The phenomenon of memory in vanadium ox nitride films

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Abstract. Vanadium oxides are representatives of a wide class of compounds possessing a metal-insulator phase transition. The properties of both the oxides themselves and their solid solutions with oxides of other metals have been well studied. The question of the possibility of dissolving nonmetallic components in the V-O system and its wobbling on the metal-insulator phase transition remain an open question. The solution of this problem, apart from the scientific side can also have practical significance: in spite of the wide possibilities and uses of vanadium oxides in optoelectronics, microwave technology and other fields, the need to search for materials with given temperatures of metal-insulator phase transition and electro-physical characteristics is obvious. In this paper, an attempt is made to dissolve nitrogen in vanadium dioxide films during their deposition by thermolysis of nitrogen-containing alkoxy derivatives.

1. Introduction

Investigations of the growth conditions of thin vanadium dioxide layers by thermal decomposition of organometallic vanadium compounds have shown that during the decomposition of compounds containing oxygen and any of the elements of Group IV, films with an admixture of this element can be obtained. In this way, thermolysis of tris (tert-butoxy) vanadium-chalcogenides and vanadium alkoxides containing nitrogen or chlorine, it was possible to precipitate: films of VOS, VON, VO-Cl systems [1]. In particular, the thermal decomposition of nitrogen-containing alkoxy derivatives of vanadium provides alloyage of nitrogen oxides of vanadium.

The deposition of the films was carried out at a temperature of 520 K, followed by annealing at a temperature of 570 K. As substrates, polycrystalline plates of pyroceramics, corundum, and single-crystal plates of lithium fluoride and magnesium oxide were used. The nitrogen content of the films was determined by gas chromatography using a "Color-I04" chromatograph using Ca-A and helium adsorbent-molecular sieves as a carrier gas. For this purpose, the films were annealed in quartz ampoules at a pressure of 300 mm Hg. and a temperature of 620 K. The accuracy of the determination of nitrogen is 15-20%. The content of vanadium was determined by spectrophotometric methods and also by methods of x-ray spectral fluorescence analysis using a spectrometer “Spark”. Vanadium determination accuracy is 5%. It was found that the nitrogen content per formula unit of vanadium dioxide is 0.1-0.2.

The temperature dependence of the electrical resistivity of the films was measured on a direct current by two-probe methods. Contacts to the samples were prepared by applying conductive glue based on polyacrylic resin to the surface of the film, as well as vacuum deposition of silver. For measurements of electrical resistance were used films with a width of 3 mm and a length of 3.5 mm. The current-voltage...
characteristics of the films were measured on a two-coordinate potentiometer, the TCP-021 recorder. For this purpose, in the circuit containing the power source, the test sample consistently included a reference linear resistor, the voltage at which the current in the circuit was determined. For measuring the A-V characteristics was used films with a width of 1 mm and a length of 0.5 mm.

2. Results and discussion
One of the results of the work was the establishment of memory in these films. While for the films of vanadium dioxide and other compounds of MIPT, the region of negative differential resistance on the volt-ampere characteristic is unstable and switching occurs in $10^{-4}, 10^{-3}$ sec in these samples a high-conductivity and low-conductivity state, differing in electrical resistance by almost two orders , can be stored at a voltage less than the threshold voltage.

Undoubtedly, the memory in these films is a consequence of the anomalous temperature hysteresis of the electrical resistivity. However, in order to quantitatively describe the effect, to establish its dependence on temperature and film dimensions, to find “coercive” voltages and other quantitative characteristics of memory, it is necessary to obtain an analytical form of the A-V characteristic, taking into account the hysteresis of the MIPT and the temperature dependences of the electrical resistivity in both phases.

Following the results of the experiment, we approximate the temperature dependence of the resistivity by the following expressions:

\[ \rho_k - \alpha (T - T_K), \quad \alpha > 0, \quad T \leq T_K; \]  
\[ \rho (T) = \rho_m - \beta (T - T_K), \quad \beta > 0, \quad T \geq T_K; \]  

Where, depending on the heating or cooling of the film, the values of $T_K, \rho_K,$ and $\rho_m$ are numbered respectively by the indices "1" and "2".

In the presence of internal energy sources (Joule heating), we take the heat transfer along the streamline to heat-intensive contacts. In this case, the heat equation becomes

\[ \frac{d^2T}{dx^2} + \frac{I^2\rho(T)}{kS^2} = 0 \]  

where is the I-current strength across the cross section of the film; $S$ is the area of the section; $K$ is the coefficient of thermal conductivity.

By solving equation (3) for given boundary conditions

\[ T \left( x = \pm \frac{\Delta}{2} \right) = T_0, \quad \frac{dT}{dx|_{x=0}} = 0 \]  

where $\Delta$ is the film width $T_0$ is the temperature of the thermostat, is

\[ T = T_x + \frac{\rho_x}{\alpha} - \frac{\rho(T_0)}{\alpha} \frac{ch \sqrt{\alpha} x}{S \sqrt{\kappa}} + \frac{ch \sqrt{\alpha}}{2h \sqrt{\kappa}} \]  

Thus, the resistivity of the plate due to the nonuniform temperature distribution along its surface is a function of the coordinate $x$. The total film resistance is then determined by integrating $\rho(x)$ from $\Delta/2$ up to $\Delta/2$ for a given thickness $h$ and $\ell$ length. The expression for the current-voltage characteristic takes the form:

\[ U = \frac{\rho(T_0) I^2 \ell}{I_a sch I / I_a arctgsh I / I_a} \]
where \(1/I_a = \frac{\sqrt{\alpha}}{2h\sqrt{k}}\).

When the temperature \(T_k\) is reached at the center of the film a high-conductivity phase channel appears in it. The corresponding threshold current is found from (4):

\[
I_n = \frac{2h\sqrt{k}}{\sqrt{\alpha}} \text{arcch} \frac{\rho(T_0)}{\rho_k}
\]

and the threshold voltage \(U_n\) is found by setting (6) in (5).

It is assumed that since the electrical resistivity of a high-conductivity phase is much less than that of a low-conducting phase, the current through the cross section of the film will be concentrated in the channel of a high-conductivity phase of width \(\delta\). In this case the heat conduction equation is divided into two:

\[
\frac{d^2T}{dx^2} = 0 \text{ in the channel};
\]

\[
\frac{d^2T}{dx^2} + \frac{I^2\rho(T)}{h\delta^2h^2} = 0 \text{ in the channel},
\]

where \(\rho(T)\) is identified by the expression (1b). Border conditions

\(T(x=\pm\Delta/2) = T_0\).  \(T(x=\pm\delta/2) = T_k, \frac{dT}{dx=x=0} = 0\), and also the "sewing" conditions for the equations (8) and (9) allow us to find the expression for the total resistance and set the type of A-V characteristic.

\[
U = \frac{I\ell\beta(T_k-T_0)}{SshI/\beta \text{arctgshI} / I_{\beta}} + \frac{\rho_m I^2\ell}{I_{\beta}SshI / \beta \text{arctgshI} / I_{\beta}}
\]

where \(1/I_{\beta} = \frac{\sqrt{\beta}}{2h\sqrt{k}}\).

The first term in (7) is determined by the expansion of the channel of the high-conductivity phase, the second term is determined by the voltage on it.

The superposition of these contributions determines the negative.

Obviously, expressions (5) and (8) are valid for the A-V characteristic both with current growth and with its decrease, with the difference that in the first case, the expressions include the parameters \(T_{x1}, \rho_{x1}, \rho_{u1}\) and in the second \(T_{x2}, \rho_{x2}, \rho_{u2}\). Hence it follows that there can exist a voltage \(U_0\), at which the negative branch of the A-V characteristic with parameters \(T_{x1}, \rho_{x1}, \rho_{u1}\) intersects with the positive IV characteristic with parameters \(T_{x2}, \rho_{x2}, \rho_{u2}\). This means the presence of stability of a high-conductivity state at a given voltage. It is obvious that since this voltage is less than the pore pressure it can also be answered by a low-conductivity state described by (5) with parameters \(T_{x1}, \rho_{x1}, \rho_{u1}\). Thus, the occurrence of the temperature hysteresis of the electrical resistivity realizes the memory on the A-V characteristic.
Based on the results obtained, the "contrast" of the record in this model is estimated. The maximum current of a stable, highly conducting state I0 is found from equation \( \frac{dU(T_1, \rho K_1, \rho M_1)}{dI} = 0 \) where U is determined by the expression (8). The current in a low-conducting state is accurately estimated from Ohm's law. In the final analysis, in the approximation of small \( \beta \) we have

\[
I_0/I_g = \frac{1}{2} \frac{\rho(T_0)}{\rho_m}
\]  

(11)

The appearance of the factor \( I/2 \) in (8) reflects the fact that, because of the boundary conditions, not all of the volume of the film passes into a highly conducting state. \( I_0/I_g \) ratio. The one determined from the \( \rho(T) \) dependence is 23, while the one determined from the A-V characteristic is 70.

The voltages \( U_1 \) and \( U_2 \), which must be applied to the film to transfer the latter from the low conductive to the highly conductive state and vice versa are measured. The operating voltage \( U_0 \) corresponding to the current \( I_0 \) is defined as

\[
U_0 = 4l_1 \sqrt{\frac{\rho_m K(T_{K1} - T_0)}{\Delta}}
\]  

(12)

The voltage \( U_1 \) necessary for transferring a film from a low conductor to a high-conductivity state is determined by the difference \( U_n \) and \( U_0 \):
\[ U_1 = \frac{\rho(T_0)l^2}{l_{\text{Sch}}I_{\text{arcsh}}/I_{\alpha}} - \frac{4l\sqrt{\rho_{\text{m1}}(T_{\text{K1}}-T_0)}}{\Delta} \]  

(13)

The voltage \( U_2 \) required to transfer the film from a high conductive to a low conducting state is determined by the difference \( U_0(T_{\text{K2}}, \rho_{\text{K2}}, \rho_{\text{M2}}) \) and \( U(T_{\text{K1}}, \rho_{\text{K1}}, \rho_{\text{M1}}) \):

\[ U_2 = \frac{4k\xi}{\Delta} - \left( \sqrt{\rho_{\text{M2}}(T_{\text{K2}} - T_0)} - \sqrt{\rho_{\text{M1}}(T_{\text{K1}} - T_0)} \right) \]  

(14)

Thus, by supporting the voltage \( U_0(T_{\text{K1}}, \rho_{\text{K1}}, \rho_{\text{M1}}) \) on the film, the voltage pulses \( U_1 \) and \( U_2 \) can be transferred from the low conducting state to the high conducting state and vice versa.

3. Conclusion

A vanadium dioxide film with a nitrogen content of 0.1-0.2 and a formula unit \( \text{V}_0 \) on various substrates was prepared by thermolysis of nitrogen-containing alkoxy derivatives of vanadium. The electrical properties of the film were investigated. A jump in the electrical resistance was observed in the 250 K region and an anomalously large temperature hysteresis of the electrical resistivity in the investigated films. The effect of electric memory was found in \( \text{V}_0 \)-0.18 N films.

A memory phenomenon was detected on the current-voltage characteristic in investigated films. It is shown that memory is considered to be a consequence of a jump in electrical resistance and its temperature hysteresis.

An analysis has been carried out to find the dependence of the effect on the temperature and film dimensions and also to establish certain quantitative characteristics of memory.

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