Resistive transitions and the critical fields of inhomogeneous superconducting vanadium films

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The resistive $R(H)$ transitions of vanadium films in a magnetic field are studied. Steplike anomalies on the $R(H_{\parallel})$ curves, which indicate that the samples are inhomogeneous, are observed in several films. The inhomogeneity caused by the film surface gives rise to a distinct group of electrons, whose properties differ from those of the electrons in the bulk of the film. This group of electrons has a weak superconductivity which typically has extremely low critical-current densities. A theoretical analysis of the experimental data and calculations of the critical field are carried out for a double-layer film model in the Ginzburg-Landau approximation. The results are in reasonable agreement with the experiment.

1. INTRODUCTION

Considerable attention has recently been focused on the study of the effect of inhomogeneities caused by structural defects on the properties of superconductors. In the presence of inhomogeneities, certain characteristics of a superconductor, such as the electron-electron-interaction constant g and the mean free path l, become functions of position. Inhomogeneities may give rise to several experimentally observable effects both in the thermodynamic and kinetic processes of superconducting systems. A typical example of an inhomogeneous superconductor is a sandwich consisting of two different superconductors which is used to study the effect of proximity. The critical temperature T_c and the critical fields H_c of such a sandwich have been studied extensively.[1] In superconducting structures strong, regularly spaced inhomogeneities are seen in the behavior of the critical fields and critical currents I_c. For example, a periodic modulation of the impurity density may lead to the appearance of an anomalous peak in the angular dependence $H_c(\theta)$ of the superconducting films.[2,3] This effect has been observed experimentally in columnar films of transition metals.[2,4,5] Inhomogeneities such as a periodic modulation of the film thickness give rise to a peculiar oscillatory dependence of $I_c(H)$.[6] The case in which g and l are random functions of the coordinates has been thoroughly studied in Refs. 7–9 in the case of strong and weak inhomogeneities. The most pronounced effect of these inhomogeneities is seen in the critical current I_c and the characteristic dependence of I_c on the temperature and magnetic field. The results for the critical currents, found in Refs. 8 and 9 for weak inhomogeneities, have been confirmed experimentally in Refs. 10-13.

A new type of structure-related inhomogeneity has recently been detected by Khaĭkin and Khlyustikov[14,15]: When electrons are trapped near the twinning plane in tin, indium, and gallium crystals, the transition temperatures are found to be higher than those of bulk single crystals of these metals. The superconductors whose electrons are trapped near the plane, line, and point defects have been described theoretically in Refs. 16-19. The inhomogeneities of superconductors caused by the trapped states give rise, in particular, to anomalous behavior of the critical fields.[15–18]

The boundaries of a superconducting film are macroscopic plane defects which can also render the superconducting properties inhomogeneous. In a previous study,[20] we have shown that a surface-related inhomogeneity of a sample causes a type-2 superconducting vanadium film to produce a special group of electrons, whose properties differ from those of the electrons in the bulk of the film. In this paper we report the results of a detailed study of the anomalous resistive properties of inhomogeneous films near the phase transitions in a magnetic field and the study of the critical fields H_c for different alignments of the field with respect to the film surface.

A double-layer model of the film, in which T_c differs from ξ in the layers, is used to interpret the results; the calculations are carried out within the context of the Ginzburg-Landau theory. The experimental data are in reasonable agreement with the results of these calculations.

2. PREPARATION OF THE SAMPLES AND THE MEASUREMENT PROCEDURE

The vanadium films ranging in thickness from 1000 to 3000 Å were deposited on glassceramic, glass, and fluorophlogopite substrates by a sharply focused electron gun. During the fabrication of the films, the temperature of the substrates and the vacuum were varied over the ranges 200–510 °C and $10^{-6}-6\times10^{-8}$ Torr. The deposition rate was varied within the range 5–15 Å/s. The film thickness was determined within ± 20 Å by the method of multiple-wave interferometry. The samples had the following dimensions: film width—1.5 mm, spacing between the potential contacts—5.8–7 mm; the normal resistance of the samples at $T = 4.2$ K was on the order of several ohms and $R_{300}/R_{4.2} = 4$–6. The superconducting transition temperatures of the various samples varied within the range 4.4–4.9 K, i.e., they were markedly lower than the value for pure vanadium, 5.3 K.

The resistance was measured by the four-probe method. The resistive transitions were recorded by an x-y recording potentiometer. A rotator was used to change the alignment

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of the sample surface with respect to the magnetic field direction. The alignment error was ~0.5°. Two methods were used to rotate the sample in a magnetic field: In the first case, the angle between the direction of **H** and the transport current I remained constant (90°); in the second case, a change in the angle between the plane of the film and the magnetic field was accompanied by a change in the angle between **H** and **I**. The bulk of the measurements were carried out when a 100-μA current was passed through the sample. The temperature of the helium bath was stabilized within 0.003 K over the interval $T = 1.7$–4.2 K. One of the samples was bombarded by 10-keV helium ions at room temperature while maintaining a pressure of 3×10^{-6} Torr in the accelerating chamber.

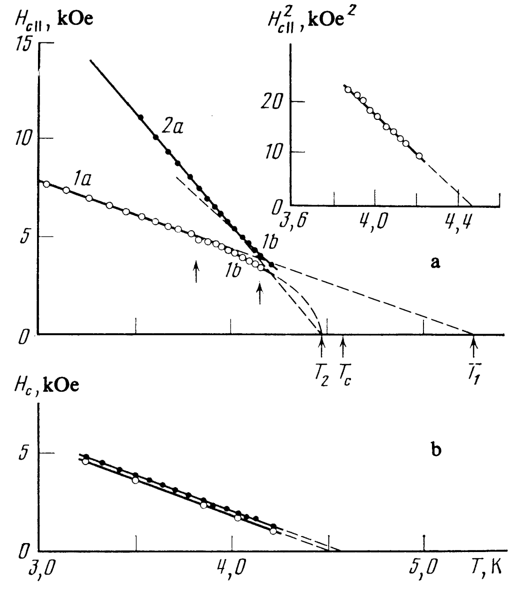

FIG. 2. Temperature dependence of the critical field. (a) The $H_{c\parallel}(T)$ curve plotted using the criterion 0.02 (1) and the criterion 0.5 (2). (b) The $H_{c\perp}(T)$ curve plotted using 0.02 (0) and 0.5 (●) (in the upper plot, curve $2b$ should be substituted for curve $1b$). The inset—The temperature dependence of $H_{c\parallel^2}$.

3. EXPERIMENTAL RESULTS

Figure 1 shows the resistive transitions in one of the samples in a parallel magnetic field. At high temperatures the $R(H_\parallel)$ curves have the usual shape, like that of the cruves for most of the vanadium films studied previously (see Ref. 21, for example). At a certain temperature T', the transition acquires a step of $V/V_n \approx 0.19$. As the temperature is lowered, the step lengthens appreciably along the field, but remains at the same level (the level changes from one sample to another).

In a field perpendicular to the film's surface, no steps have been detected down to extremely low temperatures in any of the samples studied. Also, no steps were detected in the $R(T)$ transitions which were measured at $H = 0$.

Figure 2a is a plot of the critical field $H_c\|$ as a function of temperature for one of the samples, based on the criterion $0.02\, R_n$ (i.e., curve 1, below the step) and the criterion $0.5\, R_n$ (curve 2, above the step). The inset in Fig. 2 shows the functional dependence $H_{c\parallel^2}(T)$ found from the criterion $0.02\, R_n$ for the T interval indicated by arrows in Fig. 2a. Accordingly, at high temperatures, curve 1 is well described by the dependence $H_{c\parallel} \sim (1 - T/T_2)^{1/2}$ (here T_2 is a temperature that lies below the transition temperature T_c in a zero field), while at low temperatures this curve is well described by a linear T dependence, which is extrapolated to the temperature $T_1 > T_c$. At low temperatures, curve 2 is also linear; T_2 can be found by extrapolating this curve to the field $H = 0$.

Figure 2b is a plot of the $H_{c\perp}(T)$ curve for the same sample, based on the two criteria mentioned above. As we can see in the figure, this curve has no systematic features of any sort. Steps are also seen in the $R(H)$ transitions in oblique fields. The temperature at which this feature is seen decreases as the slope angle of **H** increases with respect to the film's surface.

It has been established elsewhere[22] that stepwise transitions can be seen in two-phase samples if the transition temperature of one phase is different from that of the other and if the inclusions of the second phase are large enough to render the steps in the effect of proximity imperceptible. To show that these factors are not responsible for the $R(H)$ transitions in the films we studied, we have arranged an experiment to test a sample, to which we connected several super-conductors in series, with different T_c and H_c. A part of the vanadium film, which did not display features, was bombarded by ions (10-KeV He^+ ions, flux density 7.1×10^{16} ions/cm^2). The parts of the sample that were not bombarded with ions were found to be lined to each other only through the central part, which was subjected to ion bombardment along its entire cross section (see Fig. 3). The parameters of the test sample are given in the caption of Fig. 3. We see from the data of Ref. 23 that T_c decreases with increasing flux density of the bombarding ions and the ratio dH_c/dT increases as a result of implantation of the helium ions in the vanadium film.

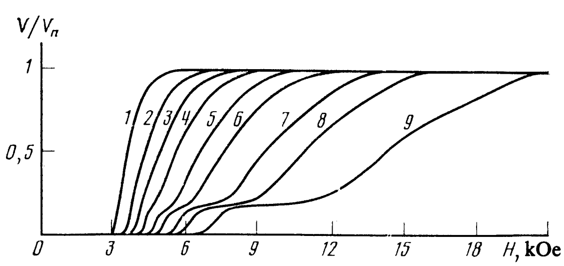

FIG. 1. The voltage across the sample versus the magnetic field $H_\parallel$ at several temperatures (K): 1—4.217; 2—4.127; 3—4.061; 4—3.985; 5—3.878; 6—3.793; 7—3.606; 8—3.451; 9—3.139.

Figure 3 shows the resistive transitions of a partially irradiated sample in a field $H_\perp$. These transitions also have a stepwise nature over an appreciable temperature interval. However, the typical values of temperatures and angles, for which the steps can be seen, are completely different from those of the samples discussed above: (1) The steps are observed at each temperature; (2) the $R(T)$ transition has a step in the absence of an external magnetic field; 3) at a given T, there is a step for each alignment of the magnetic field with respect to the film's surface. A comparison of the results presented above with the data obtained from the test sample

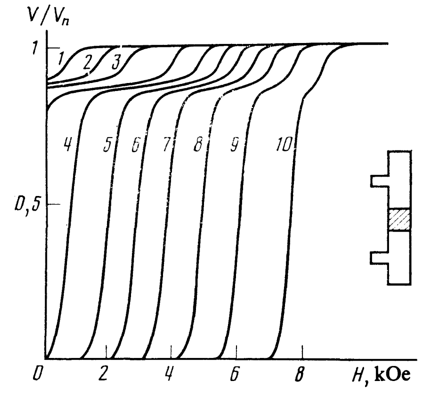

FIG. 3. The voltage across the sample, part of which was subjected to bombardment by helium ions, versus the magnetic field (the field is directed at right angles to the film). The measurements were carried out at the following temperatures (K): 1—3.829; 2—3.642; 3—3.455; 4—3.01; 5—2.869; 6—2.759; 7—2.655; 8—2.533; 9—2.369; 10—2.205. Shown at the right is the configuration of the test sample. The hatching shows the part of the sample subjected to ion bombardment. The film thickness is 420 Å. In the part of the sample subjected to ion bombardment $T_c = 3.1$ K and $dH_{c\perp}/dT = 8.5$ kOe/deg; in the part of the sample not subjected to ion bombardment $T_c = 3.987$ K and $dH_{c\perp}/dT = 4.9$ kOe/deg.

thus shows that we can rule out the bulk inhomogeneity as the cause of the anomalous behavior of the films we studied.

4. DISCUSSION AND THEORETICAL ANALYSIS OF THE RESULTS

We will show that all observable features of the resistive transitions and of the $H_c(T)$ curves can be explained by assuming that the presence of a surface—a natural, macroscopic, plane defect—causes the film to become inhomogeneous: This film produces a distinctive group of electrons, whose properties are different from those of the electrons in the rest of the film. The fact that the inhomogeneity is associated with the flat surface of the sample is evident from the particularly pronounced anomalous behavior at $\theta = 0°$ and at small values of θ. In our experiments, we have been able to determine the characteristics of each electron subsystem, because the measurements were carried out in strong magnetic fields. This was possible because the films we tested were type-II superconductors with a Ginzburg-Landau parameter[2)] $\kappa \sim 5$–9.

We easily see that curves 1 and 2 in Fig. 2 cannot be ascribed to the fields H_{c2} can H_{c3} in the case of a sample whose g constant is inhomogeneous over a narrow [in comparison with $\xi(T)$] layer D at the surface. To determine the dependence $H_{c3}(T)$, we must solve the linearized Ginzburg-Landau equation for the order parameter $\psi(x)$, $x \geqslant 0$, which is supplemented by the boundary conditions[18,25] $\psi'(+0) = \gamma\psi(0)/\xi_1$; the parameter γ characterizes the surface ($\gamma = 0$ corresponds to an ordinary[18] surface superconductivity). We can easily calculate the parameter $\gamma \neq 0$ from perturbation theory under the condition $\gamma a_H \ll \xi_1$ [$a_H = (c/2eH)^{1/2}$ is the magnetic length, and ξ_1 is the coherence length at $T = 0$]; i.e., we can easily calculate it in reasonably strong fields. The result is

$$H_{c3}(\tau)=1{,}7H_{c2}(\tau)-\zeta\frac{\Phi_0}{\xi^2}\gamma\tau^{1/2}, \quad \tau=\frac{T_1-T}{T_1}. \tag{1}$$

Here $\zeta \sim 1$ is a numerical factor, and T_1 is the critical temperature for the "bulk" electrons. A comparison of curve 2 in Fig. 2 and Eq. (1) shows that $\gamma > 0$ is the correct choice, since the two critical fields converge as the temperature is raised. Equation (1) shows that for $D \ll \xi(T)$ the low-temperature dependences of the fields H_{c2} and H_{c3} should be extrapolated to the same temperature T_1 and that their ratio H_{c3}/H_{c2} is < 1.7. These conclusions are inconsistent with the experimental data. The temperatures T_1 and T_2, which can be determined by extrapolating the linear dependences $H_{c\parallel}(T)$ to the region of small fields H, vary widely, while the field ratio is, on the contrary, larger than 1.7. We should also point out that if there is surface superconductivity, the field H_{c2} cannot be determined from the dependences $R(H)$ in the resistive measurements, since the critical current is nonvanishing in the fields $H_{c2} \leqslant H \leqslant H_{c3}$. Consequently, a model with an inhomogeneity whose scale dimension is $D \ll \xi(T)$ cannot explain these data.

A linear T dependence of $H_{c\parallel}$ at low temperatures suggests that in the experiment we have measured the critical fields H_{c3} for the two layers with different critical temperatures T_1 and T_2, different coherence lengths ξ_1 and ξ_2, and generally different thicknesses d_1 and d_2, which are appreciably greater than ξ_1 and ξ_2 (the subscripts 1 and 2 correspond respectively to curves 1 and 2 in Fig. 2). In the case of an ordinary proximity effect, under the condition $d_{1,2} \gg \xi_{1,2}(T)$, the onset of superconductivity occurs on the free surface of the film that has a smaller $\xi(T)$. In our case, we found $\xi_2(T) < \xi_1(T)$. A variation of the fields $\delta H_{c3}^{(i)}$ of the ($i = 1, 2$) layers due to a contact between them is insignificant in comparison with $H_{c3}^{(i)}$ of the single layers. This can easily be seen by using the WKB method to solve the linearized Ginzburg-Landau equation with boundary conditions at the point where the layers come in contact with each other ($x = 0$):

$$\psi'(-0)=\alpha\psi'(0), \quad \psi(-0)=\beta\psi(0). \tag{2}$$

Here α and ν are determined by the parameters of the contiguous layers.[26] The corrections $\delta H_{c3}^{(1,2)}$ to the fields $H_{c3}^{(i)}$ are

$$\delta H_{c3}^{(i)} \sim \gamma_i H_{c3}^{(i)}(T)\exp\{-2^{3/2}\pi\Phi_0^{-1}d_i^2H_{c3}^{(i)}(T)\}, \quad i=1,2; \\ \gamma_1=\beta-\alpha, \quad \gamma_2=1/\beta-1/\alpha. \tag{3}$$

The signs of δH_{c3} for layers 1 and 2 are opposite. Since the corrections to $H_{c3}^{(i)}$ are exponentially small, the transition temperature of each layer, T_1 and T_2, can be determined by linearly extrapolating the fields $H_{c\parallel}$ from the low-temperature region to small H and the values of ξ_1 and ξ_2 can be determined from the slope of the $H_{c\parallel}(T)$ curves.

We see from the data in Fig. 2 that $T_1 = 5.27$ K and $T_2 = 4.47$ K. The general transition temperature T_c, determined from the $R(T)$ curve at $H = 0$, lies between these two temperatures, as in the case of the usual proximity effect. This temperature is 4.6 K. At $T = 0$ the corresponding values of ξ_1 and ξ_2, which can be determined from the relation

$$\xi_{1,2}^{-2}=2\pi(1{,}7\Phi_0)^{-1}\,dH_{c\parallel}^{(i)}/dT,$$

are $\xi_1 = 175$ Å and $\xi_2 = 105$ Å.

The results for the critical fields presented above can thus be explained by a model consisting of two parallel layers with different T_c and ξ. We emphasize that, in contrast with the usual proximity effect in an ss' sandwich, we studied an inhomogeneous system made from a single material. Furthermore, because of the peculiar characteristics of the resistive transitions, the parallel critical fields of the two electronic subsystems can be measured concurrently in the experiment. Such measurements, in our view, are possible, because when one of the layers goes normal, the other layer, being in the resistive state, cannot shunt it. The current that passes through this layer is larger than the critical current. A reduction of the current to 1 μA does not remove the steps from the $R(H)$ curve. The density of the critical current j_c is therefore very low (we will show below that $j_c < 2$ A/cm^2); i.e., it is much lower than the density of the typical critical currents of superconductors, allowing us to assume that the superconductivity is weak.

In the case of the field $H_{c\parallel}^{(1)}$, we see a transition from a $H_{c\parallel} \sim \tau$ dependence at low temperatures to a $H_{c\parallel} \sim (1 - T/T_c)^{1/2}$ dependence at high temperatures. For homogeneous films, this sort of switch in the temperature dependence of the field $H_{c\parallel}$ is associated with the appearance of a single train of vortices.[27,28] This sort of rearrangement of the spatial distribution of the order parameter in the field occurs at the temperature T^*, with $d \approx 1.58\xi(T^*)$.[27] Hence the thickness of the layer in which an array of vortices is formed can be estimated. According to the data in Fig. 2, $T^* = (3.66 \pm 0.13)$ K. Calculations yield an estimate $d_1 \approx 600$ Å, which implies that d_1 is considerably smaller than the total width of the film, 1060 Å. Similar data have also been obtained for other samples. This situation may be viewed as evidence that a circulating eddy current cannot flow through a layer of "localized" superconductivity, because the critical current in such a layer is extremely small. Using the estimate of d_1 and the measured value of d, and assuming that the thickness of the layer in which the electrons are trapped is $d_2 \sim 450$ Å, we find an upper bound on the critical current density for the layer of trapped electrons to be $j_c < 2$A/cm^2.

Let us analyze the temperature dependence of the critical field $H_{c\perp}(T)$ for a perpendicular orientation. Performing some calculations similar to those in Ref. 29, and using the Ginzburg-Landau equation and boundary conditions (2), we easily find the equations

$$H_{c\perp} = \frac{\Phi_0}{2\pi\xi_\perp^2}\left(1 - \frac{T}{T_c}\right),$$

$$\xi_\perp^2 = \frac{T_2(T_1 - T_c)\xi_2^2 + T_1(T_c - T_2)\xi_1^2}{T_c(T_1 - T_2)}. \qquad (4)$$

Here the net superconducting transition temperature of the sample is

$$T_c = T_1T_2(\xi_1^2 + \alpha d_1\xi_2^2/\beta d_2)/(T_1\xi_1^2 + T_2\xi_2^2). \qquad (5)$$

Equations (4) and (5) are valid if the following condition is satisfied:

$$(k_id_i)^2 = d_i^2[(T_i - T)/T_i\xi_i^2 - 2\pi H_{c\perp}/\Phi_0] \ll 1. \qquad (6)$$

The quantities k_i characterize the rate at which the order parameter varies in each layer.

A calculation based on Eq. (4) yields an effective coherence length $\xi_\perp = 124$ Å, whereas from $2b$ we find $\xi_\perp = 144$ Å. Note that for the thickness d_1 and d_2 given above we have $k_id_i \sim 1$ in the temperature range 3–4 K. The experimental value of $\xi_\perp$ is therefore considered to be in satisfactory agreement with the calculated value.

The experimental data presented above can accordingly be explained by a double-layer mode. The appreciable difference between the parameters of the two layers stems from the following hypothesis concerning the structure of the films. Vanadium adsorbs all gases, especially oxygen, very effectively; the gas contaminants diffuse very effectively along the crystalline boundaries (all these films are polycrystalline substances). Consequently, if the diffusing gas impurities do not reach the substrate, the impurity-rich upper layer could acquire a granular structure, i.e., it could turn into islands of metal separated by insulating layers. The coupling between these islands would be weak. The granular films, as we know,[30,31] have much shorter mean free paths and much lower critical points than the solid films, and T_c of the transition-metal granular films is typically even lower. All these circumstances can explain the weak superconductivity of the outer surface layer, which is characterized by the parameters T_2 and ξ_2. The T dependence of the parallel critical field $H_{c\parallel}$ for this layer is represented by curve 2 in Fig. 2.

In summary, we wish to emphasize that the films we have studied clearly manifest effects which can be traced to the inhomogeneity of the sample resulting from the plane defect-the superconducting surface. We have experimentally reached a state which lies between the usual proximity effect and the superconducting state of the electrons trapped on the surface.

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[1)]For this sample the step is situated at the level $\sim 0.04\, R_n$. Note that these $H_c(T)$ curves and all similar curves are identical when either of the two methods described in the preceding section is used to rotate the sample in a magnetic field.

[2)]The values of $\varkappa$ were determined from an expression relating the measured values of H_{c1}/dT and the known derivative of the critical thermodynamic field for vanadium $dH_{cb}/dT\,|_{T_c} = -469$ Oe/K.[24]

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