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Investigation of the nonequilibrium mixed state of superconducting niobium with pinning centers

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The interaction of magnetic flux vortices that enter the sample upon increase in the magnetic field with the frozen-in magnetic flux is studied by using the anisotropy of the impedance of type-II superconductors. If the pinning is effective, the magnetic flux of the sample in the intermediate state remains parallel to the frozen-in flux. The manner in which the total magnetic flux is confined by the pinned vortices when the magnetic field is rotated relative to an immobile sample is investigated. The experimental results can be interpreted in terms of plastic deformation of the vortex lattice.

As is well known, the surface impedance of type-II superconductors depends on the inclination of the vortex lattice relative to the surface of the sample.[1] We have used this property for the study of the behavior of the vortex lattice in superconducting niobium containing pinning centers in various concentrations. Vinikov *et al.*[2] have shown that the critical current changes strongly in superconducting niobium subjected to chemical polishing, different cooling regimes, and annealing in a high vacuum. This change was attributed to the formation of niobium hydride. Thus in niobium it is easy in practice to change the concentration of pinning centers in one and the same sample.[2]

In type-II superconductors in the mixed (intermediate) state containing pinning centers, a large or small part of the vortices are pinned, depending on the concentration of the centers. These pinned vortices remain as a frozen flux upon decrease in the magnetic field to zero. In the present work, we have studied the interaction between vortices of the magnetic flux entering into the sample upon increase in the magnetic field and frozen vortices whose orientation does not coincide with the direction of the field. We also studied how the pinned vortices contain the entire magnetic flux in the rotation of a magnetic field of constant magnitude.

PREPARATION OF THE SAMPLES

The niobium samples were cut from a single-crystal ingot with resistance ratio $\rho_{300K}/\rho_{4.2K}=250$ and were subjected to mechanical polishing. The case-hardened surface layer was then removed by chemical polishing in a mixture of nitric and fluoric acids. The chemical polishing in the present work is important since a significant amount (~1 at.%) of hydrogen is dissolved in the niobium because of it.[2,3] This dissolved hydrogen was used to produce the pinning centers in the niobium. At a temperature of $T=245$ K, bonding of the hydrogen into niobium hydride occurred. Below this temperature, inclusions of the hydride phase are observed in the niobium matrix; these reached sizes of 10^{-3} to 10^{-5} cm[2] depending on the cooling rate. The change in volume of the hydride phase in comparison with the matrix amounted to ~10% and led to plastic deformation of the niobium near the place of formation, a fact also reflected in the present results. Since the niobium hydride is not a superconductor in the investigated temperature range, its segregations in the matrix constitute the pinning centers. The concentration of the dissolved hydrogen and the corresponding pinning centers varied with the temperature of the chemical polishing. We have carried out chemical polishing at room temperature ($t=23$ °C) and at a temperature of 0 °C. After completion of the cycle of measurements, the samples were subjected to annealing in a vacuum of 10^{-8} Torr at a temperature of 800 °C. This annealing had practically no effect on the dislocation structure of the niobium, but led to complete removal of the hydrogen from the sample. The dislocation clusters remaining at the location of hydride formation are also pinning centers, but they are less effective in pinning the magnetic flux vortices. One of the samples was annealed immediately after chemical polishing, i.e., it did not contain "controlled" pinning centers (at $T=4.2$ K, neither niobium hydride nor clusters of dislocations produced by the previously existing formations were produced). For an explanation of the effect of the residual impurities on the studied effect, a cycle of measurements was performed on a sample of niobium with a resistance ratio $\rho_{300K}/\rho_{4.2K}=1500$. The results showed the insensitivity of the studied effects to the residual resistance of our samples. The data on the investigated samples are given in the table.

METHOD OF MEASUREMENT

We studied the active part of the surface impedance of superconducting niobium in the intermediate state.

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Table I.

Data on samples	Number of sample*			
	2,2′	3,3′	5	6
ρ 300 K/ρ 4.2 K**	250	250	1500	250
Temperature of chemical polishing	23	0	0	0
Annealing after chemical polishing, °C	no	no	no	yes

*2′ and 3′ samples 2 and 3 annealed after the cycle of measurements.
**The resistance ratio was determined for the initial ingot from which the samples were prepared.

Measurements were carried out at a frequency of about 800 MHz. A flow-through spectrometer scheme was used.[4] The absorbing cell consisted of a helical resonator, made from aluminum wire. The sample, in the form of a strip with dimensions $15\times 4\times 0.6$ mm, was placed inside the helix. The measurements were carried out at a perpendicular orientation of the constant magnetic field and the UHF current (inset in Fig. 1). Change in the frequency of the radiation near 800 MHz by an octave did not reveal any frequency dependence of the character of the behavior of the impedance. The magnetic field created by the laboratory electromagnet could be rotated in the xy plane. Measurements were made at a temperature of 4.2 K, the sample and the resonator were insulated from the liquid helium. The absence of overheating was monitored against the change in the impedance curves upon increase in the radiated power.

Figure 1 shows the field dependence of the impedance of sample 6 of the niobium (without controlled pinning centers) for different angles of inclination of the magnetic field to the surface of the sample. The character of the field dependence for all the studied samples was similar, with the exception of the singularities (the maximum of the impedance near H_{c1} and the step near H_{c2}). These singularities are deformed as the concentration of pinning centers increases. The impedance of the sample with a small number of pinning centers, such as sample 6, in our view is of special interest. In addition to the already mentioned singularities, we can call attention to the relative minimum of the impedance as a

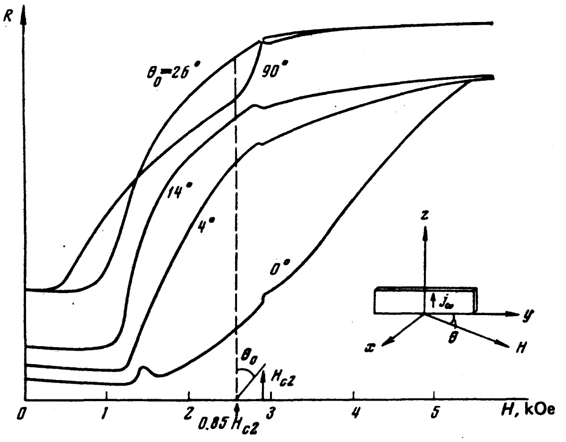

FIG. 1. Dependence of the active part of the impedance $R(H)$ on the magnetic field for different directions of the magnetic field relative to the surface of the sample. The magnetization is parallel to the magnetic field.

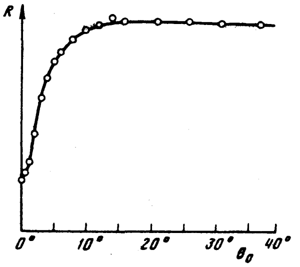

FIG. 2. Dependence of the active component of the impedance on the direction of the magnetic field relative to the surface of the sample ($H=0.85\,H_{c2}$).

function of the angle at $\theta_0=90°$ in fields somewhat smaller than H_{c2} (compare the curves at 26° and 90°, respectively). The change in the behavior of the impedance in the magnetic field $H<H_{c1}*$ (~1.4 kOe) at large angles of inclination of the field to the surface of the sample is due to the demagnetization factor of the plate. As is seen from this drawing, in the region of the intermediate state the values of the surface impedance depend on the angle of inclination of the magnetic field and, consequently, on the inclination of the magnetic flux vortices to the surface of the sample. Figure 2 shows the change in the impedance of niobium sample 2 as a function of the angle of inclination of the field to the surface of the sample in a magnetic field $H=0.85H_{c2}$. The impedance is reckoned from its minimum value when the field is parallel to the surface of the sample. It follows from this drawing that the greatest sensitity to the inclination of the vortex structure relative to the surface occurs in the range of angles $\theta_0=0$–$6°$. At large angles, the dependence $R(\theta_0)$ reaches a plateau, and the impedance is not sensitive to the inclination of the vortices relative to the surface of the sample.

All the results described above were obtained in the case of a vortex structure in equilibrium relative to the external magnetic field. The measurements were begun at a value of the magnetic field $H>H_{c3}$, when the sample is in the normal state. Upon decrease of the magnetic field below the value H_{c2}, a vortex structure with a minimum energy in an external field with a specified direction appears in the sample. The magnetic field decreased to zero and then a recording of the active component of the impedance in an increasing magnetic field was made as a function of the external field. By plotting all the curves for one direction only we can disregard hysteresis phenomena.

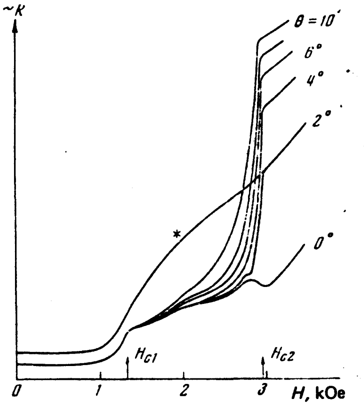

FIG. 3. Dependence of the active component of the impedance $R(H)$ for different directions of the magnetic field relative to the surface of the sample (nonequilibrium case). The frozen-in flux is parallel to the surface ($\theta_0=0$). The curve marked by an asterisk corresponds to the dependence of $R(H)$ for the angle 2° at $\theta_0=2°$.

RESULTS OF EXPERIMENT

The principal part of the experimental results refer to the study of the impedance in a nonequilibrium vortex structure relative to the external field. The procedure of measurement was as follows. The magnetic field was reduced from the value $H > H_{c2}$ (the excess above H_{c3} is unimportant for the results) at some angle θ_0 between the surface of the sample and the direction of the external field. At $H = 0$ there remains a frozen flux in the sample, connected with the pinning centers. In this state the magnet is rotated to some new angle θ and a recording is made of the impedance in the case of an increasing external field. Under these conditions, with a field larger than H_{c1}, a new system of vortex filaments begins to penetrate into the sample. We are interested in the interaction of these incoming vortices with the frozen ones. From the shape of the impedance curve, we can draw a conclusion as to the position of the vortex structure relative to the surface of the sample, by comparing the obtained values of the impedance with data similar to those shown in Figs. 1 and 2 for the corresponding sample. From the form of the dependence of the impedance R on the angle θ_0 (Fig. 2) it is clear that the initial value of the angle θ_0 should be found in the sensitive region (0–6°).

Figure 3 shows the results of such a measurement for the angle $\theta_0 = 0$ (the field is parallel to the broad side of the strip) for the sample 2 (sample with a maximum number of pinning centers). The curve marked by a star corresponds to the equilibrium magnetization of the sample $\theta = \theta_0 = 2°$. The same holds also for the initial curve ($\theta = \theta_0 = 0°$). As is seen from this drawing, the change in the impedance for some angle θ more closely corresponds to the impedance for the angle θ_0 (in the given case $\theta_0 = 0$) at which the magnetic flux is frozen than the impedance for the equilibrium configuration (compare the curves corresponding to 2° with the curve marked by the star in Fig. 3). Since the angle $\theta_0 = 0$ is a special one (the field is parallel to the surface), the analogous results are shown in Fig. 4 for the angle $\theta_0 = 6°$. The character of the data remains the same as before, the impedance remains close to the value corresponding to the angle at which the magnetic flux was frozen. A sharp change begins in the field $H = 0.95\ H_{c2}$, and at $H \geq H_{c2}$ the impedance returns to the value determined by the surface superconductivity for the existing direction of the magnetic field. Thus the change in the impedance of the sample in fields less than $0.95\ H_{c2}$, "remembers" the angle at which the

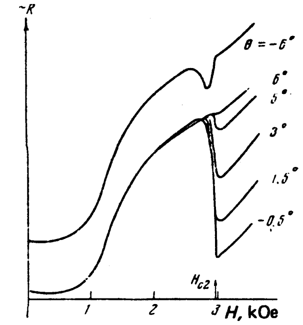

FIG. 4. Dependence of $R(H)$ for different directions of the magnetic field relative to the surface of the sample. The frozen-in flux is directed at an angle of $\theta_0 = 6°$ to the surface of the sample. The $R(H)$ curve for $\theta = 6°$ is shifted vertically.

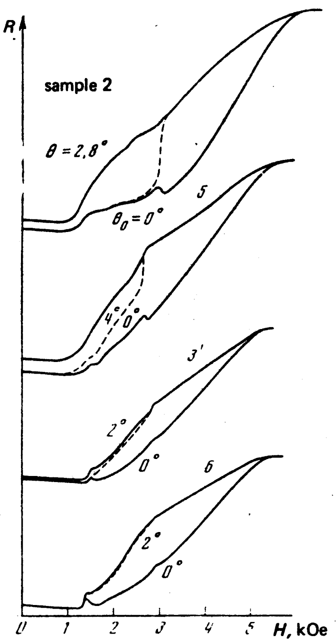

FIG. 5. Dependence of $R(H)$ for different samples. The continuous curves correspond to the impedance in the equilibrium state ($\theta = \theta_0$), the dashed to nonequilibrium (*k* the frozen magnetic flux is parallel to the surface of the sample $\theta_0 = 0$, and the field is directed at the angle θ shown in the figure).

magnetic field decreased from the value $H > H_{c2}$ to zero.

Maximum "memory" is possessed by samples that are chemically polished at room temperature and rapidly cooled (sample 2), the least memory by samples that were chemically polished at $t = 0\,°C$ (samples 3 and 5). The memory is correspondingly reduced after annealing of the samples at $t = 800\,°C$ in vacuum (samples 2′, 3′). The memory effect is completely lacking for the sample which underwent annealing immediately after chemical polishing (sample 6). Figure 5 shows for the different sample comparative data characterizing the memory effect in the change in the impedance. For the samples 3 and 2′, the data are similar to the curve for sample 5.

As is seen from the given data, in the study of the impedance under conditions of the presence of the memory effect, the magnetization of the samples connected with the vortex structure departs significantly from the direction of the external field. Here a force acts on the sample and tends to expand it. This requires rigid fastening of the sample and of the UHF resonator, so that the displacements do not affect the

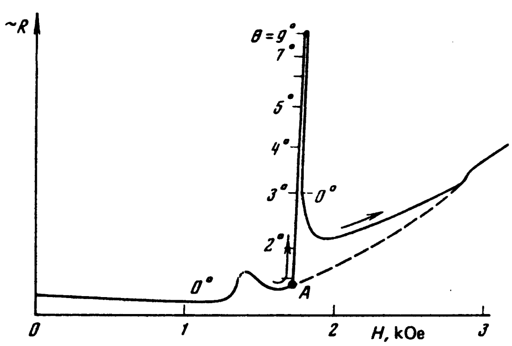

FIG. 6. Dependence of $R(H)$ at $\theta = \theta_0 = 0$. In the magnetic field H_A the field was rotated through an angle of $\theta = 9°$ and back with subsequent recording of the impedance for the sample without hydrogen (continuous curve). The dashed curve corresponds to the impedance without rotation.

impedance measurements. To this end, the sample and the resonator were secured with polystyrene linears greased by a silicone liquid (GKZh) having a low solidification temperature.

The experiment with the memory shows that the frozen-in flux determines to a significant degree the direction of the total magnetic flux of the sample. From these considerations, we carried out a simpler experiment on observation of the effect of confinement of the total magnetic flux of the sample upon departure of a constant magnetic field from some initial direction. In order to know the previous history of the sample, the magnetic field was decreased from the value $H > H_{c2}$ to zero at $\theta_0 = 0$. Then the field was increased from zero to some value between H_{c1} and H_{c2} (the point A on Fig. 6). At this point, we rotated the magnetic field and determined the impedance at certain discrete values of the angle. On reaching some angle (in Fig. 6, $\theta = 9°$), we returned the magnetic field to its original direction $\theta = 0$. This process is shown in Fig. 6 for sample 6. The increase in the impedance from point A to its value at the corresponding angle is shown in Fig. 7 (continuous curve) for sample 2 (the sample with the maximum pinning). For comparison, the increase in the impedance as a function of the angle of inclination to the surface of the sample upon removal of the anisotropy of the impedance ($\theta = \theta_0$) (curve in Fig. 2) is shown in the same drawing. Figure 7 shows that at angles of inclination of the magnetic field up to 5° the impedance is practically unchanged, i.e., the vortex structure does not depart from its initial direction. Inclination of the field by an angle ~25° leads to a mean inclination of the vortex structure by an angle ~2° (from comparison with the dashed curve).

Even a sample which does not possess the memory effect exhibits irreversibility of the change in the impedance upon completion of the rotation cycle. When the field returns to the previous position, the impedance of the sample differs from the initial value. Some memory develops in the sample after rotation of the field. In samples possessing memory, irreversibility is practically complete, i.e., upon return of the magnetic field to $\theta = 0$, the impedance remains equal to the value at the maximum angle (if the angle is not greater than 15°). These experiments show that for samples with pinning centers, the frozen-in part of the flux confines the entire magnetic flux.

DISCUSSION OF THE RESULTS

The principal factor determining the observed phenomena is the interaction of free vortex filaments with pinned ones. The memory effect is connected with the alignment of the magnetic-flux vortices entering the sample parallel to the already existing vortices of the magnetic flux. Here the competition of the interaction between vortices and the interaction of the magnetic flux vortices with the external field contribute to the formation of vortex-lattice defects that decrease the energy of interaction with the external field, and the result is incomplete memory. The vortex-lattice

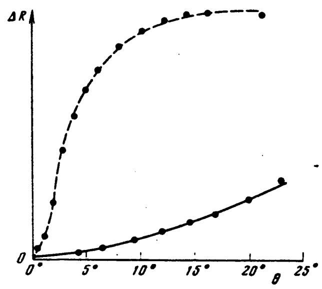

FIG. 7. Increase in the impedance for a rapidly cooled sample upon rotation of the field, as a function of the angle (continuous curve). The origin is taken to be the value of the impedance corresponding to the point A on Fig. 6. The dashed curve corresponds to the analogous increase for the anisotropy of the impedance in Fig. 2 ($H = 0.68\ H_{c2}$).

crystal grows as it were on the seeds of the vortices of the frozen-in flux in the field of the acting volume force. Near H_{c2} the vortex lattice softens and the magnetization of the sample turns towards the external field. This is clearly seen from the curve ($\theta = -6°$) of Fig. 4, where there is a minimum near H_{c2} in the impedance, corresponding to the passage of the vortices, which were previously directed at an angle of 6°, through the direction parallel to the surface of the sample. The turn occurs gradually and is reproducible upon increase in the magnetic field.

If a type-II superconductor in the intermediate state is placed in a magnetic field which is not parallel to the direction of the vortices, then the field tends to align the vortex structure parallel to the field direction. An interaction between the vortices sets in and contributes to the homogeneity of the intermediate state. The homogeneous state in the given case represents a lattice of parallel rectilinear vortex filaments. The interaction with the pinning centers tends to localize the perturbation of the vortex lattice by the external field in the region near the surface of the sample (at the distance from the surface of the sample to the pinning center). The irreversibility of the impedance change, observed in experiments with rotation of the magnetic field (Fig. 6), makes it possible to reject the near-surface bending of the vortices as a possible explanation of the observed departures. This bending should be reversible.

The qualitatively obtained results can be described by a model based on dislocations in the vortex structure of a type-II superconductor. The dislocations in the vortex structure of a superconductor are described, for example, in Ref. 5. From the deviations of the value of the impedance in zero field we can roughly estimate quantitatively the number of pinned vortices of the magnetic flux (several per cent of the maximum number of vortices at $H \to H_{c2}$). In other words, in the intermediate state the sample contains a large number of unpinned vortices which should be parallel to the pinned vortices by virtue of the interaction between the vortices. We can isolate in the sample some cells containing free vortices. For small angles of inclination, the interaction energy of the field and the vortex structure in such a cell can be written down as

$$E \sim n\Phi_0 H\varphi^2 S,$$

where n is the concentration of vortices, Φ_0 is the magnetic flux quantum, H is the magnetic field, φ is the angle of inclination of the vortex structure to the direction of the magnetic field, and S is the cross sectional area of the cell perpendicular to the vortex filaments. This energy can be regarded as the product of the generalized coordinate φ and the generalized force $\sim n\Phi_0 HS\varphi$. Then experiments with rotation of the magnetic field can be treated as the deformation of the vortex lattice (change in the generalized coordinate φ under the action of the generalized force). The value of the force here is connected with the concentration of the pinning centers through the area S. The greater S the fewer the pinning centers and the greater the force. The observed results can easily be treated here as the plastic deformation of the vortex lattice—the formation of dislocation half loops from the surface of the sample. The combination of such half loops leads to the turning of the pinned part of the vortex lattice in the direction of the acting force. The greater the dimensions of the cell S, the greater the deviation. The irreversibility here is natural. The results given in Fig. 6 for a sample without the memory effect can be treated as deformation hardening due to the crossing of the vortex filaments upon formation and motion of the dislocations as a consequence of the deviation of the magnetic field and its return to the previous direction.

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Experimental and theoretical investigation of spin-reorientation phase transitions in cubic ferromagnets and ferrimagnets in a magnetic field

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A theoretical investigation of the nature of the spin-flip phase transitions inducible by a magnetic field in infinite cubic ferromagnets is carried out with allowance for the fourth- and sixth-order terms in the expression for the magnetic-anisotropy energy. Possible (H,T) orientational phase diagrams corresponding to magnetic-field directions along high-symmetry axes of the type $\langle 100\rangle$, $\langle 111\rangle$, and $\langle 110\rangle$ are constructed. The critical points of the first-order phase transitions are determined, and the transitional domain structure accompanying a first-order transition in samples of finite dimensions is studied. An experimental verification of the results of the theory has been carried out on single crystals of the rare-earth iron garnets $Tb_xY_{3-x}Fe_5O_{12}$ $(x=0.10, 0.26)$ and $Sm_3Fe_5O_{12}$. Some (H,T) phase diagrams have been reproduced on the basis of torque, magnetization, and differential susceptibility studies. Particular attention has been paid to the detection and investigation by the Fe^{57}–NMR method of the transitional domain structure accompanying first-order orientation transitions, as illustrated by the iron garnet $Tb_{0.1}Y_{2.9}Fe_5O_{12}$.

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INTRODUCTION

Considerable attention is being paid at present to the investigation of magnetic phase transitions of the order-order type and, in particular, of spin-flip phase transitions (SFPT). It should be noted that the most intensively investigated SFPT are those occurring in uniaxial and biaxial magnetic substances, especially in the rare-earth orthoferrites (see, for example, the review published in Ref. 1). The study of the cubic ferro- and ferrimagnetic substances began only recently, the main attention having been paid to the investigation of spontaneous SFPT, which occur in zero magnetic field, and which are due to the change that occurs in the nature of the magnetic crystallographic anisotropy when the temperature changes.[2-8]

The influence of an external magnetic field on the behavior of the magnetization of an anisotropic cubic ferromagnetic substance in a narrow range of temperatures in the vicinity of the magnetic-ordering temperature has been investigated without allowance for the critical fluctuations in Refs. 9 and 10 and with allowance for the fluctuations in Ref. 11. However, the present authors do not know of any work in which a sufficiently complete analysis has been performed of the orientation phase di-

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