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Fluxoid Pinning by Small Nitride Precipitates in Niobium

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The magnetic hysteresis of superconducting niobium single crystals containing ~80-Å-diam nitride precipitates was investigated. Introducing precipitates into the samples produced no significant change in the critical-current density J_c except near the critical fields H_{c1} and H_{c2}. The samples trapped flux below H_{c1} and exhibited dramatic peaks in J_c within 3% of H_{c2}. These observations are interpreted as clear evidence of a threshold for fluxoid pinning, and reasonable agreement with theory is obtained.

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In spite of an intense research effort since the early 1960's, the fundamental understanding of fluxoid pinning by crystal defects in type-II superconductors remains incomplete. Although a number of attempts have been made to characterize the defect structure of a material and calculate its critical-current density, in most cases the theoretical value differs from the measured one by several orders of magnitude. Moreover, in contrast to experimental evidence, the theory of the statistical summation of the elementary interactions between fluxoids and defects implies that the interaction strength must be larger than a critical value before the defect can contribute to the critical-current density. This feature of the theory is known as the threshold effect.

Recently, attempts have been made to ascertain at what stage our understanding of fluxoid pinning breaks down. The absence of an experimental observation of the threshold effect and other evidence strongly suggests that the theory of the statistical summation of the elementary interactions is at fault.[1,2] However, the presence of several types of defects in most samples investigated to date obscures the interpretation of the experimental results. Here, we report the observation of the threshold effect approximately under the conditions specified by currently available theory.

We doped several single crystals of niobium with nitrogen to concentrations up to 0.1 at.%. A portion of the interstitial nitrogen absorbed at high temperature apparently formed small Nb_2N precipitates upon cooling. Throughout most of the mixed state, dc and ac magnetization measurements showed that the critical-current density in all the samples was comparable to that of the most reversible samples which we have investigated or have seen reported, and it did not change significantly upon doping with nitrogen. However, the dc magnetization measurements revealed a large increase in magnetic hysteresis near the lower critical field H_{c1} with doping. Indeed, the more heavily doped samples trapped flux when the applied field was reduced to zero from above H_{c1}. The ac measurements revealed a dramatic peak in the critical-current density within a narrow field range just below the upper critical field H_{c2}. The width of this peak increased with decreasing temperature and with increasing nitrogen concentration but was at most 3% of H_{c2}. Since reliable theoretical analysis is not available near H_{c1}, and since spurious geometrical effects and other experimental difficulties may be expected near H_{c1}, we analyze quantitatively only the data obtained in the field region near H_{c2}. The data presented below apply to the samples doped with 0.07 at.% nitrogen.

For each nitrogen concentration, a single-crystal prolate-spheroid sample (major and minor diameters are 25 and 3.2 mm, respectively) and a single-crystal cylindrical rod of similar size were loaded simultaneously by heating to 1800 °C in a dynamic pressure of 10^{-7} to 10^{-5} Torr nitrogen. A slice was cut from the center of the rod and thinned for transmission electron microscopy. The prolate spheroid was used for magnetization measurements.

Images of the precipitates were obtained by use of transmission electron microscopy (TEM) under bright-field (dynamical) and weak-beam, dark-field (kinematical) conditions. The width of the image perpendicular to the black-white direction in a bright field (dynamical diffraction condition) was taken as a measure of the precipitate size. The precipitate number density was determined to be 2.8×10^{16} cm^{-3}. The size distribution function was narrowly peaked at a diameter of about 80 Å with no precipitates observed larger than 130 Å. From the image properties (i.e., black-white direction) under different diffraction conditions, the strain distribution was found to have the symmetry properties corresponding to a platelet, coherent precipitate. From the comparison of observed black-white images with the calculated contrast,[3] the cumulative displacement of the crystal planes across the precipitate-matrix interface was estimated to be 7 Å.

The restoring force f_r due to distortion of the fluxoid lattice by a point pinning force has been calculated by Schmucker and Brandt,[4] using Ginzburg-Landau theory. Integrating Eq. (6) of Ref. 4 with approximations appropriate to our sample ($\kappa\simeq1$, $H_{c2}-B\ll H_{c2}$), we find

$$f_r/u_0=\xi H_c^{\,2}(1-\tfrac{1}{2}\kappa^2)^{1/2}(1-b)^{3/2} \qquad (1)$$

in Gaussian units, where u_0 is the displacement of the pinned fluxoid from its equilibrium position, ξ is the Ginzburg-Landau coherence length, κ is the Ginzburg-Landau parameter, H_c is the thermodynamic critical field, and $b=B/H_{c2}$ is the reduced flux density. Equation (1) is about 20% smaller than the corresponding limit of Eq. (9) of Ref. (4) because we have used the full isotropic approximation for the modulus $C_{11}(\vec{k})$.

The threshold criterion requires the pinning force f_p to vary sufficiently rapidly with the displacement u_0 of the pinned fluxoid such that

$$\partial f_p/\partial u_0 \geq f_r/u_0. \qquad (2)$$

When condition (2) is not satisfied, the $\vec{f}_p$'s due to a random array of defects have directions and their effects cancel. When condition (2) is satisfied, the presence of an elastic instability in the fluxoid lattice permits metastable configurations with nonzero sums of $\vec{f}_p$'s and a nonzero lossless bulk current density $\vec{J}_c$.

Of the elementary interactions that can be estimated between a fluxoid and a precipitate, the dominant one in our case is the second-order elastic interaction,[5] which arises from the spatial variation of the elastic moduli C_{ij} of the superconducting material in the mixed state. The strains associated with the precipitates fall off more rapidly than the spatial variation of the moduli (i.e., the fluxoid spacing). As a result, ΔC_{ij} may be removed from the integral expression for the interaction energy I. Then

$$I=\sum_{i,j=1}^{6}\Delta C_{ij}\,\partial W/\partial C_{ij}, \qquad (3)$$

where W is the elastic energy associated with the precipitate, which is approximately the same as that of an interstitial dislocation loop[6] with a Burgers vector of 7 Å. The interaction energy arises mainly from the variation of the shear modulus C_{44} and is substantially larger than previous estimates for dislocation loops.[1]

The elastic instability occurs first when the precipitate is at a fluxoid core. The force on this pinned fluxoid can be expected to be nearly the same as the total force on the fluxoid lattice so that

$$\frac{\partial f_p}{\partial u_0}=\left|\frac{\partial^2 I}{\partial u_0^{\,2}}\right|\simeq\left|\frac{\partial^2 I}{\partial x^2}\right|\simeq\left|\frac{\partial^2\Delta C_{44}}{\partial x^2}\right|\frac{\partial W}{\partial C_{44}}. \qquad (4)$$

With the estimate in Ref. 5 for $\Delta C_{44}(\vec{x})$, and the temperature dependence $\Delta C_{44}\propto H_c(T)/H_c(0)$, we find that the threshold criterion leads to the conclusion that the small precipitates in our sample contribute to J_c only for fields within ΔH of H_{c2}, where

$$\Delta H/H_{c2} \leq 3.07\times 10^{-7}(\kappa^2+0.21)H_{c2}, \quad (5)$$

when H_{c2} is in oersteds.

Both the dc and ac magnetization measurements were made with the sample in a uniform dc field parallel to its long axis. The ac applied field was smaller than and parallel to the dc field. The experimental apparatus is described elsewhere.[7,8] The total ac flux variation φ in the sample obeyed the relations $\varphi = 2\pi a^2 \mu_{\min}' h_0$ at small ac field amplitude h_0, and $\varphi = 2\pi a^2 \mu_0'(h_0 - H^*/3)$ at large h_0. Here a is the sample radius, the parameter H^* is given by $H^* = 4\pi J_c a/c$ for a uniform critical-current density J_c, and μ_0' is the equilibrium differential permeability. The parameter $\mu_{\min}'$ measures the degree of distortion the fluxoid lattice will undergo before breaking loose from pinning defects. It is related to Campbell's penetration depth λ' (Ref. 2) by

$$\mu_{\min}' = 2\mu_0'\lambda' I_1(a/\lambda')/aI_0(a/\lambda') \simeq 2\mu_0'\lambda'/a,$$

where I_0 and I_1 are modified Bessel functions. All of the magnetization measurements reported here were made after heating the sample to 400 °C in oxygen for four minutes.[7] This treatment had little effect except to reduce the hysteresis uniformly throughout the mixed state.

The dc magnetization of a prolate-spheriodal single crystal is shown in Fig. 1 before and after doping with 0.07 at.% nitrogen. The critical fields H_{c1} and H_{c2} change (and the resistivity increases) with the addition of nitrogen, as one would expect. The hysteresis near H_{c1} after doping is clear from the dc measurements, but the peak in J_c near H_{c2} could be seen only in the ac measurements. Presumably the hysteresis in the dc magnetization near H_{c2} was removed by small temperature transients that inevitably occur when we physically move the sample in order to obtain an induced voltage that is our measure of the dc magnetization.

The values of J_c near H_{c2} deduced from the ac measurements at high h_0 are plotted in Fig. 2 along with the measurements of $\mu_{\min}'$. H_{c2} is determined by noticing when μ_0' drops abruptly to unity. At the same time that J_c increases by more than a factor of 50, $\mu_{\min}'$ decreases rapidly. The most commonly accepted explanation for a peak in J_c near H_{c2} holds that the defects which contribute to J_c at fields below the peak contribute to J_c at fields below the peak contribute more efficiently within the peak because the softening of the fluxoid lattice near H_{c2} permits a high degree of correlation between the fluxoid and defect positions. The observed drop in $\mu_{\min}'$ indicates that the pinned fluxoid lattice becomes more rigid; the "softening" explanation for the peak in J_c would predict the opposite behavior of $\mu_{\min}'$. The most natural explanation for our observations is that the largest precipitates begin contributing to J_c at the low-field side of the peak. As the field is increased, more and more

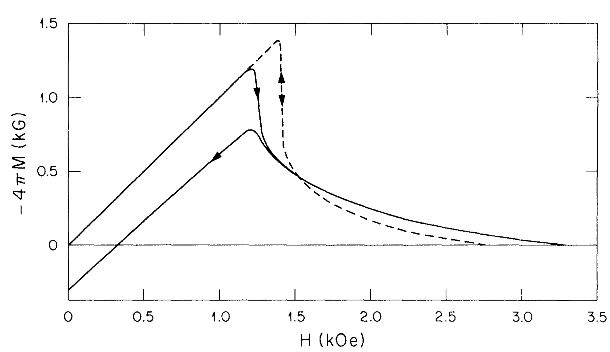

FIG. 1. Magnetization at 4.2 K of the niobium prolate spheroid doped with 0.07 at.% N (solid lines) and of the same sample before doping (dashed lines).

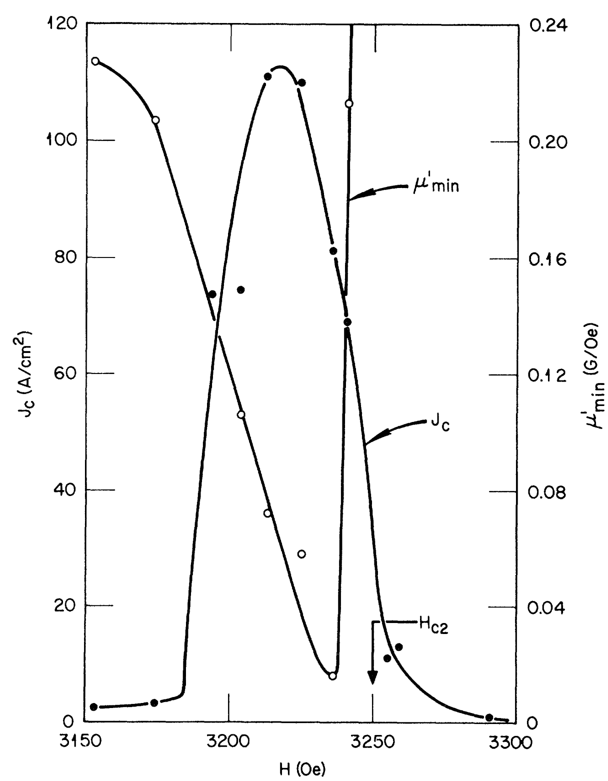

FIG. 2. The critical-current density J_c and the low-amplitude ac permeability $\mu_{\min}'$ near H_{c2} at 4.2 K.

precipitates contribute to J_c. Although the elastic moduli of the fluxoid lattice decrease with increasing field, μ_{min}' decreases (indicating apparently increased rigidity) because the lattice is pinned in more places. Sufficiently close to H_{c2} the linear decrease of I with $1-b$ must finally cause J_c to drop toward zero and μ_{min}' to approach unity.

The reduced width of the peak in J_c is plotted in Fig. 3 as a function of H_{c2} and is compared with the theoretical curve based on inequality (5). It has recently become apparent that the estimate of the elementary pinning interaction is a controversial matter. However, the controversy centers on the magnitude of I and the predicted peak width, not on the existence of the peak. The degree of agreement indicated in Fig. 3 should be regarded as tentative, order-of-magnitude confirmation of the theory that we have used.

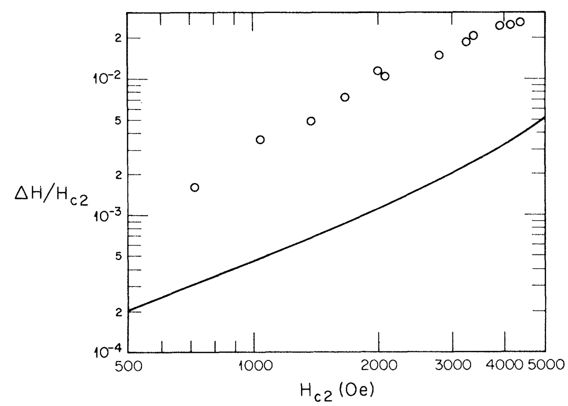

FIG. 3. The temperature dependence of the reduced width of the peak in J_c, $\Delta H/H_{c2}$, is compared with the theoretical onset of J_c given by inequality (5) with $\kappa = \kappa_1(T)$.

The data shown in Figs. 2 and 3 apply to the increasing-field history only. For the decreasing-field history, J_c was larger over a range of fields below the peak at all temperatures above 3 K. This observation indicates that disorder induced in the fluxoid lattice by passing through the high-J_c region can reduce the threshold criterion presumably by softening the fluxoid lattice as speculated by Kramer[1] and others. However, the sharp rise in J_c with increasing field and the reasonable agreement between theory and experiment indicate that fluxoid-lattice disorder does not play a major role in the increasing-field data, which we have interpreted.

We have obtained convincing evidence that small nitride precipitates in niobium contribute to the critical-current density only in narrow field ranges near H_{c1} and H_{c2}. The introduction of nitride precipitates into the samples produced magnetic hysteresis near H_{c1} and a sharp peak in J_c coincident with a sharp dip in μ_{min}' near H_{c2}. At intermediate fields no significant change in J_c was observed. The appearance of a significant increase in J_c only within narrow field ranges is itself strong evidence of a threshold effect. Careful examination of the data near H_{c2} adds further support to this interpretation. Finally, rough quantitative agreement between theory and experiment provides at least tentative confirmation of the theory.

Our results are in marked contrast to the experimental results for dislocation loops in niobium.[1] In spite of the similarity between the coherent precipitates in our sample and dislocation loops, the observed behavior of J_c is quite different. One might speculate that the presence of several types of defects in the irradiated samples could have obscured the threshold effect. In the investigations of fluxoid pinning by loops and voids, polycrystalline specimens were used, and these samples after the irradiation must invariably contain dislocation tangles, which were completely neglected in the fluxoid-pinning analysis. It should be reemphasized that the present work was done on single-crystal specimens, which contained well-characterized, platelet-type precipitates. Other defects could not be observed in our samples. The discrepancy between our results and those for dislocation loops in niobium deserves further investigation.

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