Anomalous features of the conductivity and of the galvanomagnetic properties of vanadium dioxide in strong electric fields

E. V. Babkin, G. A. Petrakovskiĭ, and A. A. Charyev
L. V. Kirenski Institute of Physics, Siberian Branch of the Academy of Sciences of the USSR

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The results of a study of the effect of a strong electric field on the kinetic properties of vanadium dioxide films are reported. The experimental method is based on the contact effects occurring in layered metal-insulator-semiconductor structures. The conductivity, magnetoresistance, and nonlinear resistance properties of the samples were found to behave anomalously in an electric field. The results are discussed in terms of the concept of a spontaneous macroscopic current in the films induced by the transition of the system to the toroidal current state.

In this letter we report an observation of the effect of a strong electric field on the conductivity, on the magnetoresistance, and on the nonlinear resistance properties of vanadium dioxide VO_2. At a temperature of 340 K, vanadium dioxide undergoes a metal-insulator phase transition. In general, the effect of the electric field on the properties of VO_2 may be linked with the difference in energies of the electric field $E^2/8\pi$ in the metal and insulator, which is seen in the removal of the field from the metal.[1] Of considerable interest is the study of the evolution of the properties of the crystal as a result of increasing the strength of the electric field, at which point its energy becomes comparable to the difference in the energies of the insulator and metal phases.

To produce a strong electric field in the crystal, we used a layered structure consisting of an insulator substrate, whose opposite sides were coated with films of nonmagnetic metal, and the compound under study. The substrates we used were 0.1-cm-thick barium titanate ceramic plates with a dielectric constant of 6100. The copper films were deposited by the vacuum evaporation method. The thickness of the copper film was 1000–3000 Å. The vanadium dioxide films were deposited by the method of vanadium acetylacetonate pyrolysis. The thickness of the vanadium dioxide film was 200–1500 Å. The contacts for the samples were fabricated by depositing a conducting cement based on a polyacrylic resin. The contacts on the VO_2 films were spaced an equal distance apart (0.1 cm) for all the samples.

An application of a voltage to the metallic electrode causes the normal component of the electric induction to jump considerably at the film-substrate interface because of the large dielectric constant of the substrate. Estimates have shown that at a voltage $U = 1000$ V the strength of the electric field in this case is 10^7–10^8 V/cm and the energy of the electric field in this case is 10^8–10^9 erg/cm^3. The difference in the energies of the insulator and metal phases of VO_2, ΔF, which is determined by the latent heat of the phase transition and by its density, is 1.8×10^9 erg/cm^3. The condition $E^2/8\pi = \Delta F$ can thus be satisfied in this experiment. Working from the estimate of the electrical capacitance of the system, the change in the density of the current carriers in this case is 10^{19}–10^{20} cm^{-3}.

The electrical measurements were carried out with direct current. The measuring apparatus which included a current generator, a detector and a shunt system, controlled and regulated the current within 0.05–12 μA. A thermally stabilized chamber with the sample was placed in the gap of the electromagnet whose magnetic field strength was varied in the range 0.1–22 kOe.

Several samples were used to measure the resistivity as functions of the voltage across the metallic electrode, the magnetic field strength, the current strength along the cross section, and the temperature. Qualitatively unambiguous results were obtained in each case. Figure 1a is a plot of the resistivity of a VO_2 film 260 Å thick as a function of the voltage U at various temperatures. The direction of the electric field is such that the VO_2 film has a positive charge. There is no resistivity minimum when the field is reversed and the resistivity increases monotonically with increasing voltage. We observed a similar behavior in the case of films of thickness 830, 1040, and 1440 Å but at a higher voltage. The voltage U_0, which corresponds to the resistivity minimum, depends monotonically on the temperature and at a constant temperature increases linearly with increasing strength of the current. The appearance of a strong magnetoresistance effect in the films at $U \neq 0$ was an unexpected result in the experiments. The samples can be restored to their original state corresponding to $U = 0$ by increasing the strength of the magnetic field which is applied normal to the plane of the film. The return of the samples to their original state can also be accomplished by going through the $\rho(H)$ minimum nonmonotonically (Fig. 1b). The magnetoresistance exhibits a

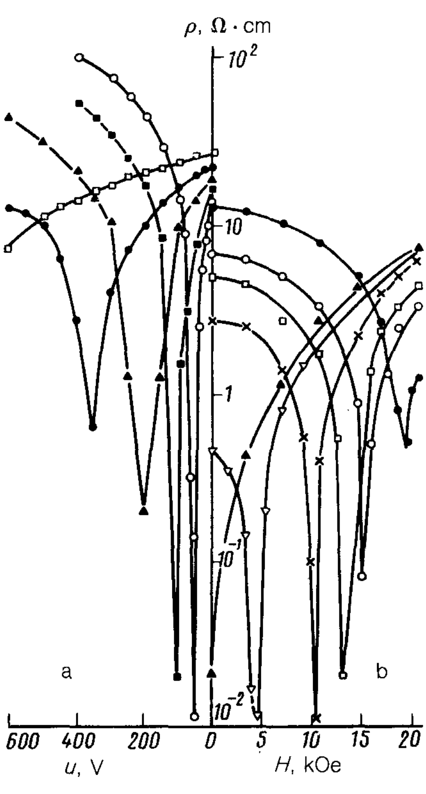

FIG. 1. (a) Resistivity of a 260-Å-thick VO_2 film versus the voltage at various temperatures: □—467 K; ●—482 K; ▲—496 K; ■—512 K; ○—529 K. The current flowing through the film is 0.112 μA. (b) Resistivity of a 260-Å-thick VO_2 film versus the strength of the magnetic field applied at right angles to the film surface at various temperatures: ▲—54 V; ▽—60 V; ×—70 V; □—80 V; ○—90 V; ●—100 V. The measurements were carried out at a temperature of 529 K; the current flowing through the film is 0.112 μA.

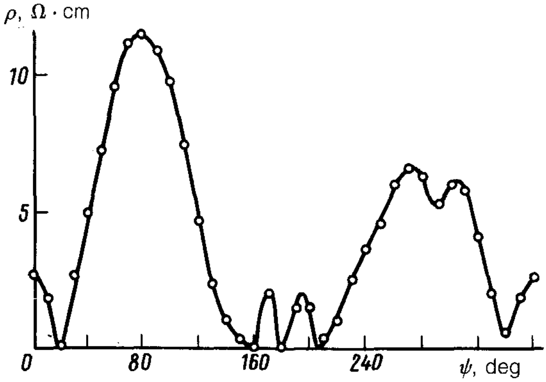

FIG. 2. The angular dependence of the magnetoresistance of a 260-Å-thick VO_2 film in a 10-kOe magnetic field at a voltage of 192 V and temperature of 490 K. The current flowing through the film is 0.112 μA. The angle ψ is reckoned from the normal to the surface of the film lying in a plane perpendicular to the direction of the electric current.

strong anisotropy which depends on the temperature and on the applied voltage (Fig. 2).

The anomalous behavior of the kinetic properties of vanadium dioxide in a strong electric field cannot be explained in terms of the classical theory which was developed for semiconductors and metals. A strong decrease in the resistivity of the samples at a certain critical voltage leads us to assume that a new state of the crystal, which is sensitive to the magnetic field and current, appears in this region. To account for the results, we can use the hypothesis of the existence of a spontaneous macroscopic current in the crystal produced as a result of transition of the system to the cooperative toroidal current state.[2–4] Under nonequilibrium conditions caused by the external current in the presence of a dissipation, there can be an anomalous conductivity of the torodial current state at the phase transition point. This anomalous conductivity is similar to the Curie-Weiss susceptibility whose amplitude depends on the current.[5] The magnetoresistance effect in this case may be the consequence of an anomalous diamagnetism. The toroidal current state cannot occur without a lowering of the symmetry of the crystal induced by the electric field. Along with the lowering of the symmetry, however, the role of the electric field reduces to a dramatic change in the state of the current carriers (a change in their density, in their energy spectrum, etc.). The matching of the conditions for the occurrence of the toroidal current state with these parameters may be the cause of a sharp decrease in the resistivity of the films at certain electric and magnetic field strengths and at certain temperatures.

Although these results can conceivably have other interpretations, we must confine ourselves to this interpretation for want of other choices. A definitive conclusion can be made after some additional experiments and in-depth theoretical analysis.

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