Characteristics of electronic properties of uranium films condensed on cold substrates

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An investigation was made of the electrical resistivity and galvanomagnetic properties of uranium films condensed on cold substrates, and also of changes in these properties during heating. A comparison was made of the electronic properties of films and bulk samples of the original uranium. A nonlinear dependence of the Hall emf on the magnetic field was observed below 43°K and analyzed. A negative temperature coefficient of the resistivity exhibited by uranium films was attributed to a possible reduction in the carrier density as a result of cooling.

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I. INTRODUCTION

Metastable amorphous modifications are observed when some metals are condensed in vacuum on liquid-helium-cooled substrates.[1] It has been suggested that these amorphous modifications are most likely to appear for metals whose lattices differ very greatly from the close-packed form.[2] One of the criteria of the formation of amorphous modifications can also be the occurrence of polymorphism of a given metal.[3] These two conditions are satisfied by uranium which has the orthorhombic lattice at temperatures below 940°K and which undergoes two polymorphic transitions at lower temperatures. Amorphous modifications of metals usually have very interesting electronic properties (for example, superconductivity[4]) and this is why they are being investigated at present. Electronic properties of amorphous metals are frequently analogous to their properties in the bulk under pressure.[5] For example, it is known that α uranium exhibits a strong increase in the superconducting transition temperature T_c on increase in pressure up to ~12 kbar (Ref. 6).

For these reasons we decided to investigate uranium films condensed on cold substrates. To the best of our knowledge, the electronic properties (electrical resistivity, Hall effect, and magnetoresistance) of uranium films have not yet been investigated. These properties are of interest, in particular in connection with a second-order phase transition which occurs in α uranium at about 43°K.[1)]

We shall report a study of the electrical conductivity and galvanomagnetic properties of uranium films immediately after their condensation on liquid-helium-cooled substrates and in the course of subsequent heating.

II. METHOD

Serious difficulties are encountered in the preparation of pure uranium films because of the high chemical activity and low vapor pressure of this metal even at temperatures much higher than the melting point.

Attempts to evaporate uranium by electron bombardment, so as to minimize the capture of impurities by films, have not been successful: a thin uranium rod acting as an anode melts right to the base without evaporation. The use of beryllium or magnesium oxide crucibles[8] is acceptable for molten uranium but it is not suitable for evaporation because uranium interacts with the crucible material at high temperatures. Evaporation from current-heated tungsten wires results in some contamination of the condensate with tungsten atoms. However, the last method is still the best under the circumstances. Mass-spectrometric investigations of uranium evaporated from a tungsten wire at $T = 1900$°K (Ref. 9) have shown that the tungsten peak in the spectrum does not exceed the background level, which represents about 3% of the uranium peak. It follows that the vapor flux contains less than 3% of tungsten atoms.

The method used to prepare and investigate uranium films used in our study had been described in detail earlier.[1] The starting material in the film preparation was uranium of 99.94% purity. The main impurities were (in at.%): 10^{-2} Mo; 2.3×10^{-3} Fe; 2×10^{-3} Al; 1.1×10^{-3} Si; 4×10^{-2} C. The ratio of the room-temperature resistivity of the original uranium to the resistivity at liquid helium temperature was 10.1.

Uranium was evaporated from a tungsten helix in a glass ampoule where a high vacuum was maintained; the ampoule was sealed off from a vacuum system and placed in a cryostat with liquid helium. Condensation of the residual gases on the cold walls of the ampoule reduced the pressure to ~10^{-12} Torr before evaporation. The rate of deposition of uranium was on the average ~200 Å/min. Films were condensed on polished glass substrates which had welded-in tungsten wires for electrical measurements. The temperature of the substrate during condensation did not exceed 30–35°K. The minimum substrate temperature, attained by reduction of its thickness and imporvement of liquid helium cooling, was 15°K when uranium was condensed at the optimal rate. The thickness of the thickest films was determined interferometrically with a maximum error <15–20%. Film thicknesses less than 300–400 Å

were estimated roughly from the condensation time.[1] The electronic properties of the films were compared with the corresponding properties of the original uranium. Our bulk sample had the dimensions $16 \times 0.4 \times 0.1$ mm.

The Hall emf and electrical resistivity were determined using an R-348 dc potentiometer. A magnetic field up to 40 kOe, directed along the normal to the film plane, was created by a superconducting solenoid. Films of thickness ~1000–1500 Å were heated to room temperature and immediately after opening of the ampoule they were subjected to an x-ray structure determination carried out using copper radiation and RKU-114M and RKD cameras.

III. EXPERIMENTAL RESULTS

1. Monitoring of heating of uranium films

The method of analysis of the singularities of temperature dependences of the electrical resistivity is sufficiently sensitive for the detection of allotropic transitions, because of the high structural sensitivity of the resistivity. The transition of a metal from one modification to another (for example, from the amorphous to crystalline form) is usually accompanied by a jump of the resistivity ρ in a fairly narrow range of temperatures.[1] The temperature dependences of the resistivity of metal films condensed on cold substrates and having the same crystal structure as the bulk metal are smooth curves without any singularities. The resistivity then changes as a result of annealing of defects and grain growth. Figure 1 shows the gradual changes in the electrical resistivity exhibited by a uranium film condensed on a substrate cooled with liquid helium and then heated to room temperature. There are no resistivity discontinuities in the dependence $\rho(T)$.

Similar $\rho(T)$ dependences were observed up to $T \approx 200°$K for all the investigated films of thickness in the range 50–1000 Å. However, in the region of $T \approx 200°$K the resistivity of thin films usually increased and the steepness of the rise became greater on reduction in the thickness. The resistivity continued to rise up to room-temperature and increased with time. This change in the behavior of the resistivity in the region of $T \approx 200°$K was in all cases accompanied by a rise of the Hall emf
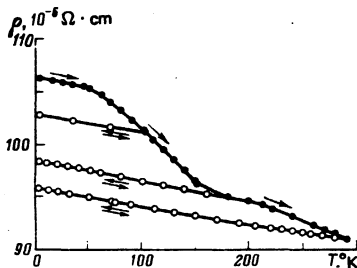

FIG. 1. Changes in the electrical resistivity resulting from the heating of a uranium field $d \approx 620$ Å thick (U-11). The open circles represent the value of ρ obtained in the course of cooling after heating to the indicated temperatures.
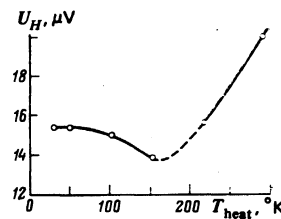

FIG. 2. Hall emf of sample U-11 in a field $H = 15.8$ kOe at $T = 4.2°$K plotted as a function of the heating temperature T_{heat}. The current through the sample was $I = 1$ mA.

U_H at $T = 4.2°$K (after heating to $T \gtrsim 200°$K), as shown in Fig. 2. The films heated to room temperature had the highest values of U_H at $T = 4.2°$K. In the case of the thin films the rise of U_H after heating to temperatures exceeding 200°K was much steeper than for the thick films. Clearly, the change in the behavior of the $\rho(T)$ curves in the region of $T = 200°$K was due to the absorption of residual gases by the films in the ampoules.

An x-ray structure analysis of the thickest ($d \approx 1000$–1500 Å) uranium films heated to room temperature indicated the presence of only the α-uranium lines in the Debye diffractograms.[2] Texture effects were exhibited by the (020), (130), and (131), (040) lines.

2. Electrical resistivity and its temperature coefficient

Immediately after condensation of a uranium film on a liquid-helium-cooled substrate the resistivity was $\sim(100-110) \times 10^{-6}$ $\Omega \cdot$cm (Fig. 1). This was a fairly high value in view of the fact that even amorphous metallic films have resistivities close to those of the molten metal.[4] The value of ρ for liquid uranium at its melting point is $\sim64 \times 10^{-6}$ $\Omega \cdot$cm (Ref. 11), which is much less than the resistivity of our films. The thickest uranium films had the room-temperature resistivity $\rho \approx 90 \times 10^{-6}$ $\Omega \cdot$cm (Fig. 1), which was again about 3 times greater than the resistivity of the original bulk uranium.

A negative temperature coefficient of the resistivity of our films (Fig. 3a) was also unexpected in view of the fact that the usual metallic-type dependence $\rho(T)$ was exhibited by the original uranium (Fig. 3b). Like the rare-earth metals,[12] which have structures similar to that of uranium, the dependence $\rho(T)$ for uranium (Fig. 3b) obeyed satisfactorily the law $\rho \propto T^3$ in the temperature range 4–30°K. The dependence $\rho(T)$ for the uranium films was nearly linear and the temperature coefficient was practically constant for films heated to different extent and having different thicknesses (Fig. 3a). The resistivity was investigated in the temperature range 1.5–300°K. Cooling to 1.5°K revealed no signs of approach to the superconducting state and this was true of freshly condensed and heated uranium films.

3. Hall effect

The Hall effect in our uranium films was investigated in the temperature range 4.2–50°K. This was preceded
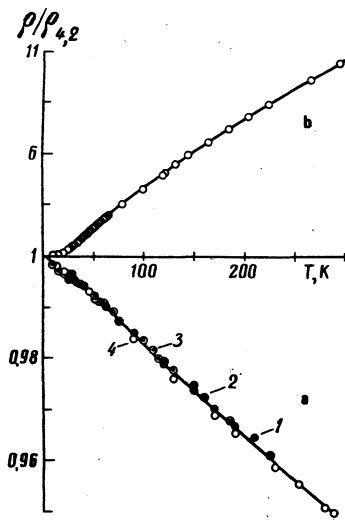
FIG. 3. Temperature dependences of the reduced resistivity $\rho/\rho_{4.2}$ ($\rho_{4.2}$ is the resistivity at $T = 4.2\,°K$). a) Heated uranium films: 1) U-2 ($d = 80$ Å, $T_{heat} = 210\,°K$); 2) U-3 ($d = 750$ Å, $T_{heat} = 260\,°K$); 3) U-8 ($d = 140$ Å, $T_{heat} = 190\,°K$); 4) U-11 ($d = 620$ Å, $T_{heat} = 295\,°K$). The points corresponding to heating of the same films to lower temperatures (50–150 °K) are plotted on the same curve. b) Similar results for a bulk sample.

by a study of the Hall effect in bulk samples of uranium[13] in the temperature range 1–300°K in fields up to 30 kOe. It was found[13] that at all temperatures the Hall emf U_H was a linear function of the magnetic field. However, our study at 4.2°K revealed nonlinear dependences $U_h(H)$ for freshly condensed and heated (to room temperature) films (Fig. 4a). A study of a bulk sample of the original uranium showed that the behavior of $U_H(H)$ was similar to that exhibited by films (Fig. 4b).

In the field range 2–40 kOe the dependence $U_H(H)$ for films and bulk samples was described by

$$U_H = a + bH^{\gamma_h},$$

where a (<0) and b (>0) are the constants of the sample. In weak fields ($H < 2$ kOe) the dependences $U_H(H)$ were not determined accurately but they were clearly linear. When the criterion of deviation of the dependences $U_H(H)$ from linearity (in fields $H > 2$ kOe) was taken to be the ratio $U_{H\,(15)}/U_{H\,(5)}$,[3] plotting of this ratio as a function of temperature indicated (Fig. 5) that near $T = 40°K$ the dependence $U_H(H)$ became linear for fresh-

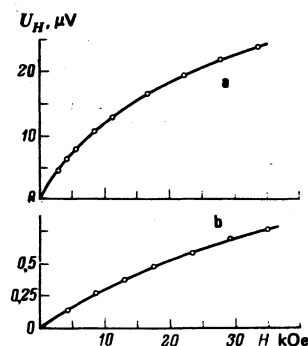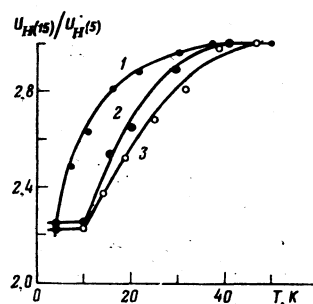FIG. 5. Ratio of the Hall emf's in fields of 15 and 5 kOe plotted as a function of temperature for film U-11 ($d = 620$ Å) after heating to temperatures (°K): 1) 50; 2) 217; 3) 295.

ly condensed and heated uranium films.

The dependence $U_H(H)$ for a bulk sample of the original uranium at $T = 77°K$ was also linear, corresponding to a Hall coefficient $R_H = +0.36 \times 10^{-10}$ m^3/C, which was close to the results obtained by other authors at the same temperature.[13] The value of the Hall coefficient for freshly prepared uranium films 300–1000 Å thick at ~50°K was $(1.9–2) \times 10^{-10}$ m^3/C, which increased to $R_H = (2.2–2.5) \times 10^{-10}$ m^3/C after heating to room temperature.

Cyclic variation of the magnetic field ($0 \to +H \to 0 \to -H \to 0$) always resulted in hysteresis of the $U_H(H)$ curves. The magnitude of this hysteresis increased on increase in $|H|$. The maximum hysteresis (after application of fields $H = \pm 40$ kOe) amounted to about 0.1–0.2% at the point $H = 0$ for the thickest freshly condensed films. After heating to room temperature the hysteresis became much greater (sometimes by almost one order of magnitude). In the case of a bulk uranium sample the strongest hysteresis at $T = 4.2°K$ amounted to 0.4–0.5%. An increase in temperature reduced the hysteresis of the $U_H(H)$ curves very strongly and this was true of films and bulk uranium.

4. Magnetoresistance

Typical field dependences of the transverse magnetoresistance of the investigated uranium films, obtained at $T = 4.2°K$, are plotted in Fig. 6a. A characteristic feature of all the samples was the small magnetoresistance ($|\Delta\rho/\rho| < 0.03\%$). The dependence of $\Delta\rho/\rho$ on H was not quadratic, in contrast to the majority of nonmagnetic metals in weak fields. In the case of some

FIG. 4. Dependences of the Hall emf (at $T = 4.2\,°K$) on the magnetic field: a) freshly condensed U-3 film ($d = 750$ Å, $I = 1$ mA); b) bulk sample ($I = 3$ A).
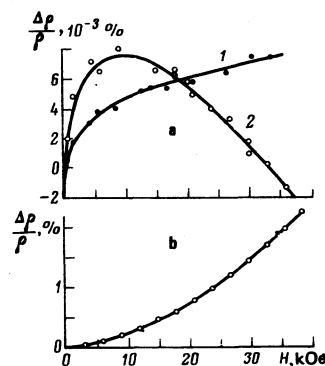

FIG. 6. Dependences of the transverse magnetoresistance on the magnetic field intensity: a) for U-3 film ($d = 750$ Å) immediately after condensation (1) and after heating to room temperature (2); b) bulk sample. The quantity $\Delta\rho$ represents the change in the resistivity under the influence of a magnetic field.

films heated to room temperature an increase in the magnetic field even reversed the sign of magnetoresistance (curve 2 in Fig. 6a).

For comparison, Fig. 6b shows the field dependence of the magnetoresistance of a bulk sample. In this case the magnetoresistance increased with the magnetic field almost quadratically. The maximum hysteresis of the $\Delta\rho/\rho$ curves in the dependence on H was a few thousandths of a percent in the case of films, whereas for a bulk sample it was 0.06%.

IV. DISCUSSION OF RESULTS

Our $\rho(T)$ and $U_H(T_{heat})$ dependences (Figs. 1 and 2), similar for all the films in the thickness range 50–1000 Å, and the results of structure analysis of the thickest ($d = 1000–1500$ Å) heated films indicate that homogeneous amorphous uranium films are not formed under the above experimental conditions. Even the thinnest of the films, least heated during condensation (by not more than 15°K), have clearly the α-uranium structure. However, we cannot exclude the possibility that the film structure immediately after condensation may be a mixture of the amorphous and α-uranium modifications. However, even in this case there should be a resistivity discontinuity at the temperature of transition of the bulk of the amorphous phase to the crystalline form (as found, for example, for beryllium[1,14]). Moreover, a polymorphic transition would have been accompanied by a change in the temperature dependence of $\rho/\rho_{4.2}$, which is not observed experimentally (Fig. 3a).

It is at present difficult to explain the relatively high electrical resistivity of our uranium films and the negative temperature coefficient. It is also surprising that the temperature coefficient of the resistivity is independent of the degree of heating and film thickness. There is little doubt that uranium films condensed on cold substrates are electrically continuous throughout the investigated range of thicknesses. Therefore, all the mechanisms of activated conduction in island films[15] are inapplicable. A negative temperature coefficient may be exhibited also by granulated films[16] composed of metal grains separated by insulating layers. However, as shown earlier,[1] the adopted method ensured the cleanest conditions for the formation of metallic condensates, which effectively excluded the possibility of formation of granulated films. Since our tungsten evaporator was outgassed and films were condensed in an ampoule whose walls were washed with liquid helium under pumping conditions ($T < 2.4$°K), no impurities, apart from those contained in the evaporated uranium charge, could have reached the film.

It is most likely that our films were contaminated with uranium dioxide UO_2, because a prepared charge naturally becomes oxidized during the preparatory stages. However, since the vapor pressure of UO_2 near the uranium evaporation temperature was approximately two orders of magnitude less than the vapor pressure of uranium,[17] the amount of the UO_2 impurity in our films was clearly slight. In fact, mass-spectroscopic investigations[9] failed to reveal the presence

of UO_2 molecules (to within 3%) during evaporation of uranium. Moreover, granulated films were characterized by an exponential temperature dependence of the resistivity. This dependence was nearly linear for our films. Granulated films with a negative temperature coefficient usually contain over 50 vol.% of the insulator and have much higher resistivities than our uranium films ($>10^{-2}$ $\Omega\cdot$cm, Ref. 16). The presence of any uranium compounds (for example, UO_2) in our films would be easy to reveal by x-ray diffraction if their amount exceeded a few percent. However, x-ray analysis of heated thick films failed to reveal the presence of other phases apart from the orthorhombic α uranium. Thus, the relatively high resistivity and its negative temperature coefficient are the properties of relatively pure films of α uranium condensed on glass.

Similar behavior has been reported earlier for bismuth films, which also have a higher value of ρ than the bulk metal and a negative temperature coefficient whereas the bulk material has a positive one.[18,19] It has been found[18] that the value of the temperature coefficient of the resistivity of bismuth is governed by the competition between the temperature dependences of the carrier density and mobility. In the case of a bulk metal a reduction in the number of carriers as a result of cooling is masked by a stronger rise of the carrier mobility, which results in a positive temperature coefficient. In the case of films the rise of the mobility as a result of cooling is much less and the more effective reduction in the carrier density is responsible for the negative temperature coefficient.

Similar behavior can be expected also in the case of uranium. The slight change in the overlap of the $6d$, $7s$, and $5f$ energy bands as a result of changes in temperature or purity may alter the number of carriers. This hypothesis is supported, in particular, by the considerable difference (up to sign reversal) between the Hall coefficients of uranium samples of different purity at low temperatures.[13] The analogy between bismuth and uranium can be extended also to the Hall effect: films of both metals are characterized by Hall coefficients which are very different from the bulk values.

Before discussing the Hall effect in uranium at temperatures below ~43°K, we must recall the physical properties of uranium in the vicinity of this temperature.

Investigations of the temperature dependences of the magnetic susceptibility,[20] specific heat,[7] atomic volume,[21] and elastic moduli[22] have led to the conclusion that a second-order phase transition occurs in α uranium at $T \approx 43$°K. It has been found[23] that the observed change in the lattice parameters and atomic volume of α uranium[21] may be due to a transfer of 0.04 electron (per atom) from the filled (with six electrons) $6d$ and $7s$ levels to a free $5f$ level. According to many authors,[7,20] the electronic transition in α uranium at 43°K may be accompanied by antiferromagnetic ordering. The magnetic moment per uranium atom is clearly very small (~0.04 μ_B). This may be

why the first attempts to determine the magnetic structure of α uranium by neutron diffraction have not been successful.[21,24]

The nonlinear dependence of the Hall emf on the applied magnetic field (Fig. 4) appears, like the above anomalies of the physical properties,[20-22] below ~40°K (Fig. 5). Thus, this nonlinearity is one of the manifestations of a new state of uranium which appears as a result of a second-order phase transition. This behavior of $U_H(H)$ may be due to a change in the overlap of the $6d$, $7s$, and $5f$ energy bands, and a consequent change in the number of carriers on increase of the magnetic field. For example, a strong change in the Hall emf for a slight change in the field is exhibited by graphite, which is a typical semimetal.[25] Clearly, the $U_H(H)$ dependence observed for uranium can be explained by a very small change in the carrier density, as indicated by the above data[13,23]: transfer of just 0.04 electrons from the $6d$ and $7s$ levels to a $5f$ level alters greatly the Hall emf (the change may include sign reversal). Another possible reason for the nonlinearity of $U_H(H)$ is the postulated magnetic ordering of uranium at $T < 43$ °K. In this case the kink of the $U_H(H)$ curve on increase of the field may be due to the deformation of the antiferromagnetic structure in the form of gradual rotation of the magnetic moments with the field.[4) The hysteresis of the $U_H(H)$ curves observed in this range of temperatures also supports the existence of magnetic order in α uranium and can be explained by the delay of rotation of the magnetic moments behind changes in the magnetic field.

The increase in the hysteresis and the shift toward higher temperatures of the dependences of $U_{H\,(15)}/U_{H\,(5)}$ on T as a result of annealing of the films (Fig. 5) is evidence of a strong influence of the degree of distortion of the crystal lattice on the kinetics of the second-order phase transition observed in α uranium. We can see that such distortions tend to displace the transition toward lower temperatures. The existence of magnetic order in α-uranium films is supported also by the negative component of the transverse magnetoresistance (Fig. 6a). In accordance with the Matthiessen rule, the total resistivity of a magnetically ordered metal can be described by

$$\rho(T) = \rho_{res} + \rho_{ph}(T) + \rho_{mag}(T), \qquad (1)$$

where ρ_{res} is the residual resistivity; $\rho_{ph}(T)$ is the resistivity due to the scattering of conduction electrons by phonons; $\rho_{mag}(T)$ if the magnetic contribution due to the disorder in the spin system (it can be considered as the result of scattering by spin waves).

In the presence of a transverse magnetic field we observe a magnetoresistance $\Delta\rho/\rho$, which—in accordance with Eq. (1)—can be described by

$$\frac{\Delta\rho}{\rho} = \frac{\Delta(\rho_{res} + \rho_{ph})}{\rho} + \frac{\Delta\rho_{mag}}{\rho}. \qquad (2)$$

The first term of Eq. (2) is always positive but the second may be negative.

The dependence of $\Delta\rho/\rho$ on H shown in Fig. 6 can thus be explained by the competition between the positive and negative terms in Eq. (2). In the case of bulk pure uranium (Fig. 6b) the usual positive magnetoresistance is relatively strong, which is due to the much greater mean free path of electrons than in films. This clearly masks the small negative contribution of the magnetic term to the total magnetoresistance. However, although our results provide an indirect confirmation of the antiferromagnetic order in α uranium at $T < 43$°K, the existence of this order cannot be regarded as finally established.

[1)A brief review of this subject can be found in Ref. 7.
[2)The x-ray diffractograms were calculated using the dimensions of the unit cell of α uranium given in Ref. 10.
[3)Here, $U_{H(15)}$ and $U_{H(5)}$ are the Hall emf's in fields of 15 kOe and 5 kOe.
[4)The Hall emf varies nonlinearly on increase of the field in the case of, for example, some rare-earth metals below the Néel temperature.[26]

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