- <sup>1)</sup>The term SQUID stands for "superconducting quantum interference device."
- <sup>2)</sup>In dimensional variables, these inequalities take the form  $L \ll \lambda_J$  and  $L \gtrsim \lambda_J$ , where  $\lambda_J \sim 0.1$  mm is the Josephson depth of penetration. In the text we use dimensionless quantities, with the lengths measured in units of  $\lambda_J$ , the current in units of  $j_c$  ( $j_c$  is the maximum value of the stationary current through the barrier), the field in units of  $H_J = \Phi_0/2\pi\lambda_J\Lambda \sim 1$  ( $\Phi_0$  is the flux quantum,  $\Lambda = 2\lambda_L + l$ ,  $\lambda_L$  is the London penetration depth,  $l \sim 10^{-7}$  cm is the thickness of the dielectric layer, and the flux in units of  $\Phi_0/2\pi$ .
- <sup>3)</sup>The numbering of the regions I-XII in Fig. 5 was chosen to preserve the correspondence with the numbering assumed previously<sup>16</sup> in the investigation of the behavior of a Josephson barrier with current in an external field.
- <sup>4)</sup>This agrees with the fact that the solutions with alternatingsign sections of the field H(x) turned out to be unstable also in the previously considered problems.<sup>12-14</sup> The ascending branches of the solutions 0', however, are stable.
- <sup>5)</sup>The term  $\frac{1}{2}\varphi_0 H_i$  in (14) (equal to  $\sigma H_i^2/8\pi$  in dimensional units) is the energy of the magnetic field inside the cavity. The expression for the free energy of a singly connected weak superconductor in an external field<sup>1,17</sup> contains a term  $H_e^2/4\pi$ , therefore the coefficient  $\frac{1}{2}$  is missing from the corresponding term.
- <sup>1</sup>I. O. Kulik and I. K. Yanosn, Éffekt Dzhozefsona v sverkhprovodyashchikh tunnel'nykh strukturakh (Josephson Effect in Superconducting Tunnel Structures), Nauka, 1970.
- <sup>2</sup>L. Solymar, Superconducive Tunneling and Applications, Wiley, 1972.
- <sup>3</sup>K. K. Likharev and B. T. Ul'rikh, Sistemy s dzhosefsonovskimi kontaktami (Systems with Josephson Junctions), Mos-

cow Univ. Press, 1978.

- <sup>4</sup>A. H. Silver and J. E. Zimmerman, Phys. Rev. 157, 317 (1967).
- <sup>5</sup>A. M. Goldman, J. Low Temp. Phys. 3, 55 (1970).
- <sup>6</sup>H. J. T. Smith and J. A. Blackburn, Phys. Rev. **B12**, 940 (1975).
- <sup>7</sup>T. C. Wang and R. I. Gayley, Phys. Rev. B15, 3401 (1977).
- <sup>8</sup>J. A. Blackburn, H. J. T. Smith, and V. Klein, Phys. Rev. **B15**, 4211 (1977).
- <sup>9</sup>V. V. Schmidt, Solid State Comm. 20, 295 (1976).
- <sup>10</sup>V. V. Shmidt and V. G. Stel'makh, Fiz. Nizk. Temp. 3, 1508 (1977) [Sov. J. Low Temp. Phys. 3, 722 (1977)].
- <sup>11</sup>G. F. Zharkov, Zh. Eksp. Teor. Fiz. **71**, 1951 (1976) [Sov. Phys. JETP **44**, 1023 (1976)].
- <sup>12</sup>G. F. Zharkov and S. A. Vasenko, *ibid*. **74**, 665 (1978) [**47**, 350 (1978)].
- <sup>13</sup>S. A. Vasenko and G. F. Zharkov, *ibid.* **75**, 180 (1978) [**48**, 89 (1978)].
- <sup>14</sup>G. F. Zharkov and A. D. Zaikin, Fiz. Nizk. Temp. 4, 586 (1978) [Sov. J. Low Temp. Phys. 4, 283 (1978)].
- <sup>15</sup>G. F. Zharkov, Zh. Eksp. Teor. Fiz. **75**, 2196 (1978) [Sov. Phys. JETP **48**, 1107 (1978)].
- <sup>16</sup>G. F. Zharkov, Proc. 20th All-Union Conf. on Low Temp. Phys., Moscow, part III, 1979, p. 257.
- <sup>17</sup>G. F. Zharkov and A. D. Zaikin, Kr. soobshch. po fizike FIAN, No. 7, 21 (1978).
- <sup>18</sup>G. F. Zharkov and A. D. Zaikin, Proc. 20th All-Union Conf. on Low Temp. Phys., Moscow, part III, 1979, p. 259.
- <sup>19</sup>V. L. Ginzburg and G. F. Zharkov, Usp. Fiz. Nauk 125, 19 (1978) [Sov. Phys. Usp. 21, 381 (1978)].

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# Critical phenomena in thin superconducting films

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Resistive transitions occuring in thin vanadium films with varying temperature, current, and magnetic field were investigated. It was observed that the excess conductivity  $\sigma'$  at  $T < T_c$  and  $H < H_c$  depends exponentially on the reduced parameters  $\epsilon = 1 - T/T_c$ ,  $h = 1 - H/H_c$ , and  $j = 1 - I/I_c$ . The resistive transitions satisfy the similarity law, i.e., they can be described by functions of  $\epsilon$ , h, and j which are universal for all the curves. The obtained regularities are discussed within the framework of the fluctuation theory of second-order phase transitions. The temperature and magnetic-field widths of the transition agree numerically with the predictions of the theory.

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Resistive transitions in the region of phase transformations into the superconducting state are measured quite frequently. These measurements determine the values of the critical parameters—the temperature of the superconducting transition  $T_c$  and the critical magnetic fields  $H_c$ . However, the nature of the resistive state in the region of the superconducting transition is as yet nowhere clear.

In the vicinity of the phase-transition point, a substantial role is played by fluctuations of the order parameter, which lead to noticeable deviations from the selfconsistent field theory (SFT), which makes use of an average value of the order parameter  $|\Delta|^2$ . In the critical region defined by the so-called Ginzburg number,<sup>1</sup> where the fluctuation corrections exceed the mean value of  $|\Delta|^2$ , the SFT cannot be used at all and an adequate description is obtained within the framework of similarity theory (see, e.g., the monograph of Patashinskii and Pokrovskii<sup>2</sup>).

The corrections that must be introduced in the SFT when the critical region is approached can be obtained from an analysis of the small fluctuations. In particular, the result of their contribution is an excess conductivity at  $T \gg T_c$ , first calculated by Aslamazov and

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Larkin.<sup>3</sup> This paper was followed by a large number of theoretical studies that supplemented and developed further the results of Ref. 3, and by an even larger number of experimental studies devoted to a check on the fluctuation theory at  $T \gg T_c$ .

The temperature regions  $T < T_c$  and  $T \sim T_c$  were much less investigated. For bulky pure superconductors, the fluctuation phenomena in this region are practically unobservable, since the thermodynamic width of the transition, defined by the Ginzburg number

$$G_{i} = \frac{1}{32\pi^{2}} \left(\frac{k}{\Delta C\xi^{3}(0)}\right)^{2},$$
 (1)

is very small, of the order of  $10^{-15}$ K. In this formula,  $\Delta C$  is the jump of the heat capacity at the phase-transition point,  $\xi(0)$  is the coherence length at T = 0. However, if the dimensions of the superconductor are bounded in one or two directions, then the volume in which the fluctuations build up becomes comparable with the physical volume of the superconductor. The fluctuations of the order-parameter phase in one- and two-dimensional systems are formally infinite in the entire temperature range, so that no ordinary phase transitions should occur in them.<sup>4</sup>

One can expect real thin films (which are not strictly two-dimensional, since they have a finite thickness) to constitute some intermediate case between the threedimensional and two-dimensional systems considered in Ref. 4. A decrease in one of the dimensions of the superconductor should correspond to a continuous transition between these two limits, which occurs apparently in the region where this dimension becomes comparable with the characteristic correlation radius of the order-parameter fluctuations. Such a "transition," which is accompanied by a decrease in the dimensionality, should also be accompanied by a growth of the Gi number.<sup>1)</sup> It may therefore be possible to interpret the physical properties of sufficiently thin films, in an appreciable vicinity of  $T_c$ , within the framework of the concepts of fluctuation theory (scaling).

The resistive state of thin superconducting films in the region of the phase transition at  $T < T_c$ , in the spirit of similarity theory, was considered by Kadanoff and Laramore.<sup>6</sup> The excess conductivity of the films was estimated in the same manner as for three-dimensional superconductors, but with the coherence length  $\xi$  replaced by the quantity

 $\xi' = \xi \exp\{-\varepsilon/\varepsilon_{c2}\}, \quad \varepsilon = 1 - T/T_c, \quad \varepsilon_{c2} = 2.48\hbar^2/m_{eff}^2 v_F^2 ld,$ 

which has the meaning of the correlation length in a two-dimensional system.

Another possible approach to the calculation of the conductivity near the critical region calls for introducing into the SFT corrections connected with the fluctuations. This approach was used by Langer and Ambegaokar<sup>7</sup> and by McCumber and Halperin<sup>8</sup> to calculate the conductivity of thin channels at  $T < T_c$ , and by Masker, Marcelja, and Parks<sup>9</sup> for thin films. It should be noted that the result obtained by this method in Ref. 9 agrees, apart from a number, with the result of Kadanoff and Laramore.<sup>6</sup>

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The number of detailed experimental studies of the resistive transition at  $T < T_c$  is very small. The behavior predicted in Ref. 7 was observed in Refs. 10 and 11 in thin whiskers, and in Ref. 12 in thin mercury filaments obtained by forcing mercury into natural asbestos channels. The theory for thin films was verified on granulated aluminum films in the already cited study of Masker, Marcelja, and Parks,<sup>9</sup> and very good agreement with the authors' own theory was obtained.

The fluctuation conductivity of films in the course of a phase transition in a magnetic field, in the same approximation as in Ref. 9, was calculated by Feat and Rickayzen.<sup>13</sup> To our knowledge, no one investigated in detail experimentally the resistive transition of films at  $H < H_c$ .

Fluctuation phenomena are not the only possible source of the temperature smearing of the phase transition. Inhomogeneities of the interelectron interaction constant, due to structural imperfections, should also broaden the transition region.

In connection with the study of the physics of the superconducting transition, a number of questions still remain unclear. We can indicate among them the following:

To what extent can the concepts of general fluctuation theory of second-order phase transitions, as applied to superconductors, find experimental confirmation?

What is the relative role of the thermodynamic fluctuations of the order parameter and of the inhomogeneities of the sample?

Can critical phenomena in superconductors be observed in pure form?

What is the width of the critical region and how does it depend on the superconductor parameters?

Attempting to answer these questions, we have investigated in detail resistive transitions in thin films of vanadium, which occur when the temperature, current, and magnetic field are varied. The choice of vanadium was dictated, in particular, by the fact that transition metals are more favorable objects for the observation of critical phenomena, since their Fermi energy  $\varepsilon_F$ , in view of the narrow electron energy bands, is much less than in nontransition metals, thus increasing the estimated Ginzburg number.

In a brief note<sup>15</sup> we first called attention to the fact that the excess conductivity  $\sigma'$  of vanadium films in the region of transitions satisfy in all cases the law of corresponding states, i.e.,  $\sigma'$  is described by a universal function of the reduced variables  $\varepsilon = 1 - T/T_c$ , h $= 1 - H/H_c$  and  $j = 1 - I/I_c$ . In this paper we report new experimental data, which we compare with the existing theories of the resistive transition in superconductors. An explanation is proposed for the similarity laws on the basis of the fluctuation theory of second-order phase transitions.

#### EXPERIMENTAL PROCEDURE

Thin vanadium films of thickness 300-400 Å were obtained by evaporating vanadium from a sharp-focus electron gun in a vacuum of  $1 \times 10^{-7}$  Torr. Prior to the sputtering of the samples, the vanadium was first heated and sputtered on a closed shutter. Next, samples were sputtered through a mask with characteristic dimensions 7-10 mm length, 0.5-1 mm width. The films were sputtered on substrates of glass, pyroceram, and mica heated to 200-300°C. The rate of sputtering was 30-40 Å/sec. The film thickness was measured by multiple-beam interferometry (by the Tolansky method) accurate to  $\pm 20$  Å.

The measurements were performed in a cryostat with a superconducting solenoid. The samples on a rod equipped with a rotating unit that made it possible to change the orientation of the sample relative to the magnetic field. Parallel orientation was established by obtaining film of minimum resistance, with accuracy not worse than  $0.1^{\circ}$ .

A constant temperature in the interval 4.2-2 K was stabilized in the course of the measurements with the aid of a manostatic unit with accuracy not worse than 0.001 K and was measured against the vapor pressure of the helium in the cryostat.

The resistance was measured with a four-probe method. In the investigation of the transition temperature, the voltage on the sample was measured with an R-348 potentiometer or else with an Shch-68000 digital voltmeter with high-resistance input, with accuracy not worse than  $1 \times 10^{-6}$  V. The constancy of the measuring current through the sample was monitored also with the aid of a digital instrument. In the measurement of each point, the direction of the current through the sample was reversed several times to eliminate the thermoelectric power and other parasitic signals that are odd in the current.

In magnetic fields, the R(H) dependence was registered either with the aid of two digital instruments or automatically with an xy automatic recording potentiometer. The field of the solenoid was determined from its current. Using a low-temperature Hall pickup we verified that there were no hysteresis phenomena in the solenoid itself. Nor was there any hysteresis in the plots of the resistance R against the magnetic field when sufficiently small measuring currents were used, i.e., the R(H) curves plotted with increasing magnetic field agreed with high accuracy with those plotted with decreasing field. This is also evidence of the absence of thermal overheating when the film goes over into the normal state—the hysteresis due to the over-heating appears at currents of the order of 10 mA.

The temperature  $T_c$  of the superconducting transition of the films turns out to be lower than  $T_c$  of the bulky vanadium and depends on the thickness of the film and on the vacuum conditions, in accordance with the published data.<sup>16,17</sup> Electron-microscopy investigations have shown that the films are made up of minute crystals with grain dimension ~200 Å. It follows from the electron diffraction patterns that the crystalline structure of the films does not differ from the structure of the bulky vanadium and there is no pronounced texture whatever.

### **RESULTS OF EXPERIMENTS**

Figure 1 shows typical plots of the resistive transition against temperature for different measuring currents I. It is seen that the temperature transition of the investigated films is strongly stretched out at all values of the current. The resistive transitions of the same film in a perpendicular magnetic field, at different temperatures, are shown in Fig. 2. All these transitions were plotted at one and the same current  $I=20 \ \mu A$ . They are also strongly stretched with respect to the magnetic field, and the width of the transition is practically independent of temperature.

It was noted in many studies that the width of the transition in superconducting films of A1, Sn, In, and others is changed, on account of the half-shadow effect when the edge of the film, whose thickness is less than that of the base film, is removed. Since we are interested here in the width and shape of the superconducting transition, experiments were performed on vanadium films deposited on mica substrates before and after the removal of the edges, It turned out that the normalized resistive transitions of films without the half-shadow effect coincide exactly with those obtained prior to the removal of the edges. Thus, there is no half-shadow effect for the investigated samples.

Inasmuch as the transitions are strongly broadened in all cases, it is easy to analyze their shape. From the experimental data we determined the values of the excess conductivity

$$\frac{\sigma'}{\sigma_n} = \frac{\sigma - \sigma_n}{\sigma_n} = \frac{V_n - V}{V_n}$$

for all the transitions. The results of such an analysis are shown in Fig. 3 in the form of a plot of  $\ln(\sigma_n/\sigma')$ against temperature. We have used here the same data as in Fig. 1. It is seen from Fig. 3 that at all measuring currents the resistive transition breaks up into two sections, within the limits of each of which the excess conductivity has an exponential dependence on the temperature. The parameters that characterize the exponentials differ somewhat for these two regions.







FIG. 2. Dependence of  $R/R_n$  on the magnetic field (*h* is perpendicular to the plane of the film), for the same sample as in Fig. 1, at various temperatures: 1-3.869 K, 2-3.84 K, 3-3.816 K, 4-3.795 K, 5-3.781 K, 6-3.76 K, 7-3.735 K, 8-3.709 K, 9-3.685 K.

Similar results were obtained also for other samples. In the present paper we analyze in greater detail the data for the "lower" exponentials.<sup>2)</sup> The region of their existence covers a rather wide interval—more than three orders of magnitude of  $\sigma'/\sigma_n$ , in which sufficiently exact measurements are possible. The parameters of the logarithmic curves corresponding to the considered exponential relations were determined by computer reduction of the data by least squares. The deviation of the experimental points from the obtained approximations do not exceed<sup>3)</sup>  $\delta = 1\%$ , and the residual variance does not exceed  $\delta_0^2 = 0.0011$ .

If we define the critical temperature as the temperature at which  $R = 0.5R_n$  [i.e.,  $\ln(\sigma_n/\sigma') = 0$ ], then on the basis of the data shown in Fig. 3, the experimentally observed dependence of the excess conductivity  $\sigma'(T)$ can be expressed in the form

$$\sigma_*/\sigma' = \exp\left\{-A\left(1 - T/T_*\right)\right\},\$$

where  $T_c$  and the coefficient A in the argument of the exponential are functions of the measuring current. The function A = A(I) is well approximated by  $A(I) = \gamma(1 - I/I_c)^n$ , where  $n \le 1$  and  $\gamma$  is a constant ( $\gamma > 0$ ). Substituting this expression in (2), we obtain the empirical equation

$$\sigma_n/\sigma' = \exp\{-\gamma (1 - I/I_c)^n (1 - T/T_c)\},$$
(3)

which describes resistive transitions at various currents. The value of  $\gamma$  sets the width of the transition. In the limit as  $I \rightarrow 0$ , Eq. (3) reduces to

$$\sigma_n/\sigma' = \exp\left\{-\gamma \left(1 - T/T_c\right)\right\}.$$
(4)

Thus, it follows from (3) that the resistive temperature transitions at various currents obey a similarity law,



FIG. 3. Dependence of  $\ln(\sigma_{m}/\sigma')$  at the temperature at different currents *I*:  $\triangle -20 \ \mu A$ .  $\blacktriangle -200 \ \mu A$ ,  $\bigcirc -600 \ \mu A$ ,  $\blacklozenge -1000 \ \mu A$ .



FIG. 4. Dependence of  $\ln(\sigma_n/\sigma')$  on the magnetic field  $H_{\perp}$  at different temperatures.

i.e., they can be described by a function of the reduced variables  $\varepsilon = 1 - T/T_c$  and  $j = 1 - I/I_c$  which is universal for all the curves.

We proceed now to consider the resistive transition in a magnetic field perpendicular to the plane of the film. Figure 4 shows plots, corresponding to the data of Fig. 2, of  $\ln(\sigma_n/\sigma')$  against  $H_{\perp}$  at different temperatures. Just as in the resistive transition in the case H=0, a change of the current through the sample leads to a shift of the curves at all temperatures. Therefore the resistive transitions measured in a magnetic field were compared at the same small value of the experimental currents. It follows from Fig. 4 that the excess conductivity varies exponentially also with the magnetic field. In this case two different regions of exponential variation of  $\sigma'$  with  $H_{\perp}$  are likewise observed. The slopes of the linear plots of  $\ln(\sigma_n/\sigma')$  against the magnetic field do not vary with temperature.

If, as before, the critical field  $H_{cl}$  is defined to be the value at the center of the transition, then the results shown in Fig. 4 can be represented analytically in the form

$$\ln(\sigma_n/\sigma') = a_1h, \quad h = 1 - H/H_{\epsilon_1}. \tag{5}$$

The experimentally obtained dependence of the coefficient  $a_1$  on the temperature is shown in Fig. 5 together with the plot of  $H_{c1}(T)$ . It is seen that  $a_1$  is a linear function of T and vanishes at a value  $T = T_c$  determined from  $H_{c1}(T)$ . It follows therefore that at any temperature the behavior of  $\sigma'$  in a perpendicular magnetic field can be described by the expression

$$(\sigma_n/\sigma')_{\perp} = \exp\left\{-\gamma_{\perp}(1-H/H_{\epsilon_{\perp}})(1-T/T_{\epsilon})\right\},\tag{6}$$

where  $\gamma_1$  is a constant for the given sample at the given  $I(\gamma_1 > 0)$ . We see thus that resistive transitions in a perpendicular magnetic field at various temperatures are described by a universal exponential function of the reduced variables  $\varepsilon$  and h. It should be noted that the



(2)

FIG. 5. Dependence of the critical field  $H_{cL}$  on the value of  $a_{\perp}$  from formula (5) on the temperature.

normalizing critical parameters  $T_c$  and  $H_c$ , as well as  $\gamma_{\perp}$ , are functions of the measuring current. Therefore, generally speaking expression (6) should contain, besides  $\varepsilon$  and h, also a function of j as a factor, but the form of this function has not been uniquely established. The values of  $\gamma$  in (4) and in (6) differ somewhat numerically.

The dependence of  $\ln(\sigma_n/\sigma')$  on  $H_{\parallel}$ , obtained in a magnetic field parallel to the plane of the film, is shown in Fig. 6. It is seen that at this orientation of the external magnetic field, too, an exponential dependence of  $\sigma'$  on H is observed:

$$\ln(\sigma_n/\sigma')_{\parallel} = a_{\parallel}h', \quad h' = 1 - H/H_{c\parallel}.$$

Just as in a perpendicular field, the coefficient  $a_{\mu}$  varies linearly with temperature, and consequently

$$(\sigma_{n}/\sigma')_{\parallel} = \exp\{-\gamma_{\parallel}(1-H/H_{c\parallel})(1-T/T_{c})\}.$$
(8)

In contrast to the case of  $H_{\perp}$ , in a field  $H_{\parallel}$  the slopes of the lines (see Fig. 6) vary with temperature. Nonetheless, as is evidenced from a comparison of (6) with (8), the dependences of  $\sigma_{n}/\sigma'$  on the reduced variables h and h' are identical. As can be easily seen from Figs. 4 and 6, the slope of the lines in the usual non-reduced coordinates are

## $D_{\perp} = d[\ln (\sigma_n/\sigma')_{\perp}]/dH_{\perp}$ and $D_{\parallel} = d[\ln (\sigma_n/\sigma')_{\parallel}]/dH_{\parallel}$ .

In the investigated films, the critical fields  $H_{cl}$  and  $H_{cll}$  depend differently on the temperature:  $H_{cl} \sim \varepsilon$  and  $H_{cll} \sim \varepsilon^{1/2}$ , as they should in fact for thin films. Consequently, with the plots in terms of the reduced fields having identical form, the slope  $D_{l}$  does not depend on the temperature, while  $D_{ll}$  decreases with temperature like  $\varepsilon^{1/2}$ .

Thus, both in a field  $H_1$  and in a field  $H_n$  there is a similarity law for the excess conductivity, and furthermore of the same form. It is important to note that in a perpendicular magnetic field the film should undoubtedly be in a mixed state, i.e., vortices should exist in it, whereas in the case of a parallel field no penetration of the vortices into the film takes place, since the film thickness is smaller than the critical thickness<sup>4)</sup> dc.<sup>18,19</sup> Since the resistive behavior in the vicinity of the critical field is the same in the cases of  $H_1$ and  $H_n$ , we can state that the resistive state in a perpendicular field in the region of the transition has no bearing whatever on the motion of the vortices.

It should be noted that the resistive behavior des-



FIG. 6. Dependence of  $\ln(\sigma_n/\sigma')$  on the magnetic field  $H_{\parallel}$  at various temperatures.

cribed above is observed only in sufficiently thin films, in which  $H_{cll} \sim (1 - T/T_c)^{1/2}$  near  $T_c$ , thus demonstrating the satisfaction of the condition  $d \ll \xi(\varepsilon)$  which is necessary for the two-dimensional situation. For thicker films, the resistive behavior is much more complicated.

In concluding this section, we note the following important fact. Noticeable noise is produced in the  $R/R_n$  interval from 0.1 to 0.9, at all values of the specified parameters (field, current, and temperature). The results presented above correspond to the mean values averaged over a sufficiently large time interval in measurements with a low-inertia digital voltmeter. The sufficiently good averaging of the value of  $R/R_n$  in the region of the transitions is obtained by automatic recording with a chart potentiometer. An analysis of the noise characteristics will be presented elsewhere.

### DISCUSSION OF RESULTS

It follows from the experimental data presented above that no matter how the superconductor goes over into the normal state-by changing the temperature, the current, or the magnetic field-the character of the resistive transition that occurs near the critical value of the chosen parameter is the same. In all cases an exponential dependence of the excess conductivity  $\sigma'/\sigma_n$ on the reduced variables  $\varepsilon$ , h and j is observed. The law of corresponding states is satisfied with respect to all three independent parameters specified in the experiment, and at any orientation of the magnetic field. It turns out that the resistive phenomena in the region of the transition are determined not by the character of the superconducting ordering (homogeneous state at  $H_{\mu}$ and vortical state at  $H_1$ , but only by the fact of the proximity to the critical point.

The observed regularities can be explained as a manifestation of the fluctuation effects that occur near second-order phase transitions. Some indication of the growth of the fluctuations is the observed increase of the noise in the resistive state. The fluctuation theory of the resistive transition in films at  $T < T_c$  and  $H < H_c$ predicts an exponential variation<sup>6,9,13</sup> of  $\sigma'$  with  $\varepsilon$  and h, as is in fact observed in our experiments. It seems to us that the identity of the form of the functions  $\sigma'(\varepsilon, h)$  in two qualitatively different situations—perpendicular and parallel magnetic fields-is sufficient proof that the superconducting transition is "fluctuating." The universal regularities that connect the reduced values of the conductivity, temperature, magnetic field, and current have properties that follow from the similarity hypothesis developed for secondorder phase transitions.

We proceed now to a detailed comparison of experiments with the theory in the region  $T < T_c$  and  $H < H_c$ . As already mentioned, the dependence of the excess conductivity  $\sigma'$  on the temperature for films at H=0 and j - 0 was analyzed within the framework of the fluctuation theory by Kadanoff and Laramore.<sup>6</sup> According to Ref. 6, at  $T < T_c$  we have

$$\sigma_n/\sigma' = A \exp\{-\varepsilon/\varepsilon_{c2}\}.$$
 (9)

The value of  $\varepsilon_{c2}$  agrees, accurate to a numerical factor of the order of unity, with the coefficient

$$A_{\infty} = \frac{0.92\hbar^2}{\tau_{tr}\mu p_{rd}} = \frac{1.84\hbar^2}{m_{eff}^2 v_{r}^2 ld},$$
 (10)

which enters at  $T \gg T_o$  in the function  $\sigma'/\sigma_n(T)$  obtained in the theory of Aslamazov and Larkin<sup>3</sup> for thin films<sup>5</sup>:  $\sigma'/\sigma_n = A_{\infty}/\varepsilon$ . In formula (10),  $\tau_{tr}$  is the transport mean free path,  $\mu$  is the chemical potential,  $p_F$  is the Fermi momentum,  $v_F$  is the Fermi velocity, l is the mean free path, and d is the film thickness.

Thus, the excess conductivity of the films at  $T < T_c$  is expressed in terms of the parameters of the superconductor as follows:

$$\sigma_{n}/\sigma' = A \exp\left\{-\frac{0.403m_{\text{eff}}^{2}v_{r}^{2}ld(1-T/T_{c})}{\hbar^{2}}\right\},$$

$$\gamma_{\text{theor}} = \frac{1}{\varepsilon_{c2}} = \frac{0.403m_{\text{eff}}^{2}v_{r}^{2}ld}{\hbar^{2}}.$$
(11)

The value obtained by Masker, Marcelja, and Parks<sup>9</sup> for  $\gamma_{\text{ther}}$  is twice as large.

When comparing the experiments with the theory, we confine ourselves to a comparison of the arguments of the exponential. The pre-exponential factor affects only the shift  $T_c$  due to the fluctuations effects, which in this case is beyond the scope of our analysis.<sup>6)</sup> The argument of the exponential determined the temperature width of the transition.

From a comparison of the empirical expression (4) with (11) it follows that the experimental temperature dependence of  $\sigma'$  agrees exactly with the theory. To explain the numerical agreement with the theory it is most convenient to equate the experimental value of  $\gamma$ to its theoretical value and determine from this the value of  $m_{eff}$ . In these calculations we use the values of d and l which we determined experimentally,<sup>7</sup> as well as the Fermi velocity  $v_F = 1.89 \times 10^7$  cm/sec obtained in experiments on critical fields of vanadium films<sup>8)</sup>.<sup>16</sup> The values of  $m_{eff}$  obtained for different samples by this method differ insignificantly from each other and lie in the interval  $(1.0-1.8)m_0$ . These values are close to the published data on  $m_{eff}$  for vanadium, determined from experiments on the de Haas-van Alphen effect and magnetothermal oscillations, where values from 1.7 to  $2.2m_0$  were obtained for different crystallographic directions.<sup>21</sup> Thus, we can conclude that the theory agrees not only with the qualitative behavior of the excess conductivity in the critical region, but describes also correctly the width of the transition.

Reference 9 contains not only the theory but also the authors' experimental data, obtained on granulated films of aluminum with different values of the normal resistance. By reducing formula (11) to the form

$$\frac{\sigma_n}{\sigma'} = A \exp\left\{-\frac{1-T/T_o}{1.04 \cdot 10^{-5} R_n^{-D}}\right\},\tag{12}$$

the authors investigated the connection between  $\gamma$  and the normal resistance per square  $R_n^{\Box}$  and obtained splendid agreement with the theory. Formulas (11) and (12) are identical in the case when the theory of the free electrons is valid. At the same time, when our data are compared with this last formula, quite significant numerical discrepancies are observed. The values of  $\gamma_{ther}$ , calculated by formula (12) using the measured values of  $R_n^{\Box}$ , exceed by almost two orders of magnitude the value of  $\gamma_{exp}$ . The cause of this disparity, in our opinion, is the following: on going from (11) to (12) we use the expression  $\rho_n = 3/2e^2N(0)v_Fl$ , which contains the density N(0) of the electronic states on the Fermi surface. For bulk vanadium, according to measurements of the electronic heat capacity,  $^{20}N(0) = 0.94 \times 10^{35}$  states/erg-atom. The effective mass corresponding to this state density is  $10.3m_0$ .<sup>20</sup>

Thus, measurements of the heat capacity and of the de Haas-van Alphen effect yield for vanadium essentially different effective masses. Calculating  $\gamma_{\text{ther}}$  from (12), we actually use a large effective mass (which enters in  $\gamma$  in the form  $m_{\text{eff}}^2$ ), and this is why we obtain such a large numerical discrepancy with experiment. These data indicate primarily that the theory of free electrons describes poorly the properties of transition metals. The fact that in our experiments we determined  $m_{\text{eff}} \approx (1-2)m_0$  indicates either that the resistivity in the region of the transition is determined by light carriers, or that the films are strongly disordered compared with bulky vanadium, on account of which the effective masses become smaller.

Summarizing the foregoing we can state that the experimental data obtained in the present study for the resistive temperature transition agree well both qualitatively and quantitatively with the predictions of the fluctuation theory. From the interpretation proposed here for the nature of the resistivity in the vicinity of the phase-transition point it follows, in particular, that in many cases the appreciable width of the superconducting transition can be explained without assuming structural inhomogeneities in the superconductors. From our point of view, the films used in the present investigations are most convenient for the observation of critical phenomena in the region of phase transitions. On the other hand, they are little influenced by structural inhomogeneities (static fluctuations), and on the other hand the conditions in these films are most favorable for the observation of effects of dynamic fluctuations of the order parameter, which are particularly substantial in the Ginzburg region ( $\varepsilon \leq Gi$ ). This conclusion can be drawn from the following considerations. According to the structural investigations,<sup>22</sup> the investigated vanadium films are made up of minute crystals, and the boundaries of the crystallites are strongly contaminated by diffusion of oxygen. This makes these objects closer to granulated films where, as is well known, the influence of the structural inhomogeneities is extremely small. On the other hand, because of the small thickness of the films (the twodimensional character of the system), the small mean free paths  $(l \sim 10^{-7} \text{ cm})$ , and the specifies of the transition metals, in which the values of  $\xi_0$  are smaller by a factor 5-6 than in simple metals,<sup>9)</sup> the estimate of the Ginzburg number is greatly increased. The width of the Ginzburg region for thin films is determined according to Ref. 23 by

$$Gi_z = (Gi_s)^{u_z} \xi(0)/d$$
 (13)

and for the investigated films it should be ~0.04 K. In the experiment, the width of the transition for the investigated samples is 0.12-0.2 K. It appears that this rather appreciable discrepancy should not be taken very seriously, inasmuch as the estimate of the Ginzburg number is accurate only in order of magnitude.<sup>24</sup> We note also that for one-dimensional superconductors, as indicated by Hohenberg,<sup>23</sup> the transition region observed in Ref. 10 is wider than Gi<sub>1</sub>.

Thus, for dirty films of transition metals the region of the smearing of the phase transition due to the critical fluctuations, turns out to be large. At the same time, the quasigranular character of the films decreases the role of the static fluctuations and consequently their contribution to the smearing of the transition, so that the critical phenomena can be observed in pure form.

We proceed now to the interpretation of the results of the investigations in a magnetic field. The excess conductivity  $\sigma'$  in magnetic fields near the critical region, where the interaction of the fluctuations is substantial, was calculated for  $H < H_c$  by Feat and Rickayzen.<sup>13</sup> For a magnetic field parallel to the film they obtained the expression<sup>10</sup>:

$$\left(\frac{\sigma_n}{\sigma'}\right)_{\parallel} = A \exp\left\{-\frac{\pi e^2 \hbar^2 d^3 (H_{c\parallel}^2 - H^2)}{3c^2 m_{eff}^2 k T_c \beta_{GL}}\right\},$$
(14)

where  $\beta_{\rm GL} = 1.02\hbar^2/nl^2m$ , *n* is the carrier density. In the transition region, the quantity  $\delta H = H_{\rm cll} - H$  is a small parameter. Expanding (14) in terms of this parameter and substituting in the resultant equation the expression for  $H_c$  from Ref. 25:

 $H_{e\parallel} = \sqrt{12} \Phi_0 / 2\pi \xi(\varepsilon) d,$ 

we obtain

$$\left(\frac{\sigma_n}{\sigma'}\right)_{\mu} = A \exp\left\{-\frac{1.608m_{\text{eff}}v_F^2 ld(1-H/H_{c_{\parallel}})(1-T/T_c)}{\hbar^2}\right\}.$$
 (15)

A comparison of formulas (8) and (15) shows that the experimentally observed temperature and field dependences of the excess conductivity in a parallel magnetic field agree with the theoretical ones. From a comparison of (15) and (11) it follows that the ratio of the coefficients  $\gamma_{\parallel}$  and  $\gamma$ , which determine respectively the width of transitions in  $H_{\parallel}$  and at H=0, is equal to 4, if we use the results of Kadanoff and Laramore,<sup>6</sup> or to 2 if we use the results of Ref. 9. In the experiments  $\gamma_{\parallel}/\gamma \approx 1.5$ , i.e., the order of magnitude of  $\gamma_{\parallel exp}$  is close to the theoretical value.

We have thus compared the theoretical result of Ref. 13 with the experimental data for  $H_{\rm w}$ . Unfortunately, the corresponding expression obtained in Ref. 13 for the case  $H_{\rm i}$  is not suitable for comparison with experiments, and we know of no other theoretical treatments of the field dependences.

We present below simple calculations which enable us to interpret the experimental results also for a perpendicular field. We use for this purpose the connection between the values of the order parameter  $|\Delta_{H1}|^2$ and  $|\Delta_{H1}|^2$  obtained in magnetic fields perpendicular and parallel to the film, and the mean value of the order parameter  $|\Delta_T|^2$  in the absence of a field. As is known from Refs. 26 and 27,

$$|\Delta_{T}|^{2} = \frac{8\pi^{2}k^{2}T_{o}^{2}(1-T/T_{o})}{7\zeta(3)},$$
(16)

$$|\Delta_{H\perp}|^{2} = \frac{ec\rho_{n}kT_{c}H_{c\perp}(1-H/H_{c\perp})}{\pi^{2}\beta_{A}(\kappa^{2}-0.5)},$$
(17)

$$|\Delta_{\pi_{\parallel}}|^{2} = \frac{ec\rho_{n}kT_{o}dH_{e\parallel}(1-H/H_{e\parallel})}{3^{\eta_{n}}\pi^{2}\xi(\varepsilon)\,\varkappa^{2}},$$
(18)

where  $\varkappa$  is the Ginzburg-Landau parameter and  $\xi(3) = 1.202$ . Using the expression for  $\varkappa$  from Ref. 28 ( $\gamma_c$  is the temperature coefficient of the electronic heat capacity)

$$\kappa = \frac{ec\rho_n \gamma_c^{\eta_n}}{k\pi^3} \left(\frac{21\zeta(3)}{2\pi}\right)^{\eta_2}$$
(19)

and putting<sup>11</sup>)  $\kappa^2 \gg 0.5$ , we obtain after simple transformations

$$|\Delta_{H\perp}|^2 = \beta_A^{-1} |\Delta_T|^2 (1 - H/H_{c\perp}), \qquad (20)$$

$$|\Delta_{H_{\parallel}}|^{2} = 2 |\Delta_{T}|^{2} (1 - H/H_{c_{\parallel}}).$$
(21)

Thus, the order parameter in a magnetic field is expressed only in terms of an order parameter that depends only on the temperature and in terms of the value of the reduced field.

By virtue of the general character of the similarity laws one should hope to be able to take into account the influence of the magnetic field on the excess conductivity by introducing in (11) a suitable field dependence of the order parameter. We then obtain for transitions in a perpendicular field

$$(\sigma_{\rm n}/\sigma')_{\rm I} = A \exp\{-[0.403m_{\rm eff}^2 v_{\rm F}^2 ld(1-H/H_{\rm c\perp})(1-T/T_{\rm c})]/\beta_{\rm A}\hbar^2\}; \quad (22)$$

and for transitions in a parallel field

$$(\sigma_n/\sigma')_{\parallel} = A \exp\{-[0.806m_{eff}^3 v_F^2 l d(1 - H/H_{c\parallel})(1 - T/T_c)]/\hbar^2\}.$$
(23)

It is easily seen that the expression (23) obtained by us agrees, apart from a numerical factor, with the more rigorously derived formula (15). If we start from the results of Masker, Marcelja, and Parks,<sup>9</sup> then we obtain for the parallel field exact agreement between the numerical coefficient and formula (15). This gives grounds for assuming that formula (22) for the perpendicular field is also reliable.

It is seen that the analytic expression (22) reflects the empirical similarity law (6) obtained above. As follows from (22) and (11), the ratio  $\gamma_{\rm L}/\gamma$  should equal



FIG. 7. Schematic representation of the fluctuation region in the three-dimensional space T, H, and I. Solid lines—lines of phase-transition points. The dashed lines delimit the fluctuation region.

to  $1/\beta_A(\beta_A = 1.16)$ . The experimental values of  $\gamma_L/\gamma$ differ from the indicated quantity by  $\pm(10-40)$ %. Such an agreement should be regarded as perfectly satisfactory.

It follows from the foregoing analysis that the resistive phenomena in the region of the superconducting transitions in the investigated films are due to fluctuation changes of the order parameter. The critical region in which the fluctuations are significant can be represented with the aid of a three-dimensional diagram in the space of T, H, and I (Fig. 7). The phenomena investigated by us are localized in the immediate vicinity of the three-dimensional surface of the superconducting phase transition, which is shown in Fig. 7. Outside this region, in a perpendicular magnetic field, when a mixed state exists, the resistivity is determined to a considerable degree by the dynamics of the variation of the order parameter when the vortices move,<sup>29,30</sup> while inside the region they are determined by the fluctuation change of the order parameter.

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- <sup>1)</sup>A similar situation occurs also in zero-dimensional superconductors (small particles with dimensions smaller than the coherence length  $\xi_0$ ). This question was first investigated theoretically by Shmidt.<sup>5</sup>
- <sup>2)</sup>Laws similar to those used described below for the lower exponentials are observed also for the upper exponentials.
- <sup>3)</sup>All the empirical relations obtained in the present paper were reduced in the same manner. The values of  $\delta$  and  $\delta_{res}^2$  at measuring currents  $I \ge 100 \ \mu A$  do not exceed in any case the values indicated in the text.
- $^{\rm 4)} {\rm For\ example}\,,\, {\rm a\ film\ 370}$  Å thick, the results for which were given above, goes over according to our measurements into a vortical state at T = 2.49 K. The temperature dependence of  $H_{cl}$  is changed in this case from  $(1 - T/T_c)^{1/2}$  to  $1 - T/T_c$ . All the data considered above pertain to sufficiently high temperatures.
- <sup>5</sup> In later papers they use customarily the more compact expression  $A_{\infty} = e^2 R_n^{-1} / 16\hbar$ , which is obtained from (10) by using the formulas of the free-electron theory.
- <sup>6</sup>) The empirical expression (4) does not contain at all a preexponential factor, inasmuch as it contains the experimentally measured value of  $T_c$  and not the quantity  $T_{c0}$  which would be obtained in the absence of fluctuations.
- <sup>7</sup>) The length *l* is determined from the relation  $\xi^2(\varepsilon) = 0.72\xi_0 l \varepsilon^{-1}$ , and  $\xi^2$  (E) is obtained from the measured  $H_{cL}$  (E) dependence. In the estimate of  $\xi_0$  we took into account the change of  $T_c$  of the films compared with the bulk vanadium, and for the latter we used the value  $\xi_0 = 450$  Å.<sup>20</sup>
- <sup>8)</sup>We note that this value is very close to  $v_F = 1.77 \times 10^7$  cm/sec for bulk vanadium.<sup>20</sup>
- <sup>9</sup>)For three-dimensional systems<sup>1</sup> Gi<sub>3</sub> ~  $(a_0/\xi_0)^4$   $(a_0$  is the lattice parameter of the metal), so that for transition metals it is approximately 10<sup>3</sup> times larger than for such metals as tin.
- <sup>10</sup>)Bearing in mind temperatures close to the transition point,

we replace T in (14) by  $T_c$ .

- We replace I in (127 by -c. <sup>11</sup>)For the samples investigated by us  $\varkappa$  ranges approximately from 8 to 10.
- <sup>1</sup>V. L. Ginzburg, Fiz. Tverd. Tela (Leningrad) 2, 2031 (1960) [Sov. Phys. Solid State 2, 1824 (1961)].
- <sup>2</sup>A. Z. Patashinskii and V. L. Pokrovskii, Fluktuatsionnaya teoriya fazovykh perekhodov (Fluctuation Theory of Phase Transitions), Nauka, 1975.
- <sup>3</sup>L. G. Aslamozov and A. I. Larkin, Fiz. Tverd. Tela (Leningrad) 10, 1104 (1968) [Sov. Phys. Solid State 10, 875 (1968)].
- <sup>4</sup>T. M. Rice, Phys. Rev. **140A**, 1889 (1965).
- <sup>5</sup>V. V. Shmidt, Pis'ma v ZhETF 3, 141 (1966) [JETP Lett. 3, 89 (1966)].
- <sup>6</sup>L. P. Kadanoff and G. Laramore, Phys. Rev. 175, 579 (1968). <sup>7</sup>J. S. Langer and V. Ambegaokar, Phys. Rev. 164, 498
- (1967). <sup>8</sup>D. F. McCumber and B. I. Halperin, Phys. Rev. 1B, 1054,
- (1970).
- <sup>9</sup>W. E. Masker, S. Marcelja, and R. D. Parks, Phys. Rev. 188, 745 (1969).
- <sup>10</sup>R. J. Warburton, B. R. Patton, W. W. Webb, and J. W. Wilkins, Physica 55, 3211 (1971).
- <sup>11</sup>W. W. Webb and R. J. Warburton, Phys. Rev. Lett. 20, 461 (1968)
- <sup>12</sup>V. N. Bogomolov, Yu. A. Kumzerov, D. D. Prokof'ev, A. P. Smirnov, and S. I. Tsypkin, Fiz. Tverd. Tela (Leningrad) 18, 971 (1976) [Sov. Phys. Solid State 18, 556 (1976)].
- <sup>13</sup>G. R. Feat and G. Rickayzer, J. of Physics, ser F., 5, 307 (1975).
- <sup>14</sup>A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Teor. Fiz. 65, 1704 (1973) [Sov. Phys. JETP 38, 854 (1974)].
- <sup>15</sup>N. Ya. Fogel and A. S. Sidorenko, Phys. Lett. **68A**, 456 (1978).
- <sup>16</sup>A. A. Teplov, M. N. Mikheeva, V. M. Golyanov, and A. N. Gusev, Zh. Eksp. Teor. Fiz. 71, 1122 (1976) [Sov. Phys. JETP 44, 587 (1976)].
- <sup>17</sup>N. E. Alekseevskii, V. M. Zakozarenko, K. Blüthner, and H. J. Köhler, Phys. Stat. Sol. 34A, 541 (1976).
- <sup>18</sup>A. A. Abrikosov, Zh. Eksp. Teor. Fiz. **47**, 720 (1964) [Sov. Phys. JETP 20, 480 (1965)].
- <sup>19</sup>I. O. Kulik, *ibid.* 52, 1632 (1967) [25, 1085 (1967)].
- <sup>20</sup>R. Radebaugh and P. H. Keesom, Phys. Rev. 149, 217 (1966).
- <sup>21</sup>R. D. Parker and M. H. Halloran, Phys. Rev. **9B**, 4130 (1974).
- <sup>22</sup>A. S. Sidorenko, L. F. Rybal'chenko, and N. Ya. Fogel', Proc. 20th All-Union Conf. on Low Temp. Phys., part III, 1979, p. 140.
- <sup>23</sup>P. Hohenberg, Paper at 11th Internat. Conf. on Low Temp. Physics, Scotland, August, 1968.
- <sup>24</sup>L. P. Kadanoff, W. Götze, D. Hamblen, R. Hecht, E. A. S. Levis, V. V. Palciauskas, M. Rayl, and J. Swift, Rev. Mod. Phys. 39, 395 (1967).
- <sup>25</sup>V. L. Ginzburg and L. D. Landau, Zh. Eksp. Teor. Fiz. 20, 1064 (1950).
- <sup>26</sup>L. P. Gor'kov, Zh. Eksp. Teor. Fiz. **36**, 1918 (1959) [Sov. Phys. JETP 9, 1364 (1959)].
- <sup>27</sup>E. Guyon, Advan. Phys. 15, 417 (1966).
- <sup>28</sup>L. P. Gor'kov, Zh. Eksp. Teor. Fiz. 37, 1407 (1959).
- <sup>29</sup>M. Tinkham, Phys. Rev. Lett. 13, 804, 1969.
- <sup>30</sup>N. Ya. Fogel', Zh. Eksp. Teor. Fiz. 63, 1371 (1972) [Sov. Phys. JETP 36, 725 (1973)].

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