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Oscillations of the critical temperature and resistance in lamellar thin-film vanadium-carbon structures

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The superconducting properties of vanadium-carbon sandwiches with vanadium layers $d_V = 100$ Å thick and variable thickness carbon liners are studied. The thickness of the carbon liners was varied between 0 and 100 Å. The investigations are performed with samples having one, two, and five vanadium layers. The dependence of the sandwich superconducting temperature T_c on the carbon liner thickness is oscillatory, the oscillations being particularly pronounced for the samples with two layers. The first peak on the $T(d_C)$ curve (d_C is the thickness of the carbon coating) occurs at $d_C = 3$ Å for single-layer samples (vanadium film with carbon coating of variable thickness). In two- and five-layer sandwiches the first peak is observed for a carbon layer thickness of 5 Å. A correlation is obtained between the critical temperature and the quantities R_{300}/R_n, ρ_{300}, and ρ_n for multilayer samples.

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We have previously reported[1,2] observation of an oscillatory dependence of the critical temperature of films of a number of superconducting metals on the thickness of a diamond coating. The oscillations took place at very small carbon coatings ($d_C < 20$ Å) and for sufficiently thin ($d < 300$ Å) metallic layers. Quite recently, Sixl[3] reported observation of a similar behavior of the critical temperature of thin aluminum films as a function of the thickness of a silicon-oxide coating. Although the aluminum films were obtained by another method and the coating was of a different material, the general laws governing the $T_c(d_C)$ dependence were the same as in our experiments. We have continued the investigations on vanadium films coated with carbon, and also studied the effect of a thin dielectric layer (C) of variable thickness on the properties of metallic vanadium film in layered sandwich-type systems.

In the method used to produce the samples, the sandwich structure has a number of advantages over a superconductor coated with a dielectric of variable thickness. Thus, in the case of a sandwich the sample can be covered from above a thick protective dielectric film, and this excludes completely the possibility of oxidation of the investigated metallic layer. In addition, in the layered sample the thin dielectric layer borders upon two identical metallic layers, so that the action of the dielectric on the superconductor becomes more effective. All this gives grounds for hoping to obtain, in the case of a sandwich, a more distinct oscillatory dependence of the properties of the metallic layer on the thickness of the dielectric.

PREPARATION OF SAMPLES AND MEASUREMENT PROCEDURE

The sample were prepared in a high-vacuum ion-evaporation installation described by us in[2]. The evaporation was carried out in spectrally pure krypton, the working pressure of which was 5×10^{-6} Torr. The discharge current was 1 mA at 4 kV in a magnetic field 0.7 kOe. During the sputtering time all the parts of the working chamber and the substrate had a temperature close to that of liquid nitrogen.

The film thickness was calculated from the evaporation rate and evaporation time. The evaporation rate of each material was determined with the aid of control samples 1000 Å thick. The thicknesses of these samples were determined by an interference method. The error in the determination of the investigated films did not exceed 5% in the entire range of thicknesses. The film growth rates were 10 Å/min for vanadium and 2 Å/min for carbon films$^{1)}$. The determination of the growth rate was repeated after several evaporation experiments.

The raw materials for the cathode were metallic vanadium and graphite of purity not worse than 99.94%. The investigated samples were rectangular in shape and measured 5×9 mm. The substrates were polished glass plates into which platinum-wire leads were sealed. To exclude edge effects, the edges of the films were trimmed with a diamond microcutter. The superconducting-transition temperature and the sample resistance were measured both before and after trimming the edges.

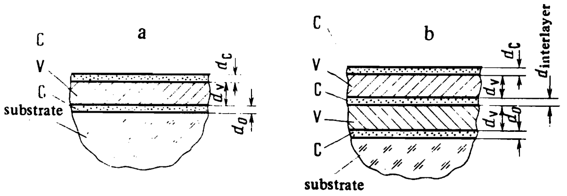

FIG. 1. a) Vanadium film with fixed carbon sublayer and coating of variable-thickness (section); b) sandwich with two layers of vanadium and fixed carbon sublayer and coating (section).

Figure 1 shows schematically the two investigated sample types: a–metallic film with dielectric coating, and b–two-layer sandwich. To prevent the sample properties from being affected by the possible imperfections and contaminations of the substrate prior to evaporation of the sample, a fresh layer of carbon (sublayer) of thickness $d_0 = 15$ Å was deposited on the sample. All the samples were coated after sputtering by a protective film of synthetic diamond, with thickness likewise 15 Å.[2)] The protective sublayers and coatings were deposited in the same ultra-high-vacuum evaporation installations used for the investigated samples. It should be noted that the carbon films obtained by this method remain solid and strong, and form no island structure when their thickness is decreased down to 3 Å.

The construction of the installation made it possible to obtain eight pairs of samples during in one evaporation cycle. The samples making up the pair were prepared simultaneously, and the difference between their properties was determined only by random factors. After the end of the evaporation, the installation was heated to room temperature, unsealed, and the samples transferred to the measuring helium cryostat.

The critical temperature was measured by a resistive method.[3)] The superconducting transition temperature was defined as the temperature at which $R = 0.5R_n$, where R_n is the residual resistance of the sample. The temperature of the measuring block of the cryostat was measured with a Scientific Instruments Co. germanium thermometer, and the sample resistance was measured by a potentiometer method. To calculate the resistivity of the samples, the geometric dimensions of the samples were determined with an optical comparator. The error of ρ did not exceed 7%.

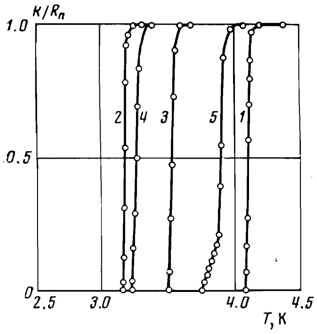

FIG. 2. Superconducting-transition curves of vanadium films with carbon coating and of two-layer vanadium-carbon sandwiches with carbon interlayers of varying thickness: 1) $C_{15}-V_{200}-C_{15}$, 2) $C_{15}-V_{100}-C_8-V_{100}-C_{15}$, 3) $C_{15}-V_{100}-C_{10}-V_{100}-C_{15}$, 4) $C_{15}-V_{100}-C_{11}-V_{100}-C_{15}$, 5) $C_{15}-V_{120}-C_{15}$ (the subscript denotes the interlayer thickness in angstroms).

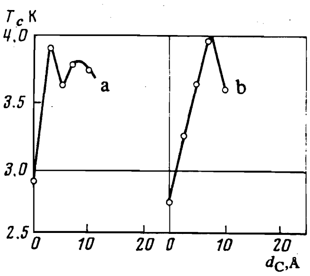

FIG. 3. Critical temperature of vanadium films with $d_V = 120$ Å and fixed carbon sublayer, as a function of the thickness of the carbon coating: a) vanadium films with carbon sublayer $d_0 = 15$ Å, b) vanadium films with carbon sublayer $d_0 = 10$ Å.

RESULTS

Figure 2 shows plots of the transition of pure vanadium films in two-layer V–C–V sandwiches with different carbon interlayer thickness. The width of the superconducting transition changed little from sample to sample and amounted to only several hundredths of a degree K. It should be noted that the vanadium films investigated by us, with thickness $d \geq 500$ Å, had a critical temperature close to T_c of the pure bulky metal and a sufficiently large resistance ratio, $R_{300}/R_n = 6$.

Figure 3 shows the dependence obtained by us for the superconducting transition temperature of pure vanadium films on the thickness d_C of the carbon coating for two batches of samples that differ only in the thickness d_0 of the carbon sublayer. The samples of the first batch (Fig. 3, curve a) had a sublayer thickness $d_0 = 15$ Å. The samples of the second batch (Fig. 3, curve b) had $d_0 = 10$ Å. The vanadium film thickness was constant ($d_V = 120$ Å). Both curves are nonmonotonic. The maximum values of T_c for both curves are close to each other. However, the positions of the maxima differ quite strongly. For the samples of the first batch (Fig. 3, curve a) the first maximum takes place at $d_C = 3$ Å, whereas for the samples of the second batch (Fig. 3b) the first maximum is observed at $d_C = 7$ Å. The period of the oscillations is ~ 5 Å. Here, just as in[2], we observe a mutual influence of the dielectric layers located on opposite sides of a metallic film.

The oscillatory character of the $T_c(d_C)$ curve is less pronounced for the vanadium films than for the other superconductors investigated by us. There is no clear-cut dependence of the resistance on the thickness of the carbon coating. The apparent reason is the high sensitivity of the unprotected vanadium film to oxidation and the ensuing difficulty of obtaining in pure form the dependence of the properties of the metallic film on the thickness of the carbon coating.

As already mentioned, in the case of sandwich-type structures the effect of the oxidation is completely eliminated and all the dependences should manifest themselves more clearly. Figure 4 shows the superconducting-transition temperatures of two-layer (relative to vanadium) vanadium–carbon–vanadium samples as functions of the thickness of the carbon interlayer. The thickness of the metallic layers was constant at

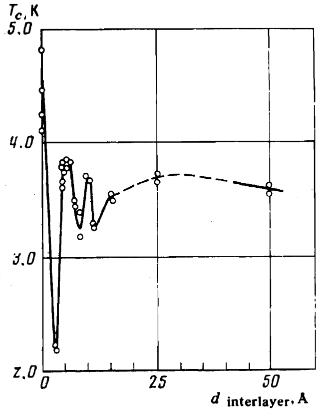

FIG. 4. Critical temperature of two-layer vanadium–carbon sandwiches as a function of the thickness of the carbon interlayer. The thickness of the carbon sublayer and of the coating is constant at 15 Å.

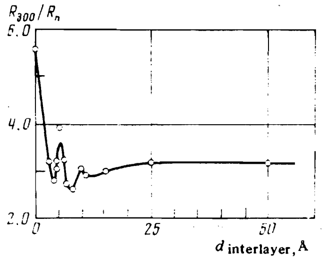

FIG. 5. Resistance ratio R_{300}/R_n of two-layer vanadium-carbon sandwiches vs the thickness of the carbon interlayer. Thickness of carbon sublayer and coating 15 Å.

100 Å. The curve was plotted for three batches of samples prepared in different sputtering experiments. The agreement between the critical temperatures of different samples having the same carbon-interlayer thickness allows us to state that we are not dealing with a random scatter of the points.

Figures 5–7 show plots of the normalized resistivities $\rho^*_{300}/\rho^0_{300}$, ρ^*_n/ρ^0_n and of the resistance ratio R_{300}/R_n against the carbon liner thickness $d_{\text{interlayer}}$ for the same samples (ρ^*_{300} is the resistivity of the sandwich at room temperature, ρ^*_n is the residual resistivity of the sandwich; ρ^0_{300} and ρ^0_n are resistivities of a film of pure vanadium of thickness $d_V = 200$ Å).

All the curves for the sandwiches are oscillatory, the oscillations being much more pronounced than in the case of vanadium films with variable carbon coating. The oscillation period is 5 Å, and the first two maxima on the $T_c(d_{\text{interlayer}})$ curve occur at liner thickness 5 and 10 Å. Attention is called to the fact that for sandwiches with carbon sublayer $d_0 = 15$ Å and with a coating $d_C = 15$ Å and with a coating $d_C = 15$ Å the first extremum on the $T_c(d_{\text{interlayer}})$ curve is always a minimum at $d_{\text{interlayer}} = 3$ Å, whereas for a pure vanadium film with $d_0 = 15$ Å interlayer the first extremum is a maximum at a carbon coating thickness $d_C = 3$ Å.

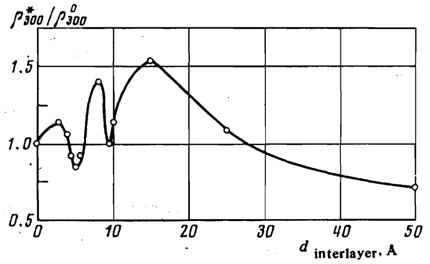

FIG. 6. Normalized resistivity at room temperature $\rho^*_{300}/\rho^0_{300}$ of two-layer vanadium-carbon sandwiches vs the thickness of the carbon interlayer. The carbon sublayer and coating are 15 Å thick.

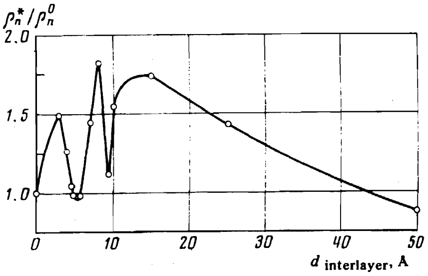

FIG. 7. Normalized residual resistivity ρ^*_n/ρ_n of two-layer vanadium-carbon sandwiches vs the thickness of the carbon interlayer. The carbon sublayer and coating are 15 Å thick.

In the case of sandwiches, a perfectly distinct correlation is observed between the critical temperature of the samples and the quantities ρ_{300}, ρ_n, and R_{300}/R_n. Whereas the resistance ratio R_{300}/R_n varies in phase with the changes of the critical temperature, then the resistivities ρ_{300} and ρ_n vary oppositely: the maximum resistance corresponds to the minimum T_c. The amplitude of the residual-resistance oscillations are much larger than the amplitudes of the oscillations of ρ_{300}. The critical temperature at any of the maxima of the $T_c(d_{\text{interlayer}})$ curve of two-layer sandwiches with metal-layer thickness $d = 100$ Å never exceeds T_c of a pure vanadium film 200 Å thick and having the same conditions on the boundary.

We have investigated also the properties of vanadium-carbon samples with five vanadium layers. Figures 8 and 9 show the plots of T_c and R_{300}/R_n against the thickness of the carbon interlayer for this case. The vanadium layer thickness in five-layer samples was constant at 100 Å, just as in the two-layer samples. The curves remained oscillatory, but the amplitude of the oscillations decreased with increasing number of layers in the sandwich. The oscillations were observed against the background of a monotonic variation of the critical temperature and of R_{300}/R_n, from a value corresponding to a vanadium film with $d_V = 500$ Å to a value equal to T_c of a $d_V = 100$ Å vanadium film. The electron-microscopy and electron-diffraction investigations of all the sandwiches, in transmission, have shown that the vanadium in such a layered system has a structure and lattice parameter that are typical of pure bulk metal. The

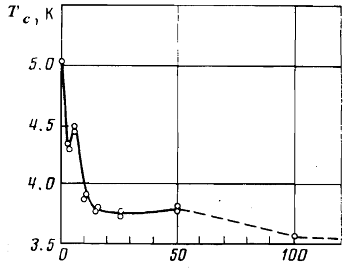

FIG. 8. Critical temperature of vanadium-carbon sandwiches with five vanadium layers vs thickness of carbon interlayer. The carbon sublayer and coating are 15 Å thick.

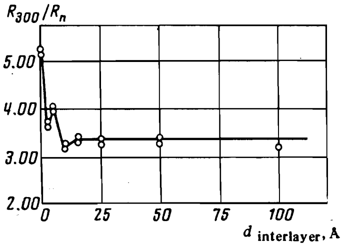

FIG. 9. Resistance ratio R_{300}/R_n of five-layer vanadium-carbon sandwiches vs the thickness of the carbon interlayer.

invariance of the lattice parameter and the great sharpness of the lines on the electron diffraction patterns of the sandwiches point to the absence of a large mixing (more than 2–3% in terms of carbon) of different layer to a great depth.4) The vanadium grain dimension in the sandwiches remains unchanged with changing thickness of the carbon liner. Thus, e.g., for $d_{\text{interlayer}} = 3$ Å the average grain dimension is $a \approx 500$ Å and for $d_{\text{interlayer}} = 25$ Å we have $a \approx 400$ Å.

DISCUSSION OF THE RESULTS

The similarity of the $T_c(d_{\text{interlayer}})$ curves for sandwiches and samples with dielectric coatings (the periods and the amplitudes of the oscillations coincide) indicates that the same physical mechanism is in operation in both cases. The results of electron-microscope and electron-diffraction investigations allow us to state that this mechanism is not connected with structure changes in the samples when the thickness of the carbon layer is varied.

Until recently there was no satisfactory explanation of the observed effect. All the mechanisms proposed so far lead to a monotonic dependence of T_c on the thickness of the dielectric layer, with saturation at large coating thicknesses. As already indicated,[2] to explain the nonmonotonic dependence of the critical temperature we must assume either a superposition of several known mechanisms, that make contributions of different signs, or the existence of a new mechanism that leads to an oscillatory dependence of the properties of the metallic film. Since in our experiments on vanadium we obtained besides oscillations of T_c, also pronounced oscillations of the resistance, which correlated with the changes of the critical temperature, we can propose that the mechanism responsible for the observed effect is connected with changes of the electron density of states.

Our experiments on superconducting films with dielectric coatings and on metal–dielectric sandwiches have shown that in such systems it is impossible to obtain a noticeable increase of the critical temperature as a result of the mechanisms discussed in[6]. None of the investigated samples had T_c higher than the critical temperature of bulk metal, although we were able to work with very thin metallic layers, and the properties of the dielectric layer were sufficiently distinctive.

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1)The physical, chemical, spectral, optical, mechanical, and structural investigations of the carbon films obtained by deposition of the carbon by the method described above[2,3] have shown this film to be a finely dispersed crypto-crystalline diamond body of the "carbonado" type. It can be assumed that the carbon layers in multilayered systems are of the same modification.

2)We have verified by special experiments that a synthetic-diamond layer 10 Å thick protects any metallic film completely against oxidation.

3)T_c of some samples was determined also by a contactless method, from the change of the inductance of a flat measuring coil placed near the sample. Both methods of measuring the critical temperature yielded practically the same results.

4)The absence of an appreciable and deep penetration of carbon atoms into the vanadium layer indicates also that with increaseing thickness of the carbon liner the value of T_c of a two-layer sandwich $V_{100\,\text{Å}}$–C–$V_{100\,\text{Å}}$ tends to the value of T_c of a pure vanadium film $d_V = 100$ Å thick. In addition, the sample in which special homogeneous mixing of V and C was carried out, with a carbon content ~3 at.%, revealed no superconductivity down to 1.2 °K.

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