EFFECT OF PRESSURE ON THE SUPERCONDUCTING PARAMETERS AND CRITICAL SURFACE CURRENT IN NIOBIUM

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The effect of hydrostatic pressure P up to 16 kilobars has been investigated on the transition temperature T_c to the superconducting state, the critical surface current j_c^S, and the second (H_{c2}) and third (H_{c3}) critical fields in niobium. The measurements were made by the harmonic-analysis method in single-crystal samples of niobium with a resistance ratio $\rho(300°K)/\rho(4.2°K) = 20$ and for various processings of the surface. The maximum change in T_c for P = 16 kbar does not exceed 0.1°K. The values of H_{c2} and H_{c3} decrease approximately linearly under the influence of pressure with respective rates of 11 and 20 G/kbar. The critical surface current decreases reversibly with compression, the maximum pressure effect (~50% at P = 16 kbar) being observed in the sample with the largest value of critical surface current at P = 0. It is shown that the observed strong decrease of surface current is not explained by the theories of Abrikosov, and Fink and Barnes.

1. INTRODUCTION

VERY few studies have been made of the effect of hydrostatic pressure P on the critical parameters (I_c, H_{c2}) of superconductors of the second kind. Studies are known of the effect of pressure on the critical current I_c only in Nb_3Sn (up to 1.75 kbar)[1] and in Nb-Zr alloys (up to P = 20 kbar)[2] and on the parameter κ in the Ginzburg-Landau theory[3] only in In-Tl alloys.[4] The present work is devoted to the experimental study of the pressure effect up to P = 16 kbar on the critical surface current j_c^S in niobium and to determination of the dependence of the second (H_{c2}) and third (H_{c3}) critical fields on pressure P.

The theory of ideal (reversible) superconductors of the second kind has been developed by Abrikosov.[5] The properties of superconductors of the second kind whose crystal structure is not uniform differ strongly from properties of ideal superconductors. In particular, ideal superconductors of the second kind in a mixed state cannot carry a volume current without loss, and in superconductors of the second kind with nonuniform structure, a high critical current density is often observed. The nonequilibrium properties of superconductors of the second kind are usually explained on the basis of the ideas of critical state.[6-8]

Existence of a surface superconducting layer in a parallel magnetic field in the interval $H_{c2} < H < H_{c3}$ (when the volume of the massive sample is in the normal state) was predicted by Saint-James and de Gennes.[9] According to their article, the critical field H_{c3} is determined by the relation

$$H_{c2} = 1.69\, H_{c3} \qquad (1)$$

Investigation of surface superconductivity (see, for example, ref. 10) has shown that a surface superconducting layer is capable of carrying a nonzero superconducting current. By means of an approximate solution of the Ginzburg-Landau equations, Abrikosov[11] (and later Park[12]) obtained the following expression for the critical surface current density j_c^S:

$$j_c^s = \frac{5H_{c2}}{3\sqrt{3\pi}\,\varkappa^2}\left(1-\frac{H}{H_{c3}}\right)^{3/2}\left[\frac{a}{\text{cm}}\right]. \qquad (2)$$

Fink and Barnes[13] considered the fact that in real, massive samples (for example, in cylinders) the surface forms a multiply connected superconducting system and in this case the addition to the free energy arising from screening of the external field can comprise an appreciable part of the total energy. If magnetic energy is taken into account, the theory of Fink and Barnes leads to an expression for j_c^S:

$$\frac{4\pi}{c}j_c^s = \eta\frac{H_c}{\varkappa}\left(\frac{2\lambda}{R}\right)^{1/2}\frac{\Delta}{\xi}\left[\frac{f(R)}{\psi_0}\right]^2, \qquad (3)$$

where $\eta \sim 1$, λ is the depth of penetration of the magnetic field, and R is the diameter of the cylindrical sample. Numerical values for the quantity $(\Delta/\xi)[f(R)/\psi_0]^2$ as a function of H/H_{c2} can be found in refs. 13 and 14.

The value of j_c^S calculated from Eq. (2) usually exceeds the experimentally observed value by an order of magnitude. The Barnes-Fink theory agrees better with the experimental data. A significant disagreement is observed only for fields close to H_{c3}.[10]

In order to explain the disagreement between experimental data and theory[11-13], Hart and Swartz[15] proposed a surface-pinning model. According to this model the magnetic field component normal to the surface, which the early theories in general did not take into account, plays an important role. The presence in real experiments of the normal magnetic field component leads to the fact that the magnetic flux intersecting the surface is quantized. The surface defects form pinning centers, on which quantized vortices of magnetic flux are pinned. A more highly developed theory of surface pinning, which qualitatively agrees with the experimental data, is given in the work of Akhmedov, Karasik, and Rusinov.[16,17] For example, Akhmedov's dissertation[18] reports observation of a decrease in the critical surface current in very pure niobium by several orders of magnitude and establishes a direct connection of the quantity j_c^s with the quality of the surface.

EXPERIMENTAL METHOD

Hydrostatic pressure up to 20 kbar was produced with a multiplier using kerosene-oil mixture as the pressure-transmitting medium.[19] The pressure in the working channel of the multiplier was determined from the displacement of the superconducting transition temperature in a tin manometer, which was measured by a induction method.[20] The external magnetic field was produced by superconducting solenoids.

In order to measure the critical volume and surface currents j_c^v and j_c^s, a contactless harmonic-analysis method was used. A cylindrical sample was placed in a modulated longitudinal magnetic field $H + h_0 \cos \omega t$ and a signal V(t) was measured in a measuring coil wound on the sample.

The penetration of an alternating field into a hard superconductor has been investigated theoretically (and experimentally) by Bean.[6] The measurements made by us showed that Bean's calculations are applicable also for massive superconductors of the second kind (for a field $H < H_{c2}$), if in addition we take into account the screening role of the surface. These results will be published separately. The critical volume current is determined from the amplitude V_1 of the first harmonic of the signal V(t) from the formula[1)]

$$V_1 = 17.05 \cdot 10^8 \frac{NR\nu}{j_c^s} (h_0 - h_1)^2 \tag{4}$$

(where N is the number of turns in the measuring coil, R is the sample diameter, and ν is the frequency of the alternating magnetic field), which differs from Bean's formula in that the "effective amplitude" of the alternating field $h_0 - h_1$ enters into Eq. (4). The field h_1 is determined by the screening surface currents.

The density of the critical surface current for $H > H_{c2}$ is determined from the condition[10]

$$^4/_{10}\,\pi j_c^s\,(H) = h_k(H) = {}^1/_2 h_0 \quad \text{for} \quad V_1'(H) = {}^1/_2 V_0, \tag{5}$$

where V_1 is the amplitude of the real part (i.e., coinciding in phase with dh/dt) of the first harmonic of the signal V(t); V_0 is the amplitude of the signal V(t) for a field $H > H_{c3}$.

A diagram of the apparatus is shown in Fig. 1. The variable magnetic field $h_0 \cos \omega t$ is produced by a coil wound of copper wire. The coil is placed inside the superconducting solenoid and is supplied from a type GZ-33 audio-frequency generator. The variable current through the coil is measured by a voltmeter (type VZ-13) and a standard resistance. The measuring coil K_1 (Fig. 1) of 20-micron copper wire with 300 turns is close-wound on the sample. Use of a multilayer measuring coil is undesirable in measurements under pressure and in measurements in magnetic fields less than H_{c2}.

A parasitic signal is produced as the result of pres-

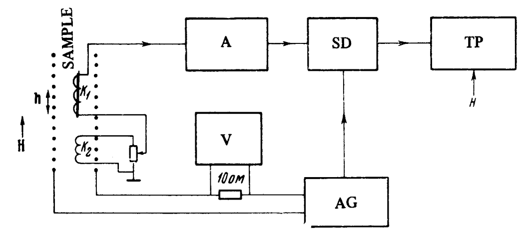

FIG. 1. Experimental arrangement for measurement by the harmonic-analysis method: A – amplifier, SD – synchronous detector, TP – two-dimensional plotter, AG – audio generator, V – voltmeter.

ence of a gap between the measuring coil and the sample. For this geometry in measurements of the critical surface current above H_{c2} this signal is so small that it can be neglected. However, in measurements below H_{c2}, when the depth of penetration of the alternating field is small in comparison with the diameter of the sample, the parasitic signal becomes comparable with the working signal. The parasitic signal was compensated by an auxiliary coil K_2 (Fig. 1) with 10 turns, with which it was possible to reduce the parasitic signal by an order of magnitude. The working frequency was 120 Hz. At this frequency the attenuation of the alternating field in passing through the wall of the pressure multiplier was small.

The signal from the measuring coil was amplified and analyzed by a U2-6 measuring amplifier and SD-1 synchronous detector. The SD-1 detector was adjusted to the phases of signals V_1' and V_1'' for H = 0, with respect to the phase of the parasitic signal.

The measurements were made in single-crystal samples of niobium of cylindrical shape, 2 mm in diameter and 12 mm long, with a resistance ratio $\Gamma = \rho(300°K)/\rho(4.2°K) \sim 20$. The surface of each of the cylinders was processed by different means. The surface of sample III/3 was mechanically polished and then etched in a solution of 75% HNO_3, 25% HF. Sample III/4 was mechanically polished, etched, and then subjected to electropolishing. Sample III/5 was mechanically polished, chemically etched, and electropolished, and then annealed in a vacuum of $\sim 10^{-8}$ mm Hg at t = 1500°C for two hours.

Above H_{c2} we measured V_1' and sometimes V_1'' for fixed amplitudes of the alternating field and with a slow increase of the external field. The values of V_1' and H were recorded on a two-dimensional plotter. Below H_{c2} we measured the amplitude of the first harmonic V_1 and third harmonic V_3 as a function of the external field H for fixed values of h_0, and also the harmonics V_1 and V_3 as a function of the alternating field h_0 for fixed values of H.

EXPERIMENTAL RESULTS

The critical temperature T_c in all samples was 9.1°K. The maximum change in T_c with pressure up to P = 16 kbar did not exceed 0.1°K. The resistivity ρ_n in the normal state at T = 4.2°K was determined from the skin depth and for samples III/3 and III/4 was 8×10^{-7} ohm-cm, and for sample III/5 was 7.1 $\times 10^{-7}$ ohm-cm.

The second critical field H_{c2} for P = 0 was determined by various methods. The values obtained are in

1)We use the subscripts to designate the harmonic number (V_1, V_3), and the superscript primes (V′ and V″) to represent respectively the real and imaginary components.

good agreement with each other and with the Gor'kov-Goodman formula.[21] For samples III/4 and III/4 the parameter κ was ≈ 1.71 and $H_{c2} = 3.82$ kG, and for sample III/4, κ was 1.66 and $H_{c2} = 3.7$ kG. Under pressure the field H_{c2} was determined from the location of the peak in the curves of $V_1'(H)$ with increasing magnetic field for sufficiently small values of h_0 ($h_0 < \sim 1$ G).[10,22] This method is sufficiently accurate to determine the dependence of H_{c2} on P. The second critical field H_{c2} in the niobium samples studied decreases with pressure with a rate 11 G/kbar (Fig. 2).

The third critical field H_{c3} was determined from the moment of disappearance of the critical surface field (in the linear approximation). For P = 0 the ratio H_{c3}/H_{c2} in the samples III/3 and III/4 was 1.75 and in sample III/5—1.7. The critical field H_{c3} decreases approximately linearly with pressure with a rate 20 G/kbar (Fig. 2). The ratio $(\partial H_{c3}/\partial P)/(\partial H_{c2}/\partial P)$ was ~ 1.9.

The critical surface current density $j_c^S(H)$ for $H > H_{c2}$ was determined from curves of $V'(H)$ according to formula (5). Figure 3 shows j_c^S as a function of H for different pressures P. The critical surface current in all samples studied decreases reversibly with pressure. The largest pressure effect on j_c^S was observed in sample III/3. In this sample the critical surface current for P = 16 kbar decreases by approximately a factor of two for $H = 1.2\ H_{c2}$. It is interesting to note that the greatest pressure effect corresponds to the highest critical surface current density for P = 0. (At a field H = 4 kG and P = 0 the critical surface current density in sample III/4 was 4, in sample III/4—2.5, and in sample III/5—2 A/cm; see Fig. 3). The j_c^S curves shift toward lower values of the magnetic field H with increasing pressure (Fig. 3) as a result of the fact that the critical fields H_{c2} and H_{c3} decrease with pressure.

The nature of the penetration of the alternating field into the superconductor below H_{c2} is shown in Fig. 4. In the same figure we have shown the first and third harmonic as a function of the magnetic field H for a constant amplitude of alternating field ($h_0 = 46.5$ G). In the curves of $V_1(H)$ and $V_3(H)$, minima are observed at a field $H = H_p$ (denoted by the arrows ↑ in Fig. 4). The minima appear only for rather large values of h_0, but their position does not depend on the amplitude of h_0. The values of H_p are close to H_{c2} but do not coincide with them. In fields above H_p the amplitude of the first harmonic increases rapidly (curve 2 in Fig. 4), approaching the value of V_0 for $H > H_{c2}$ (a weak screening effect of surface currents appears in the function $V_1(H)$). The nature of the function $V_3(H)$ changes considerably near H_{c2} (Fig. 4, ↓). The existence of values of V_3 different from zero above H_{c2} is the consequence of distortion of the signal by the screening surface currents. We note that the features in the $V_3(H)$ curves (↓) for all values of P agree with $H_{c2}(P)$ determined by another method (from the maximum in the $V_1'(H)$ curve.

The field H_p decreases monotonically under the influence of pressure with approximately the rate as H_{c2} (Fig. 2). The minima in the curves of $V_1(H)$ and $V_3(H)$ at $H = H_p$ correspond to the peak effect—the maximum in the curve of the transport critical current j_c as a

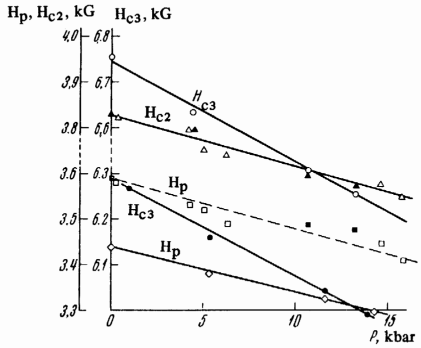

FIG. 2. Pressure dependence of H_{c2} (Δ – sample III/3, ▲ – III/4), H_{c3} (○ – sample III/4, ● – III/5), and field values H_p corresponding to the peak effect (■, □, and ◇ – respectively samples III/3, III/4, and III/5).

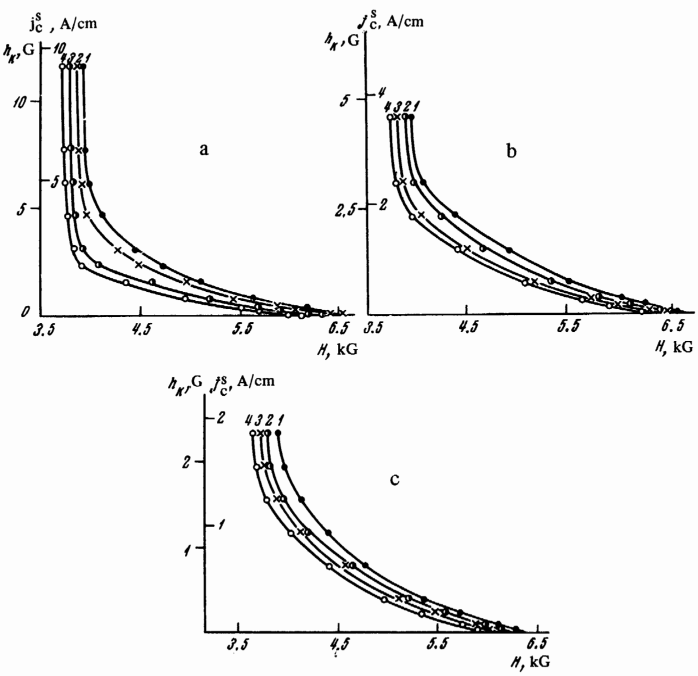

FIG. 3. Critical surface current density j_c^S and corresponding values of h_c as a function of field H for different pressures: a – sample III/3, curve 1 – P = 0, 2 – P = 4.4, 3 – P = 5.1, 4 – P = 14.8 kbar; b – sample III/4, curve 1 – P = 0, 2 – P = 4.5, 3 – P = 10.8, 4 – P = 14.8 kbar; c – sample III/5, curve 1 – P = 0, 2 – P = 5.4, 3 – P = 11.7, 4 – P = 14 kbar.

function of the field H, which is observed in superconductors of the second kind.[23]

The dependence of V_1 and V_3 on h_0 for fixed values of the magnetic field H indicate the following nature of the penetration of the alternating field.

A. Penetration of the alternating field into the sample begins only above a certain value h_c of the amplitude h_0 of the alternating field (Fig. 5). In the interval $0 < h_0 < h_c$ the sample volume is completely screened by the surface current from penetration of the alternating field (the straight line in Fig. 5 illustrates the incompletely compensated parasitic signal from the gap between the measuring coil and the sample). It is possible to deter-

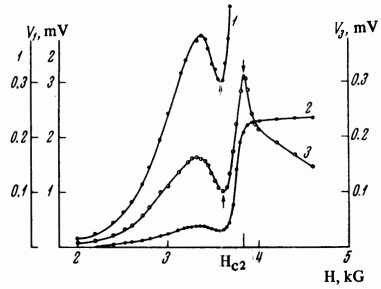

FIG. 4. V_1 (curves 1 and 2) and V_3 (curve 3) as a function of field; $h_0 = 46.5$ G and P = 0 (sample III/4).

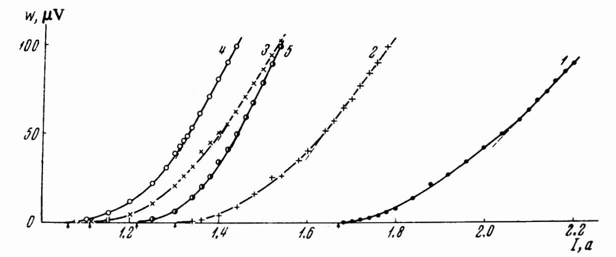

FIG. 8. Potential difference W in a sample of niobium ribbon for fixed field values and T = 4.2°K, as a function of current. Curve 1 – H = 1.2, 2 – H = 1.6, 3 – H = 2, 4 – H = 2.3, 5 – H = 2.72 kG (equal to H_p). The arrows indicate values of j_c for the corresponding magnetic fields.

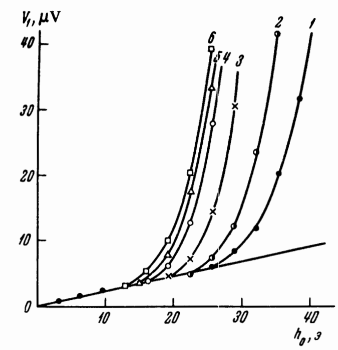

FIG. 5. Initial stage of penetration of alternating field (sample III/4) for different values of external field (below H_{c2}) for P = 0. (Straight line – uncompensated part of the parasitic signal from the gap between the coil and the sample). Curve 1 – H = 2.8, 2 – H = 3, 3 – H = 3.2, 4 – H = 3.36, 5 – H = 3.6, 6 – H = 3.68 kG.

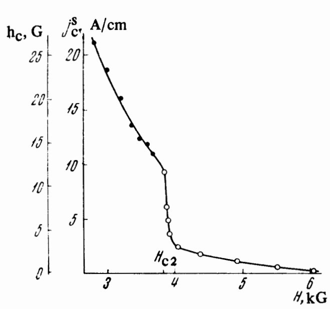

FIG. 6

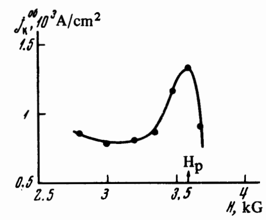

FIG. 7

FIG. 6. Critical surface current density j_c^S (and h_c) as a function of H (sample III/4) for P = 0. Below H_{c2} the value of j_c^S was determined from curves of $V_1(h_0)$, and above H_{c2} by curves of $V_1'(H)$.

FIG. 7. Critical volume current density as a function of H for P = 0 (sample III/4).

mine h_c more accurately by plotting $V_1(h_0)$ on a semi-logarithmic scale.

The density of the critical surface current, which is determined from the condition $(4/10)\pi j_c^S(H) = h_c(H)$, decreases monotonically with increasing magnetic field H (Fig. 6). Near H_{c2}, where the sample goes into the region of surface superconductivity, the critical surface current density falls sharply.

B. At large amplitudes h_c of the alternating field the dependence of V_1 on h_0 (and of V_3 on h_0) becomes quadratic. The nature of the change in the critical surface current with compression below H_{c2} is similar in sign and approximately in magnitude to its change above H_{c2}.

The density of the critical "volume" current j_c^V, which is calculated from formula (4) according to the curves of $V_1(h_0)$, is shown in Fig. 7. It can be seen that the volume component of the critical current has a maximum at a field value $H = H_p$, and therefore it is just this component which is responsible for appearance of the "peak effect." The critical volume current in the niobium samples studied apparently depends on pressure in a complicated way.

Additional measurements were made of the critical volume current (from appearance of the potential difference W in the sample) in niobium ribbon at P = 0 and T = 4.2°K. The ribbons were prepared from the same niobium as samples III/3 and III/4, by cold drawing (with a degree of deformation of 95%). The peak effect is preserved, but the field H_p after deformation decreases to a value ~2.6 kG.

From the functions W(I) at H = const (Fig. 8) we determined the differential resistivity ρ_f in the resistive state (Fig. 9). It is evident that the "features" associated with the peak effect disappear.

DISCUSSION OF RESULTS

A decrease in the second critical field $H_{c2} = \sqrt{2}\kappa H_c$ under pressure (Fig. 2) can occur as the result of a change in H_c and κ under compression:

$$\frac{\partial H_{c2}}{\partial P} = \sqrt{2}\frac{\partial \varkappa}{\partial P} H_c + \sqrt{2}\varkappa \frac{\partial H_c}{\partial P}. \tag{6}$$

The value of $\partial H_c/\partial P$, unfortunately, is unknown and therefore it is impossible to determine $\partial\kappa/\partial P$ from Eq. (6). We can only estimate its upper limit by assuming that $\partial H_c/\partial P = 0$: $|\partial\kappa/\partial P| < 5 \times 10^{-3}$ kbar^{-1}. (It is interesting to note that Fischer and Olsen[4], working with In-Tl alloy, obtained $\partial\kappa/\partial P \sim 4.5 \times 10^{-3}$ kbar^{-1}.)

According to Eq. (1) the field H_{c3} (Fig. 2) should change under pressure 1.7 times more rapidly than H_{c2}. The experimentally obtained value $(\partial H_{c3}/\partial P)/(\partial H_{c2}/\partial P) \sim 1.9$ is somewhat greater than the theoretical value. This difference can be explained, on the one hand, by the possible experimental errors and, on the other hand, by the possible effect of the change of critical surface current under pressure on the determination of the value of H_{c3}.

The critical surface current density in niobium decreases reversibly with pressure in the magnetic field interval $H_{c2} < H < H_{c3}$ (Fig. 3). The relative variation of the critical surface current density for a field $H = 1.2\ H_{c2} = 4.5$ kG is shown in Fig. 10.

The decrease in critical surface current with pressure apparently follows from the theories of Abrikosov, and Fink and Barnes. However, according to these theories the decrease should be considerably smaller than is experimentally observed. Comparison of the experimental data with theory is hindered also by the fact that the data obtained for j_c^s at P = 0 do not agree with the theoretical values. The value of j_c^s calculated from formula (2) is more than an order of magnitude greater than the experimental data. Furthermore, according to the theory of Fink and Barnes the values of j_c^s in all samples studied should be almost identical, since the parameters H_{c2} and κ are nearly the same in these samples. However, the experimental data for j_c^s in samples III/3 and III/5 differ by two to three times (Fig. 3).

The variation of j_c^s with pressure, according to the theories mentioned above, occurs as the result of change in the parameters H_c (or H_{c2}) and κ. With a decrease of H_c (or H_{c2}) the critical surface current should decrease, and with a decrease of κ should increase (see formulas (2) and (3)). It is easiest to estimate the effect of pressure on j_c^s from formula (2). Without taking into account the change (apparently a decrease) in the parameter κ with the pressure, and using the value $\partial H_{c2}/\partial P \sim 11$ G/kbar, from formula (2) for P = 15 kbar and $H = 1.2\ H_{c2}$ we obtain $\Delta j_c^s/j_c^s(P = 0) \sim 15\%$ ($\Delta j_c^s = j_c^s(P) - j_c^s(0)$). The experimental data (Fig. 10) are two to four times greater than the theoretical value (which for the same reason can only be decreased as the result of the assumption that κ does not change with pressure).

The different values of j_c^s in the niobium samples studied at P = 0 and the nature of the change of j_c^s with pressure indicate that in formation of the surface current an important role is apparently played by the surface-pinning mechanism, which is not taken into account in the theories of Abrikosov and of Fink and Barnes. From this point of view[15-18] the change of j_c^s with pressure may be associated with a reduction in the pinning force for "surface vortices" under compression. This conclusion agrees with the fact that the pressure effect is considerably less in the samples (III/4 and III/5, Fig. 10) with a more perfect surface (lower values of j_c^s).

The possibility of existence of a critical surface current at fields less than H_{c2} has been shown theoretically by Fink.[24] According to Fink's theory, j_c^s varies smoothly near H_{c2}. The data obtained by us on j_c^s as a function of H below H_{c2} (Fig. 6) agree with Fink's theory in the respect that j_c^s is observed even below H_{c2}. However, the sharp increase of j_c^s at $H = H_{c2}$ with decrease of magnetic field does not find explanation in terms of Fink's theory. A sharp rise of j_c^s near H_{c2} is observed also by Hart and Swartz[15] and is explained by them on the basis of the surface-pinning model.

The data obtained for the critical current in niobium by the harmonic-analysis method show that the peak effect in niobium is a volume effect (see Figs. 6 and 7). The field H_p decreases with compression approximately at the same rate as H_{c2} (Fig. 2). The differential resistivity ρ_f in the resistive state, which characterizes the collective motion of the quantized vortices, increases monotonically near H_p with increase of H (Fig. 9).

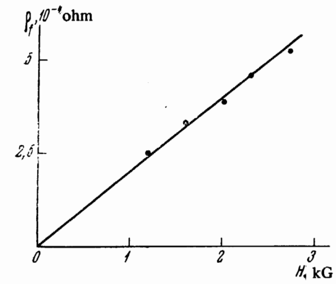

FIG. 9

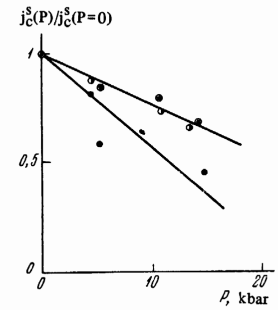

FIG. 10

FIG. 9. Differential resistivity ρ_f as a function of field in niobium ribbon at T = 4.2°K.

FIG. 10. Relative variation $j_c^s(P)/j_c^s(0)$ of critical surface current density as a function of pressure for H = 4.5 kG: ● – sample III/3, ◑ – sample III/4, ○ – sample III/5.

A possible explanation of the peak effect in niobium is as follows. Inclusion of only the rigidity of the Abrikosov lattice of quantized vortices near H_{c2}[8] cannot explain the peak effect. In this case the peak effect would be observed in all superconductors of the second kind. Therefore, in order to explain the peak effect it is necessary to take into account also the role of pinning. We will assume that in niobium the pinning centers are divided into two groups. In one group are pinning centers having a high density but producing comparatively low potential barriers which pin the vortices. In the second group are pinning centers which produce higher potential barriers than the pinning centers of the first group but which have considerably lower density. In fields $H \ll H_{c2}$ the pinning of vortices is accomplished mainly at the centers of the first group, since the centers of the second group pin an insignificant fraction of the vortices. With increase of the magnetic field the number of pinning centers of the second group does not change, but their efficiency increases considerably as the result of the increasing interaction between the vortices. One pinning center, which at fields $H \ll H_{c2}$ pins only one quantized vortex, as the field approaches H_{c2} pins a larger and larger number of vortices. As the result of the increased interaction between the vortices with increasing H, the interaction distance between the pinning centers of the second group and the quantized vortices is increased. As a result the critical current increases with increasing H.

De Sorbo[25] has shown that the peak effect is observed most frequently in those niobium samples which contain impurities (for example, oxygen) which form interstitial solid solutions. The pinning centers of the first group may be associated with impurities of this type, and the pinning centers of the second group with dislocations. Under the influence of pressure, H_{c2} decreases, and the Abrikosov lattice of quantized vortices becomes more rigid at lower field values. As the result, H_p decreases.

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