Magnetic flux pinning in a type-I superconductor in the intermediate state

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A study is made of the conditions under which a transport current can flow without loss in a type-I superconductor in the intermediate state. It is found that the critical current does not vanish in a comparatively broad range of field strengths in the intermediate state. The results are interpreted from the viewpoint of pinning of the magnetic flux. The dependence of the average pinning force on sample geometry and heat treatment of the samples is discussed. It is pointed out that type-I superconductors in the intermediate state may be convenient for experimental study of pinning mechanisms.

1. INTRODUCTION

By intermediate states of a type-I superconductor is meant a state in which the magnetic field penetrates into the superconductor in a homogeneous fashion, because the volume of the sample breaks up into a system of regions of normal and superconducting phases. This state is realized when the superconductor is placed in an external magnetic field whose intensity lies in the interval $H_c(1 - n) \leq H \leq H_c$ (n is the demagnetization factor and H_c is the critical magnetic field).

One can imagine two types of idealized intermediate-state structures: a) a layered structure in which the normal and superconducting phases form a system of alternating layers[1], and (b) a filamentary structure, which can constitute a system of normal filaments or tubes of magnetic flux in a superconducting matrix (when the concentration of the normal phase is small) or else a system of superconducting filaments in a normal matrix (when the normal-phase concentration is large). The hypothesis that a structure of normal filaments stretching along the external field may be energy-wise more favored in weak magnetic fields was first advanced by Landau[2]. Andrew[3] calculated the free energy of the filamentary structure and showed that in weak fields such a structure corresponds to a minimum of the energy. Recently Goren and Tinkham[4] proposed an idealized intermediate-state model of plates in a perpendicular magnetic field, according to which the flux penetrates into the sample through isolated regions of the normal phase, which form a triangular lattice in a superconducting matrix. The free energy of such a system differs little from the free energy of the layered model.

It should be noted that all the estimates of the free energy within the framework of any model pertain to the case of complete thermodynamic equilibrium. The free energy is then relatively insensitive to the details of the topology of the intermediate state. One can therefore observe in experiments a great abundance of intermediate-state structures, including a structure in which the external magnetic field penetrates into the sample in the form of macroscopic tubes of magnetic flux that pass through the regions of the normal phase and are "interspersed" in the superconducting matrix.

The realization of such a structure, at least during the initial stage of the intermediate state, is evidenced by a number of recent studies[4-9], particularly the visual observations of Träuble and Essmann[7] and of Solomon[8], and the data of Goren and Tinkham[4] on the topography of the magnetic field in thin plates, which were obtained with the aid of a scanning Hall pickup.

The existence of such a filamentary structure in the intermediate state allows us to speak, in a certain sense, of an analogy with the mixed state of type-II superconductors. We have in mind, first, the possible motion of the macroscopic flux tubes under the influence of an electric current and, second, their interaction with inhomogeneities of the sample structure. The former should lead to the onset of a new mechanism of resistance to the electric current, similar to that observed in the mixed state during the motion of Abrikosov vortices (the resistive effect[10]). The latter is connected with the influence of pinning in the intermediate state and therefore enables us to speak of a critical current in the same sense as introduced for type-II superconductors (the concept of the critical state[11,12]).

The usual loss mechanism in the intermediate state is the ohmic resistance which occurs when current flows perpendicular to layers of the normal phase. The only possibility for the nature of the losses to become resistive within the framework of the layered model can be realized in the case when the layers of the normal phase are elongated along the current and move in a perpendicular direction[13,14]. This specific case can be realized in single-crystal samples in a magnetic field almost parallel to the sample surface. As shown by Sharvin[15], one obtains under such conditions a regular layered structure oriented along the projection of the magnetic field on the surface of the sample. Sharvin did not observe the motion of such a structure under the influence of current[16]. Another case when the resistive loss mechanism can be realized in the intermediate state can arise only out of motion of isolated regions of the normal phases. This case was observed by us in tin plates in a perpendicular magnetic field[9].

The present paper is devoted to a study of the conditions under which the pinning of isolated regions of the normal phase becomes manifest in the intermediate state. We discuss an experimental method of measuring the critical current density in the intermediate state. By critical is meant the upper limit of the current density, at which there are still no resistive losses, in full analogy with the concept of the critical state for type-II superconductors. We present below data on the dependence of the critical current density on the external magnetic field, on the sample dimensions, and on their heat treatment, and estimate the average pinning force in the intermediate state on the basis of these data. We discuss the possible connection between the pinning force and the dimensions of the normal-phase regions. In conclusion, we advance some ideas concerning the feasibility in principle and the advantages of investigating the mechanism of pinning of the magnetic flux with the aid of type-I superconductors in the initial stage of the intermediate state.

2. EXPERIMENTAL PROCEDURE

Usually the critical current is determined by a potentiometer method from the appearance of a voltage drop across the sample when the current reaches a certain threshold value. In type-II superconductors in the mixed state, the voltage drop is interpreted quite uniquely as the start of the resistive losses connected with the motion of the Abrikosov vortex filaments. In the absence of a pinning force, such losses intervene at arbitrarily small currents, but the presence of "dry friction" between the vortices and the defects of the sample structure leads to nonzero values of the critical current. The dependence of the critical current density on the magnetic field characterizes the establishment of the so-called "critical state" of the type-II superconductors, whereby the forces exerted on the vortices by the transport current become equal to the forces exerted by the structure defects (the so-called "pinning centers").

In the intermediate state, however, the appearance of a potential difference across the sample when current flows through the sample from an external source can be a consequence either of the resistive loss mechanism or of the trivial ohmic mechanism. These two cases can be distinguished by eliminating the external current source and exciting current in a closed superconducting circuit in which the sample is connected in series. Then any loss mechanism leads to attenuation of the current in the circuit. If the loss is ohmic, there can be no current flow in the circuit. If the loss is connected with motion of isolated regions of the normal phase, then two possibilities arise. 1) The regions move freely in space under the influence of the current, being created on one surface and vanishing on the other. The current in the circuit attenuates to zero. 2) The regions are capable of being pinned to inhomogeneities of the sample structure. The current in the circuit attenuates to a certain critical value, at which it is no longer capable of causing motion of the regions. Measurement of the relaxation time of such an attenuation makes it possible to assess the effective resistance[9], and the function $\bar{j}_c(H)$ ($\bar{j}_c$ is the average critical current density and H is the external field) characterizes the effectiveness of the pinning.

A schematic diagram of the measurements is shown in Fig. 1. The sample was connected in series with a coil W_1 wound of lead wire of 0.5 mm diameter. To excite an undamped current in such a circuit, a pair of excitation coils W_2 was mounted coaxially with the coil W_1 and produced a field H_e. The magnetic field **H** was applied perpendicular to the surface of the sample with the aid of another pair of coils W_3. The coils W_1, W_2, and W_3, together with the sample, were mounted on a German-silver tube that passed to the outside through a seal in the cover of the cryostat. This system was set in reciprocating motion with the aid of a synchronous motor and a crankshaft and rod mechanism, at a frequency ~0.1 Hz and amplitude 7 mm. The detector coil W_4 was placed in the immediate vicinity of coil W_1, at the bottom of the cryostat. Motion of the coil W_1 relative to W_4 induced in the latter an alternating emf proportional to the current in the superconducting circuit. The measuring system consisted of a type F116/1 photocompensation microvoltmeter, a type ÉPP-09 electronic potentiometer with a time constant of 1 sec, and a system of variable resistances, with the aid of which the sensitivity of the circuit could be varied in a wide range. The apparatus made it possible to measure currents from 30 mA to tens of amperes and to depict a relative change of current ~5% in the superconducting circuit.

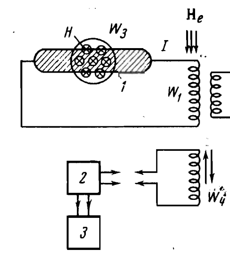

FIG. 1. Schematic diagram of the measurements: 1—sampe, 2—F116/1 photocompensation microvoltmeter, 3—ÉPP-09 electronic potentiometer.

The measurement procedure consisted of the following: at a temperature $T < T_c$, the system was transformed to the normal state by turning on a field $H > H_c(T)$. At the same time, the field H_e was turned on with the aid of the excitation coils. The sample was again made superconducting by turning off the field H, and an undamped current I_0 was excited in the circuit by turning off the field H_e. The value of the current depended on H_e. Thus the system was returned to the initial state. By applying to the sample fields H with different values from zero to $H_c(T)$, we determined each time the equilibrium value of the current I in the superconducting circuit, corresponding to a given H. We thus plotted the dependence of the average critical current density $\bar{j}_c$ on the external field.

3. SAMPLES

The samples were polycrystalline tin plates 22 mm long, 6 mm wide, and from 1.3 to 0.04 mm thick, obtained by rolling the original ingot. The resistance ratio was $R(293°K)/R(4.2°K) = 560$. The resistivity was $\rho_n = 2.4 \times 10^{-6}\ \Omega$-cm at $T = 4.2°K$. The Ginzburg-Landau parameter calculated from the known formula

$$\varkappa = \varkappa_0 + 7.53 \cdot 10^3\ \rho_n \gamma^{1/2}$$

(where $\kappa_0 = 0.325$ for pure tin in accordance with the data of Chambers[17]) was 0.33. The measurements were made on unannealed samples as well as on samples annealed at $T = 170°C$ for 10–15 hours.

4. CRITICAL CURRENTS

Plots of the average critical current density against the external field were obtained for samples with different thicknesses and at different temperatures, before and after the annealing. Typical curves of this type are shown in Fig. 2. We call attention to the following features.

1) The critical current differs from zero in a relatively wide field interval in the region of the existence of the intermediate state. For samples of thickness $d \geq 0.1$ mm, the critical currents differ from zero up to fields $H^{max} \approx 0.7 H_c$. For smaller thicknesses,

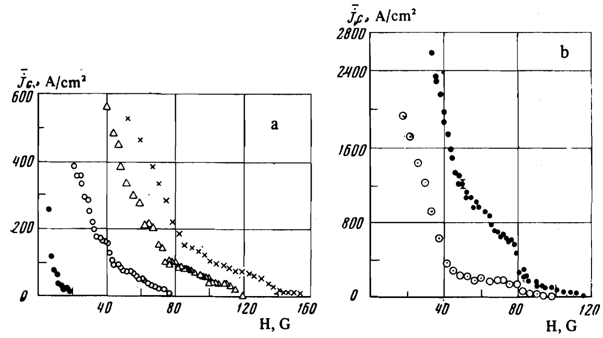

FIG. 2. Dependence of the average critical current density on the external magnetic field: a—sample No. 6, d = 0.1 mm, ●—T = 3.48°K, O—T = 2.93°K, Δ—T = 2.49°K, X—T = 2.12°K; b—sample No. 5, d = 0.04 mm, T = 2.12°K, ●—prior to annealing, O—after annealing.

$H^{max} \approx 0.5H_c$. When the transition temperature is approached, H^{max} decreases. Annealing does not noticeably change the value of H^{max}.

By varying the initial current I_0, we can reach a critical state at different values of the external field, namely such for which the initial current I_0 is critical. In other words, the current in the circuit does not attenuate in fields for which the sample is known to be in the intermediate state. This means, first, that up to fields $H \sim H^{max}$ the intermediate-state structure is a system of isolated regions of the normal phase in a superconducting matrix and, second, that for the motion of such a structure it is necessary to have a current of finite magnitude, i.e., the pinning effect obviously is present.

At $H > H^{max}$, one observes the usual ohmic loss mechanism, since there are no superconducting through channels in the sample along the current direction.

At first glance it may seem strange that the field interval in which the critical current differs from zero extends to field values such that the concentration of the normal phase in the sample is large. It should be remembered, however, that the development of the intermediate state in our case always takes place in the presence of an electric current. The establishment of some topology of the intermediate state is apparently governed in many respects by the interaction of the regions of the normal phase with the structural inhomogeneities in the sample. As a result, one of many possible metastable states is produced. The presence of current contributes to the establishment of a more equilibrium state. The current, in particular, can prevent the coalescence of normal regions and by the same token hinder the formation of continuous normal layers up to larger values of the external magnetic field.

The foregoing is confirmed, for example, by the results of Solomon[8], who observed, using the powder method, a "filamentary" structure of the intermediate state in the presence of current, up to fields $\sim 0.5H_c$. In fields stronger than $0.7H_c$, solid layers of the normal phase appeared, oriented perpendicular to the current direction.

2) The critical current density depends on the sample thickness. Other conditions being equal, it increases with decreasing thickness, and remains at the same time smaller by two or three orders of magnitude than those values that are typical of type-II superconductors subjected to cold working. This indicates that the pinning is relatively ineffective in the intermediate state. It is clear that an important role is played here by the ratio of the dimensions of the normal regions to the dimensions of the inhomogeneities of the structure.

We shall discuss this question in greater detail later on, in connection with the discussion of the dependence of the average pinning force on the sample thickness.

3) Annealing does not noticeably influence the critical current density in the case of "thick" samples ($d \sim 1$ mm). With decreasing thickness, however, the influence of the annealing becomes more and more significant. Thus, annealing of a sample 0.04 mm thick led to a decrease in the critical current density by approximately one order of magnitude in the region of weak fields (Fig. 2b).

5. PINNING FORCES

The dependence of the average critical current density $\bar{j}_c$ on the applied magnetic field contains information on the so-called volume pinning force, i.e., on the average density of the force of the interaction of the magnetic flux in the sample with the structure defects. Usually the $\bar{j}_c(H)$ are cited as the principal characteristics of type-II superconductors in the resistive state. These curves are used to draw qualitative conclusions concerning the relative pinning force under various conditions. It is easy to show, however, that by measuring $\bar{j}_c$ as a function of H and by using the concept of the critical state[11] it is possible in a number of cases to estimate the volume pinning force. The penetration of a magnetic field into a superconductor (in the form of Abrikosov vortices in the mixed state or in the form of magnetic-flux tubes so long as such a structure is realized in the intermediate state), always starts at the surface of the sample. If nothing prevents such a magnetic structure from moving into the interior of the sample, then the distribution of the magnetic induction in the interior is homogeneous and corresponds to a reversible magnetization curve, B(H).

Inhomogeneities in the sample, generally speaking, hinder the penetration of the vortices (or flux tubes) to the interior, and this leads to a nonzero gradient in the density of their distribution. Under these conditions, a magnetic-pressure force proportional to this gradient is produced. By critical state is meant a state in which the magnetic-pressure force at each point inside the superconductor is equal to the pinning force F_p. If the superconductor boundary is a plane parallel to the external field, then the critical state corresponds to satisfaction of the equation

$$F_p(x) = -\frac{b(x)}{4\pi\tilde{\mu}}\left[\frac{\partial b(x)}{\partial x}\right] \quad (1)$$

(the x axis is perpendicular to the surface, $F_p(x)$ is the pinning force per unit volume at the point x; b(x) is the local value of the magnetic induction; $\tilde{\mu} = dB/dH$ is the derivative of the reversible magnetization curve).

Relation (1) determines the critical value of the magnetic-induction gradient $(\partial b(x)/\partial x)_c$ at each point inside the superconductor. This gradient corresponds to a critical current density

$$j_c(x) = -\frac{c}{4\pi}\left[\frac{\partial b(x)}{\partial x}\right]_c.$$

Thus,

$$F_p(x) = \frac{b(x)}{c\tilde{\mu}} j_c(x). \quad (2)$$

The average pinning force $\bar{F}_p$ can be determined if one knows the dependence of j_c on b. As shown by Fietz and Webb[18], this dependence can be reconstructed in a certain approximation from data on the magnetization curves of the samples. Thus, by measuring the magnetic properties of the samples we can determine the average pinning force. When measuring the electric properties, when the critical state is reached in the presence of a transport current, it is easy to show, by assuming the current $j_c(x)$ to have a nearly uniform distribution (Bean[12], Fietz and Webb[18]), that the average pinning force can be estimated with the aid of (2) by using the $\bar{j}_c(H)$ dependence obtained directly from experiment. In particular, for a type-I superconductor in the form of a thin plate in a perpendicular field, $\bar{F}_p$ can be estimated from the formula

$$\bar{F}_p = \bar{j}_c H / c. \qquad (3)$$

We have estimated $\bar{F}_p$ in accordance with formula (3). By way of example, Fig. 3 shows the values of $\bar{F}_p$ as functions of H/H_c for two samples of different thickness before and after annealing. A characteristic feature of curves of this type is the presence of a maximum in a range of fields from $0.1H_c$ to $0.5H_c$, depending on the sample thickness. (In Fig. 3a, there is no maximum for the sample 0.04 mm thick; it can only be stated that such a maximum should, in any case, occur at $H/H_c \lesssim 0.1$, since the pinning force should vanish with vanishing of the intermediate state.) The presence of a maximum becomes understandable if it is recognized that the volume pinning force, on the one hand, should increase with increasing field, since the number of normal regions per unit volume increases. On the other hand, the pinning effectiveness apparently decreases with increasing field. The reason for this may be the decrease of the local force exerted on the normal region by the defect. In addition, the dimensions of the normal regions themselves increase with increasing field, and these regions may merge with one another. This should make them less sensitive to disturbances of the structure. With increasing temperature, the value of $\bar{F}_p$ pertaining to one and the same value of the relative field H/H_c decreases, and becomes vanishingly small in practice at $T \gtrsim 0.9T_c$.

Figure 4 shows the dependence of the maximum pinning force $\bar{F}_p^{max}$ on the sample thickness at different temperatures before and after annealing. We note, first, that $\bar{F}_p^{max}$ increases with decreasing sample thickness. One can clearly distinguish here between two thickness ranges: at $d > d_c$ ($d_c \sim 0.1$ mm) one observes a relatively small increase of the pinning force with decreasing thickness. At $d < d_c$, the pinning force increases sharply with decreasing thickness. With increasing temperature, the curves shift towards smaller values of $\bar{F}_p^{max}$, but the value of d_c and the character of the curves remain practically unchanged. Another interesting feature is that the influence of annealing is not the same for "thick" and "thin" samples. For $d \lesssim d_c$, i.e., in the same region where a sharp dependence of $\bar{F}_p^{max}$ on the thickness appears, annealing leads to a decrease in the pinning force by a factor of several times. At $d > d_c$, the annealing is not very effective and at thicknesses $\gtrsim 1$ mm it exerts no noticeable influence on the pinning force at all. This indicates that in the former case the pinning is due to disturbances in the structure in the interior of the sample, whereas in the latter case it is due to surface inhomogeneities.

The interaction of the normal regions with the structure disturbances can be appreciable only if the characteristic dimensions of these regions are commensurate with the dimensions of the disturbances. Experimental data obtained by Träuble and Essmann[7], the data of Solomon[8], and the measurements of Goren and Tinkham[4] indicate that the characteristic dimensions a_n of the normal regions can range from several microns to several hundred microns, increasing with increasing field and with increasing sample thickness. Goren and Tinkham[4] have compared the experimental values of a_n with the results of their theoretical estimates of this value within the framework of the "filamentary model" proposed by them, and also with the known Landau calculation[1] for the layered model of the intermediate state. In the field interval $0.2H_c < H < H_c$, both models give approximately the same estimates for a_n, which are in fair agreement with the experimental data. To estimate a_n in accordance with our experiments, we used the results of Goren and Tinkham's simplified calculation[4], according to which

$$a_n = [2\Delta(T) d / h(1-h)(1-h^{1/2})]^{1/2}, \qquad (4)$$

where d is the sample thickness, $h = H/H_c$ is the rela-

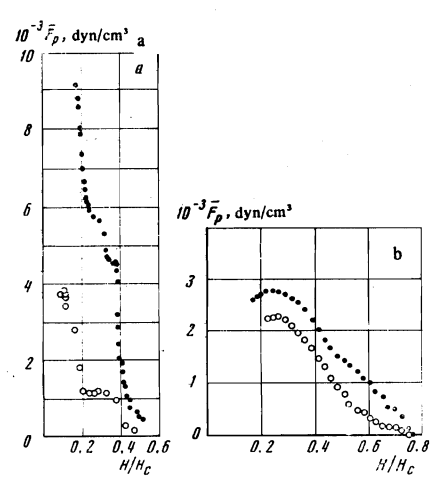

FIG. 3. Dependence of the average pinning force on the relative magnetic field: a—sample No. 5, d = 0.04 mm, T = 2.12°K, ●—prior to annealing, ○—after annealing; b—sample No. 7, d = 0.4 mm, ●—prior to annealing, ○—after annealing.

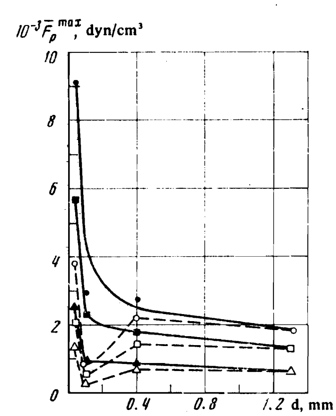

FIG. 4

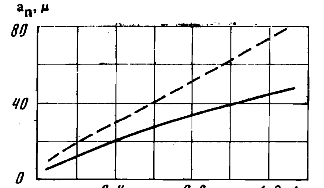

FIG. 5

FIG. 4. Maximum pinning force vs. sample thickness at different temperatures. T = 2.92°K: ▲—prior to annealing, △—after annealing; T = 2.49°K: ■—prior to annealing, □—after annealing; T = 2.12°K: ●—prior to annealing, ○—after annealing.

FIG. 5. Results of estimation of the characteristic dimensions of the normal region as functions of the sample thickness: solid line—T = 2.12°K, dashed—T = 3.48°K.

Sample thickness, mm	Average grain dimension, μ	
	Prior to annealing	After annealing
0.04	4.5±1.2	10.6±2.4
0,1	5.7±2.2	13±3
1.3	10±2	27±4

tive magnetic field, and $\Delta(T)$ is a surface-energy parameter, the value of which for tin was taken by us from the data of Cody and Miller[19].

Figure 5 shows the results of the calculation of a_n as a function of the sample thickness in accordance with formula (4). (In the calculation of a_n, we chose those values of h for which the average pinning force is maximal for a given sample thickness.) The values of a_n lie in the interval from ~5 to 80 μ. The value of a_n increases with increasing temperature, but for thin samples ($d \lesssim 10^{-1}$ mm), the influence of the temperature is negligible and the "critical" sample thickness d_c corresponds to one and the same value $a_n \sim 10\ \mu$. This allows us to assume that the maximum dimensions of the defects in the sample volume that are effective for pinning are of the same order of magnitude. This is precisely why a decrease in the sample thickness below d_c gives rise to a sharp increase of the pinning force. For the same reason, annealing is effective only in the case of thin samples.

The normal regions can apparently be pinned by dislocation grids. One cannot exclude the possibility that the grain dimension also influences the pinning effectiveness. The table lists the average grain dimensions revealed by etching of the sample surface. One can assume that if the grain boundaries do exert an influence on the pinning, this takes place only in sufficiently thin samples, when the dimensions of the normal regions become smaller than the grain dimensions. Annealing leads to an increase of the average grain dimension, and should therefore increase the contribution of this type of pinning. The fact that the pinning force decreases after annealing in thin samples offers evidence favoring the assumption that the principal role is played by the dislocation density.

6. CONCLUSION

In conclusion, we wish to note one circumstance which in our opinion is of fundamental significance.

If we disregard applications, then the pinning problem should be taken to mean the search for mechanisms that determine the interaction between the magnetic flux in a superconductor and various types of defects or inhomogeneities in its structure. The solution of this problem encounters two main difficulties. In a real situation one deals with the simultaneous action of different mechanisms. To determine the contribution of each of them it is necessary to satisfy simultaneously at least the following conditions: a) the presence of samples with controllable amounts and types of defects, b) the use of control methods to obtain exhaustive quantitative information on the structure of the investigated samples.

The second difficulty is of fundamental character and casts doubt on the very possibility of uniquely interpreting the experimental data. The point is that in experiments one always estimates, in one manner or another, the pinning force F_p averaged over the volume. It is necessary to extract from these data the value of the local interaction force f_p between the vortex and the given defect. It might seem that, having a sample with a strictly defined type of defects, knowing their concentration and the vortex concentration, there would be no difficulty in connecting the local pinning force with the volume force. However, as shown by Labusch[20], this connection is not so simple and should contain the elastic properties of the Abrikosov vortex lattice. If the vortex lattice were absolutely rigid, then the averaging of the local forces exerted on the vortices by the statistically distributed system of defects would set such a vortex lattice in motion under the influence of a vanishingly small external force, i.e., $\overline{F}_p = 0$, and the critical current in type-II superconductors would always be equal to zero. However, as a result of the interaction of the vortices with the defects, the vortex lattice becomes deformed. The response of the lattice to the local force of interaction with each defect depends on the magnitude of this force and on the elastic properties of the lattice as a whole. As a result, the average pinning force turns out to be different from zero and is expressed in terms of the local force of interaction of the vortices with the defects and a combination of the elastic moduli of the vortex lattice. Calculation of the concrete form of this coupling encounters considerable difficulties. The calculations of Labusch[20] and also of Fietz and Webb[18] lead to a quadratic dependence of $\overline{F}_p$ on f_p. All these calculations, however, are based on a large number of rather arbitrary assumptions, and hardly allow us to regard the problem as solved.

The "filamentary structure" in the intermediate state of type-I superconductors differs appreciably from the vortex structure of the mixed state. First, each such filament (flux tube) is a normal region of macroscopic size, and the period of the structure greatly exceeds the depth of penetration. This allows us to assume that the filamentary structure, regarded as a unit, acts as an absolutely inelastic system in the course of interaction with the defects. In other words, in this case there is no problem of the coupling between the volume and local pinning forces. Even this circumstance alone enables us, in our opinion, to state that type-I superconductors can be suitable objects for research from the point of view of the problem of pinning as a whole. Second, since the dimensions of the "flux tubes" can vary considerably, depending on the external conditions, it becomes possible to "sift out" the pinning centers that are ineffective for these characteristic dimensions. In contrast to type-II superconductors, this enables us to realize in experiments conditions such that the sample behaves as if there were no defects in it or as if it contained only defects of one kind.

From this point of view, type-I superconductors may turn out to be useful, for example, in the study of the mechanism of pinning on the boundaries between superconductors and vacuum or between superconductors and normal metals, a mechanism considered by Shmidt[21].

One must not forget, of course, that the quantitative characteristics of the interaction between the "flux tubes" and the defects or the interface should differ from those to which theoretical estimates lead for the vortex, although the interaction mechanisms themselves hardly change, since in the final analysis we deal in both cases with interactions of similar nature. The need for having such estimates as applied to type-I superconductors is obvious.

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