N-shaped current-voltage characteristics and the temperature-electric instability in tin at low temperatures

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The current-voltage characteristics (CVC) of pure tin ($\rho(300\text{ K})/\rho(4.2\text{ K})\approx 10^4$) are measured at temperatures between 4.2 and 30 K. A simple contactless technique is suggested for the measurement of the characteristics. At current densities $j\approx 5\times 10^4$ A/cm^2 and at a heat dissipation from the sample $\sim 10^{-1}$ W/cm^2, a descending branch is observed on the CVC, and can be ascribed to the fact that the resistivity depends more strongly on temperature than the heat dissipation from the sample surface. The stability of the sample state on this branch of the CVC with respect to homogeneous (coordinate-dependent) or inhomogeneous perturbations of the crystal-lattice temperature is analyzed.

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1. INTRODUCTION

According to theoretical estimates[1] there can be realized in metals at low temperatures and at a current density ~10^4–10^5 A/cm^2 conditions such that the homogeneous distribution of the electric field becomes unstable, and this leads to the spontaneous onset of spatially separated regions with different electric field intensities—domains.

Domain electric instability was observed about 20 years ago in semiconductors and by now had found extensive use not only in research but also in practical applications (the Gunn effect). The observation of such an instability in crystals would therefore be the next important step towards the investigation of the onset and development of electric domains in a solid with a dense electron plasma, and their possible technical utilization.

It is known[2] that one of the necessary conditions for the onset of a domain structure is the presence of a current-voltage-characteristic section with negative differential conductivity (N-type CVC). The appearance of such a section on the CVC of a metal can be expected under conditions of coherent magnetic breakdown,[3] as well as when the sample is heated by the current to a temperature at which its resistance $R(T)$ has a stronger temperature dependence than the heat flux $P(T)$ carried away from its surface—the so-called "temperature" mechanism of formation of an N-type CVC (Ref. 1).

The first experimental confirmation of the existence of a decreasing branch of the CVC of metals, due to the "temperature" mechanism, was obtained for gallium in Ref. 4. However, no temperature-electric instability of the states of the investigated sample was observed in these experiments. On the basis of the results of Ref. 1, it can be assumed that the stability of the CVC of gallium[4] on the section of the characteristic with the negative differential conductivity is due to the following causes. First, the procedure chosen in Ref. 4 to measure the CVC of metals did not provide for a uniform dissipation of the Joule heat from the different sections of the investigated sample. Second, owing to the limited length of the sample ($l\approx 2$ cm), the condition for its stable "domain" state was not realized. It was therefore of interest to carry out a detailed investigation of the CVC of metals under conditions of electric heating at low temperatures, with allowance for the arguments advanced above.

The experiments referred to in the present paper were performed on tin samples of length exceeding 10 cm using a procedure that ensured uniform removal of the Joule heat from the entire surface of the investigated sample.

2. BASIC PREMISES

The criterion for the onset of a CVC section with negative differential conductivity $\sigma_d=dI/dU$ of the sample (I and U are respectively the current and the voltage) under conditions of temperature heating is satisfaction of the inequality[2]

$$\sigma_d=R(T,f)\,\frac{(d/dT)\,[P(T)/R(T,f)]}{(d/dT)\,[P(T)R(T,f)]}<0, \tag{1}$$

which follows from the energy conservation law and from Ohm's law. Here $P(T)$ is the heat flux from the sample to the ambient at a given temperature T and under given heat-transfer conditions; $R(T,f)$ is the resistance of the sample to current of frequency f at a temperature T.

In many cases the $P(T)$ and $R(T,f)$ dependences of a pure metal at a fixed frequency f can be approximated by power-law functions. We put

$$R(T)=\alpha_0(T/T_0)^m, \tag{2a}$$

$$P(T)=\beta\left(\frac{T}{T_0}\right)^s\frac{T-T_0}{T_0}, \tag{2b}$$

where α_0 is the sample resistance at an ambient temperature T_0, $\beta(T/T_0)^s$ characterizes the heat-transfer coefficient and its dependence on the temperature, $(T-T_0)/T_0$ is the relative temperature rise, and the parameters m and s are determined by the dominant energy-scattering mechanism.

With allowance for (2a) and (2b), it follows from (1) that formation of an N-type CVC calls for

$$m-1>s\geqslant -m-1, \tag{3}$$

with $\sigma_d=0$ at a temperature $T_N=(m-s)T_0/(m-s-1)$.

We introduce the dimensionless parameter $t=T/T_0$. We then obtain from the energy conservation law, with

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allowance for Ohm's law and expressions (2a) and (2b), if the inequality (3) holds,

$$\frac{I(t)}{I_c} = \left(\frac{t}{t_c}\right)^{(s-m)/2} \left(\frac{t-1}{t_c-1}\right)^{1/2}, \tag{4a}$$

$$\frac{U(t)}{U_c} = \left(\frac{t}{t_c}\right)^{(s+m)/2} \left(\frac{t-1}{t_c-1}\right)^{1/2}. \tag{4b}$$

Here I_c and U_c are the critical current and voltage corresponding to the temperature T_N, and $t_c = T_N/T_0$.

Analyzing expressions (4a) and (4b), it is easily seen that the voltage is a monotonically increasing function of temperature, and the current reaches a maximum at $t = t_c$. Eliminating t from these equations, we can obtain the explicit form of the current-voltage characteristic of the sample. This characteristic will have a region of values of U where the differential conductivity reverses sign and becomes negative.

3. SAMPLES AND EXPERIMENTAL TECHNIQUE

Starting from the foregoing, we determine now the basic requirements that must be satisfied by the samples and by the experimental technique to observe N-type CVC in metals.

In the most general case, with account taken of the different heat-transfer mechanisms (cooling by a liquid or gas, heat conduction via materials in thermal contact with the sample, radiation) the value of s can reach ~3. To satisfy the inequality (3), the temperature dependence of the resistivity $\rho(T)$ of the investigated sample must be described by a power-law function with an exponent $m > 4$.

The samples used in the experiments must therefore be of sufficiently high purity [the ratio of the resistivities at room and helium temperatures must be $\rho(300\ \text{K})/\rho(4.2\ \text{K}) \approx 10^2-10^3$], and the measurements must be carried out at $T \ll \Theta$ (Θ is the Debye temperature), where such a $\rho(T)$ dependence is easily obtained. For most metals these are temperatures close to that of liquid helium. At these temperatures, the most convenient mechanism of heat transfer from the sample, which permits investigation of the CVC of metals under conditions of electric heating at relatively low current densities, is radiation. However, owing to the small slope of the descending section of the CVC ($m - s \lesssim 2$) and to the difficulty of ensuring uniform heat exchange between the investigated sample and the container that encloses it, such an experimental setup offers from our point of view little promise for the solution of the problem of observing a domain structure in metals.

In the present study we have chosen a mechanism of convective heat removal from the sample in an atmosphere of helium gas. Since the heat dissipation at a temperature ~10 K is under these conditions $10^{-1}-10^{-2}$ W/cm^2, heating a sample with $\rho \sim 10^{-9}-10^{-10}\ \Omega\cdot\text{cm}$ calls for current densities 10^4-10^5 A/cm^2. To reach such current densities we used an iron-core transformer, so as to decrease the loss of magnetic flux contained in the area surrounded by the metallic ring.

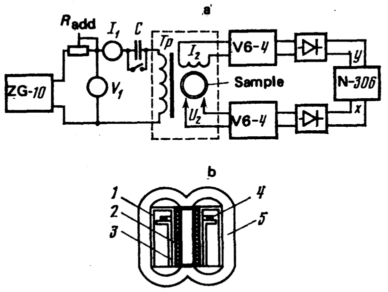

FIG. 1. a) Block diagram of experimental setup for the measurement of the CVC; b) measuring transformer: 1) inverted Teflon cup, 2) primary winding of transformer, 3) Teflon sample holder, 4) sample, 5) magnetic core of transformer.

The investigated samples were made of OVCh-000 tin in a graphite crucible in the shape of long cylindrical tube, using zone crystallization in an inert-gas atmosphere. The tin was cut next into rings of diameter ~40 mm and cross section ~1 mm^2, with a resistivity ratio $\rho(300\ \text{K})/\rho(4.2\ \text{K}) \approx 10^4$. The structure of the samples was not monitored.

The ring was placed in an inverted Teflon cup and placed over the primary winding of a transformer located in liquid helium [Fig. 1(b)]. The sample was the secondary winding of the transformer and, being located in a helium-gas medium, was cooled by convection in the gas. The primary winding was thermally stabilized by the liquid helium.

The power source was a ZG-10 generator [Fig. 1(a)]. The electric field strength in the ring and the current were determined from the voltage drop and from the value of the current in the primary circuit of the transformer, which were monitored with VK 7-10A and Ts-4311 instruments.

The feasibility of determining the current-voltage characteristics of the sample from measurements in the primary circuit can be easily verified by considering the equivalent circuit of Fig. 2(a). The elements r_1, L_1, and L_2' of this circuit take into account the losses in the primary winding of the transformer and the stray fluxes in the primary and secondary windings, respectively. The coil L_0 through which the magnetizing current $I_0 = I_1 + I_2'$ flows, determines the effective magnetic flux of the transformer, and r_0 determines the core losses to hysteresis and to eddy currents. The equivalent sample resistance is $R_T' \approx n^2 R_T$, where $R_T = R_0 + R(T)$ are the temperature-dependent and temperature-independent parts of the sample resistance; n is the transformation ratio; R_{in} is the internal resistance of the generator; R_{add} is the additional resistance. In a real transformer $L_0 \gg L_1$ ($L_1 \approx L_2'$), $r_0 \gg r_1$, and $r_0 \gg R_T'$, so that the equivalent circuit can be represented

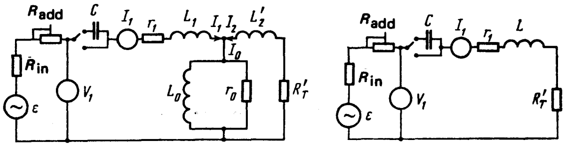

FIG. 2. Equivalent circuit of the setup.

with sufficient accuracy in the form shown in Fig. 2(b). The experimentally measured current I_1 and voltage V_1 in the primary circuit are given by

$$I_1=(P(T)/R_T')^{1/2}, \tag{5a}$$

$$V_1=I_1[(r_1+R_T')^2+4\pi^2f^2L^2]^{1/2}, \tag{5b}$$

which describe a current-voltage characteristic of an electric circuit that contains not only a nonlinear element R'_T, but also an active resistance r_1 and an inductance L.

Since the primary winding of the transformer was placed directly in the liquid helium and was made of copper wire with a resistivity ratio $\rho(300\ \text{K})/\rho(4.2\ \text{K}) \approx 100$, the value of r_1 did not exceed 0.1 Ω in our experiments and its contribution compared with R'_T (~30 Ω) could be neglected. In individual experiments, the current and voltage in the ring were measured directly. In this case a previously calibrated Rogowski loop placed around the sample measured the current, and copper leads of 0.1 mm diameter were soldered with indium at two asymmetrical points of the ring. The voltages picked off the ring and the Rogowski loop were measured with V6-4 selective voltmeter and were recorded, following linear detection, with an H-306 x-y recorder [Fig. 1(a)]. It was established by experiment that the connection between the current (voltage) measured in the secondary circuit by the method described above, and the current (voltage) in the primary circuit, remained linear in the entire range of the measurements. The heat transfer via the potential leads did not influence significantly the shape of the CVC, since it was smaller by three orders of magnitude than the heat transfer through helium gas under convection conditions.

These measurements, however, did not exclude the influence of the inductance on the measured voltage drop, thus leading to a distortion of the CVC. Therefore, to reveal the true form of the current-voltage characteristic of the investigated sample, the measurements were performed under conditions of resonance of the voltages in the primary circuit. In this case the primary winding of the transformer was fed through a capacitor C and at the resonant frequency the resistance of the measured circuit became purely active, determined mainly by the sample resistance R'_T.

4. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Investigations of the current-voltage characteristics of tin under conditions of low-temperature electric heating were carried out in the frequency interval 66–5200 Hz and in the temperature range 4.2–30 K. The shapes of these characteristics, obtained at pump-current frequencies 191, 416, and 722 Hz are shown in Fig. 3.

As seen from the figure, these characteristics have a complicated nonmonotonic character. First, with increasing voltage, the current in the measuring circuit increases linearly and then, after reaching a critical value I_{1c}, which causes the sample to be heated to a temperature 6–7 K, decreases to a certain value that is practically independent of frequency and varies very little with increasing voltage.

In the entire investigated frequency interval, the critical current was well described by a linear function of $f^{-1/4}$, in agreement with the shape that follows from (5a) under conditions of the normal skin effect, for which $R'_T \sim f^{1/2}$.

Calculation of the absolute value of the critical current density and of the average field intensity in the sample, which was carried out for a frequency 722 Hz, yielded $j_c = 5\times 10^4$ A/cm² and $E_c \approx 5\times 10^{-7}$ CGS.

The independence of the CVC of tin of frequency at $V_1 > 10$ V (see Fig. 3) can be easily attributed to the size effect. Under these conditions, the sample is heated to temperatures ~15 K, at which its resistivity increases to such an extent that the penetration depth (δ_n) of the alternating current exceeds the thickness of the sample. Indeed, at this temperature and at a frequency 722 Hz we have $\delta_n \approx 6\times 10^{-2}$ cm for tin of the indicated purity, whereas the thickness of the samples investigated in the experiments was $\sim 1\times 10^{-1}$ cm.

The region with $V_1 > 10$ V (see Fig. 3) can be regarded as the region where the current is stabilized, since the current changes only 10% when the voltage is changed by more than 1.5 times. By varying the sample dimensions, its initial temperature, and the heat-exchange conditions it is easy to control both the absolute value of the stabilization current and the width of this section of the CVC; this may be of definite interest from the practical point of view.

A typical feature of the obtained characteristics is the presence of a decreasing section, on which the differential conductivity σ_d is negative. The appearance of this branch on the current-voltage characteristic of tin is undoubtedly due to the stronger temperature dependence of the sample resistance compared with the temperature dependence of the heat flux $P(T)$ from its surface.

At high frequencies, the CVC branch with $\sigma_d < 0$ is stable, but instability was observed with decreasing frequency. In this case, a small increment of the generator voltage transferred the sample spontaneously

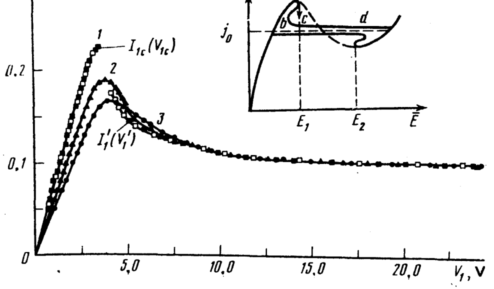

FIG. 3. Current-voltage characteristics of tin at various pump frequencies: 1) 191 Hz, 2) 416 Hz, 3) 722 Hz. Values measured in the forward direction are marked by black symbols. The inset shows a typical form of a CVC of a semiconductor with an electric-field domain.[5]

from the state $I_{1c}(V_{1c})$ into the state $I_1'(V_1')$, and hysteresis was observed in the reverse direction along the CVC (see curve 1 of Fig. 3).

The appearance of this instability and of the hysteresis might seem to be evidence in favor of the formation of a domain structure in tin, but a more detailed examination readily explains this behavior as being due to the influence of the external circuit. In fact, assume that a homogeneous (independent of the coordinate) temperature fluctuation δT is produced in the sample and changes the rate of change of the crystal-lattice energy by an amount $\delta[I_1V_1-P(T)]$. Then, recognizing that under resonance conditions we have

$$I_1=\varepsilon/(R_T'+Z), \quad V_1=\varepsilon R_T'/(R_T'+Z) \quad (r_1\ll R_T'),$$

where ε is the effective value of the generator voltage and $Z=r_1+R_{\mathrm{in}}+R_{\mathrm{add}}$ [see Fig. 2(b)], we can write

$$\begin{aligned}\delta[I_1V_1-P(T)]&=\left[I_1^2\frac{dR_T'}{dT}(Z-R_T')(Z+R_T')^{-1}-\frac{dP(T)}{dT}\right]\delta T\\&=-R_T'Z\left(\frac{dP(T)}{dT}+\frac{dR_T'}{dT}I_1^2\right)(Z+R_T')^{-1}\\&\times\left[R_T'\frac{(d/dT)(P(T)/R_T')}{(d/dT)(P(T)R_T')}+\frac{1}{Z}\right]\delta T.\end{aligned} \tag{6}$$

The first term in the square brackets is, by definition, σ_d [see Eq. (1)], so that with allowance for $dR'_T/dT>0$ it follows from (6) that

$$\delta[I_1V_1-P(T)]\sim-(\sigma_d+1/Z)\delta T.$$

As seen from this expression, the homogeneous fluctuation of the temperature of the sample on the section with $\sigma_d<0$ will be damped only if the following inequality is satisfied

$$|\sigma_d|<1/Z. \tag{7}$$

The validity of this conclusion is clearly illustrated in Fig. 4, which shows the current-voltage characteristics of tin for different values of an additional resistance R_{add} connected in the measuring circuit. With increasing R_{add}, the inequality (7) is violated and the CVC acquires a broad region in which the sample state is unstable. This inequality is not satisfied at low frequencies because of the large σ_d.

From the point of view of finding the region of the existence of a domain structure in metals, principal interest attaches therefore to the characteristics investigated in a given-voltage regime ($Z\ll R'_T$), for which the sample is stable to homogeneous fluctuations even at $\sigma_d<0$. In this case it becomes possible to observe the evolution of fluctuations that are inhomogeneous along the current and do not change the total resistance of the sample, and lead to formation of electric-field domains.[1,5]

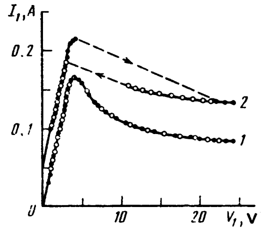

FIG. 4. CVC of tin at a frequency $f=416$ Hz: 1) $R_{\mathrm{add}}=0$; 2) $R_{\mathrm{add}}=100\,\Omega$. Curve 2 is shifted upward 0.05 A. The black and white circles correspond to passage in the forward and backward directions, the dashed lines designate the instability regions.

Under conditions close to the given-voltage regime, we plotted the characteristics shown in Fig. 3 (curves 2 and 3). Comparing them with the CVC of a semiconductor with a domain (see the inset of Fig. 3), it is easily noted that there are no singularities that would attest to domain formation in the tin. In all probability this can be attributed to the insufficient slope of the CVC in the region with $\sigma_d<0$, and this leads, at the given sample dimensions and at the thermal conductivity of the metal, to a stable homogeneous distribution of the electric field E. According to Ref. 1, for the "temperature" mechanism there exist three ranges of the parameters $\bar{E}$ and $z=(2\pi l_0)^2/l^2$, with qualitatively different structure of the electric field [$l_0\approx(\varkappa Td/q)^{1/2}$, $\varkappa$ is the thermal conductivity of the metal, and d and l are the thickness and length of the sample]. The boundary of the region with stable homogeneous distribution of E is determined by the equation $\sigma_1 z=\max|dj/dE|$, where $\sigma_1=j_1/E_1$ is the conductivity at the point a (see the inset of Fig. 3). Using these equations, it can be shown that for a sample with an electric field domain to be stable it is necessary to satisfy the inequality

$$\frac{l}{d^{1/2}}\geqslant\frac{\pi}{c_1}\left(\frac{\varkappa T_N}{c_2}\right)^{1/2}. \tag{8}$$

The constants c_1 and c_2 are determined from experiment with the aid of the expressions

$$V_{1c}=2c_1l/d^{1/2} \text{ and } dI_1/dV_1=c_2d^2/l.$$

Assuming $\varkappa\approx20$ W · cm^{-1} · deg^{-1} (Ref. 6), $T_N=7$ K, and $l=12$ cm, we find that at pump frequency 416 Hz and at $d\approx10^{-1}$ cm the sample length needed to satisfy the inequality (8) must be larger than 36 cm. In the case of shorter samples, however, a stable state should be realized with uniform field distribution, as is apparently indeed observed in the present experiments.

Even though the result is by way of estimate, an unambiguous answer to the question of existence of electric-field domains in metals calls for research on longer and thinner samples, and also with direct current. At the present time such experiments are difficult to perform for the lack of sufficiently pure and thin samples (<0.1 mm diameter and length $l>10$ cm). Nevertheless, they will apparently become feasible in the nearest future.

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[1] A. A. Slutskin and A. M. Kadigrobov, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 219 (1978) [JETP Lett. **28**, 201 (1978)].
[2] V. L. Bonch-Bruevich, I. P. Zvyagin, and A. G. Mironov, Domennaya élektricheskaya neustoĭchivost' v poluprovod-

nikakh (Domain Electric Instability in Semiconductors), Nauka, 1972, Chap. 1.
[3] A. A. Slutskin and A. M. Kadigrobov, Fiz. Nizk. Temp. **4**, 536 (1978) [Sov. J. Low Temp. Fiz. **4**, 262 (1978)].
[4] Yu. N. Chiang and A. G. Ordynets, *ibid.* **6**, 1279 (1980) [**6**, 622 (1980)].
[5] A. F. Volkov and Sh. M. Kogan, Usp. Fiz. Nauk **96**, 633 (1968) [Sov. Phys. Usp. **11**, 881 (1969)].
[6] M. P. Malkov, I. B. Danilov, A. G. Zel'dovich, and A. B. Fradkov, Spravochnik po fiziko-tekhnicheskim osnovam kriogeniki (Handbook of Physical and Technical Fundamentals of Cryogenics), Energiya, 1978, p. 131.

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