to an applied field which was monotonically increased from zero at a constant temperature is completely outside of the framework of a critical-state model.

We will now discuss the possible relationship of the phenomena we have observed to the mixed-intermediate state.[15] The mixed-intermediate state is the response of a type-II superconductor such as niobium which is similar to the intermediate state of a type-I superconductor. In very clean single crystals of niobium the unusually long mean free path allows for large values of the coherence length ξ, bringing the Ginzburg-landau parameter κ very close to the value $1/\sqrt{2}$. Close to this limit which defines the boundary between type-I and type-II superconductors, it has been shown that at low fields an attractive interaction between vortices develops which leads to a number of interesting observations. There are a number of arguments which suggest to us that the phenomena which we describe in this paper is not due to this effect. For our films, κ is well above the value $1/\sqrt{2}$ because

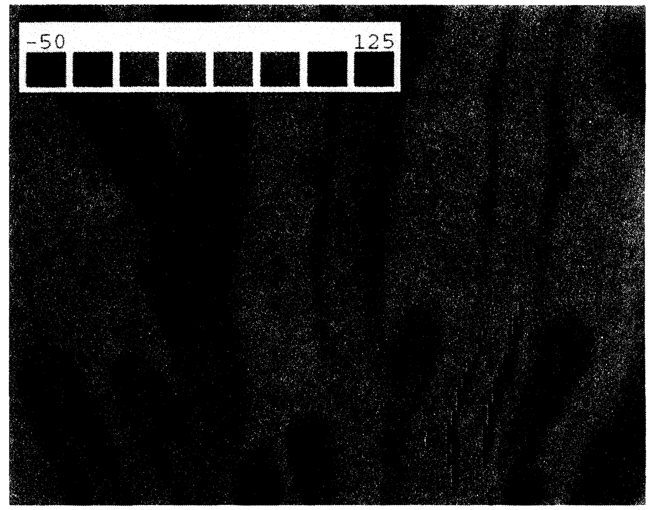

FIG. 2. Shown is an enlargement of a region of dendritic growth. For this structure, the field is applied in such a way as to produce the dark regions of dendritic growth. The regions of the dendrite in which antivortices are nucleated are shown in red. This data was taken at 5.9 K. The image corresponds to an area of 275×230 μm^2, and the magnetic units in the colored scale are Gauss.

they are thin films with a moderate mean free path. An estimate can be made for κ from the measured resistivity, using the dirty limit equation[16] $\kappa=\kappa_0+7.53\times10^3\rho\gamma^{1/2}$, where κ_0 is the clean limit value, ρ is the normal state resistivity in Ω cm, and γ is the electronic specific heat coefficient in ergs/cm^3/K^2. Using our measured $\rho=1.7$ $10^{-6}\Omega$cm, and a lower estimate of $\gamma\approx10^4$ ergs/cm^3 K^2, we get $\kappa=2$, well within the type-II regime. Second, the structures are observed only upon changes in the applied field at low temperatures. Third, the attractive vortex interaction responsible for the intermediate-mixed state fixes the value of the local field in the penetrated regions at a value given by the optimum lattice parameter. This is not observed in our experiments where the field intensity of the structures is controlled by the external field at the moment at which the structure nucleates. We have seen this in both the magneto-optical experiments where the different gray levels gives us a measure of the field scale of the dendrites as well as more microscopically in the Bitter decorations in which different field scales in the dendrites are given by differing vortex separations. Finally, the mixed-intermediate state could never result in a field inversion as shown in Fig. 2.

Shown in Fig. 3 are two images of dendrites taken using Bitter decoration. The upper frame shows a large scale picture of the overall dendrite structure and the lower shows a dendrite at a larger magnification where one can see the individual vortices which comprise the structure. Seeing the dendrites with Bitter decoration makes it clear that the magneto-optical results are not an experimental artifact. The decorations also show that the magnetic field in the dendrites is penetrating the sample as vortices and that different field intensities correspond to different vortex densities as is usually the case for

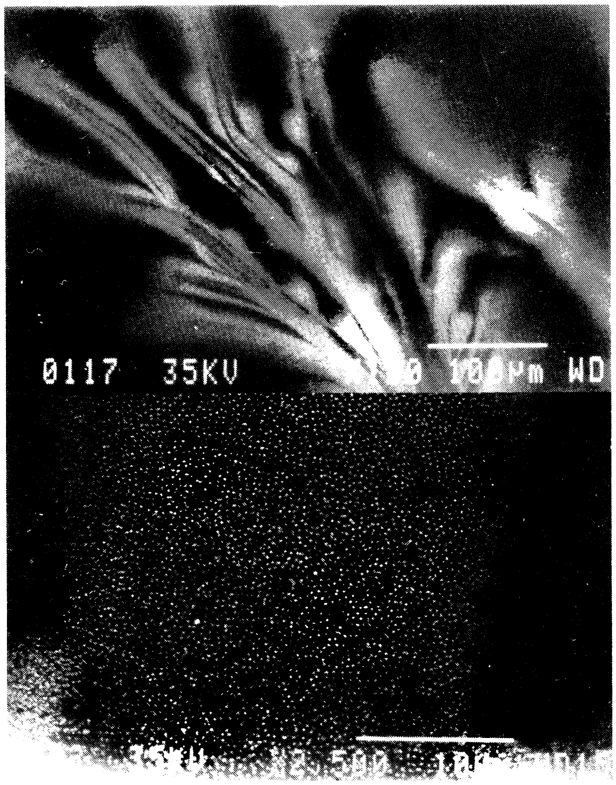

FIG. 3. Shown are the results of magnetic decoration studies of the dendrite growth. The upper figure shows a large area of the film and the lower figure is an expanded scale in which it is possible to see the individual vortices which make up the dendrite. The scale bars represent 100 and 10 μm for the top and bottom panels, respectively. This data was taken after cooling the sample to 4.2 K.

type-II superconductors. A comparison between the two techniques shows that the vortices are singly quantized with the normal value of flux, $hc/2e$. We do not see any evidence of a different kind of penetration such as lamellae or vortex clusters which would result from bound states of vortices in the intermediate-mixed state (for an example, see the images in Ref. 15). The lower frame of Fig. 3 shows clearly that the vortices in the dendrites are disordered on a very short length scale. The Fourier transforms show a liquidlike structure factor with no sixfold modulation evident and rapidly decaying correlation functions. From a structural point of view, the dendrites look as though they are filled with a vortex liquid. However, we cannot argue that this lack of order is evidence that the dendrites are due to the sudden quench of a penetrating vortex fluid as similarly disordered patterns are seen in these films for decorations done in field-cooled homogeneous magnetic fields.

In a recent paper, Leiderer *et al.*[6] have seen magnetic structures in thin films of Y-Ba-Cu-O that are similar to some of those reported here. In those experiments, they nucleated the structures by using a laser pulse fired at a localized spot on the sample and presumably heating it up above T_c. The data in Ref. 6 were explained by a model in which the normal state propagated at the expense of the superconducting one. Such an explanation does not seem to be appropriate for our observations, given the strong temperature dependence of the nucleation process, and its suppression at high temperatures. In addition, such a model cannot explain the appearance of antivortices. Either the origin of that effect and this one are different or the model they propose is incorrect. Generally speaking, the strong temperature dependence argues against a thermal runaway as a possible explanation for our observations.

In conclusion, we have used both high-resolution magneto-optical imaging and Bitter decoration to study the magnetic penetration into thin films of niobium. We have found that over a wide region of the phase diagram the critical-state model breaks down and that the field penetration proceeds via nucleation of dendrites. In addition to dendritic growth we have also observed roughening of the flux fronts in the region of the phase diagram approximately described by the critical-state model. We also see that the dendrites can grow by nucleating large regions of antivortices. The value of $\kappa \geq 2$, the presence of antivortices, the lack of an observation of vortex clusters, and the intrinsic nonequilibrium nature of the dendrites are evidence against an interpretation in terms of the mixed intermediate state in a marginally type-II superconductor. An interpretation in terms of a thermal runaway into the normal state does not seem viable in view of the strong temperature dependence of the effect, with its suppression at higher temperatures.

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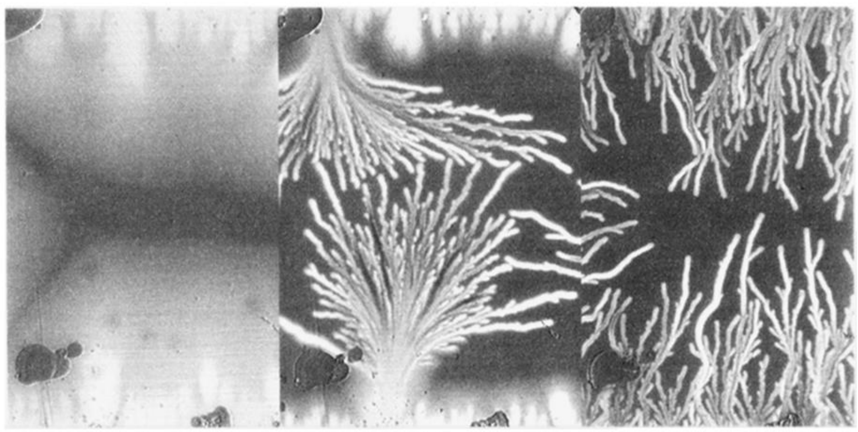

(a) (b) (c)

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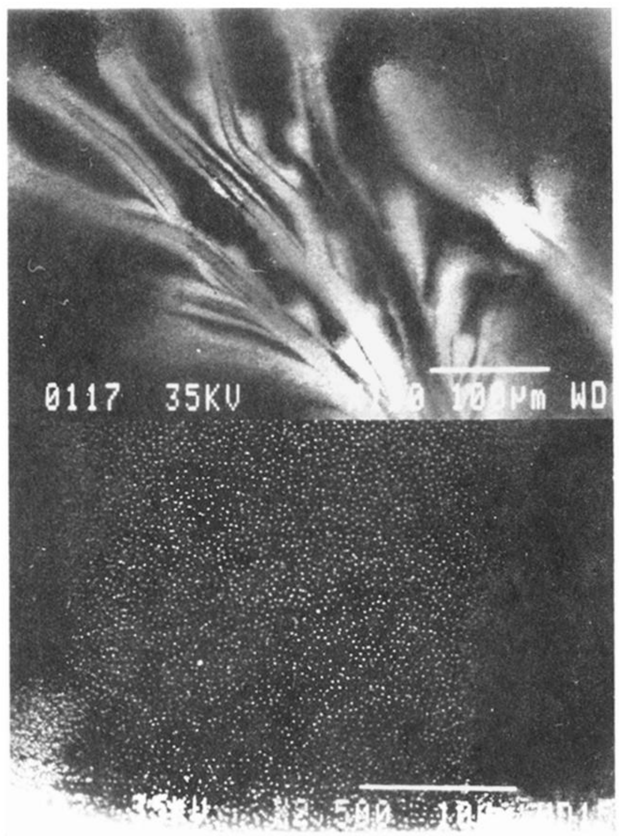

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