Thin Monocrystalline Films of High-Melting Metals for Metal Nanoelectronics

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Abstract

Monocrystalline thin (10–100 nm) films of high-melting metals (W, Mo, Nb, Ta) are obtained by means of pulsed laser vaporization in ultrahigh vacuum. The films are characterized by perfect bulk structure and interfaces. The mean amplitude of the roughness of outer surface is about 0.2 nm. As a result, unusually long residual free paths of electrons are achieved. They exceed film thickness by an order of magnitude. The results obtained in the present study open the possibility to produce ballistic electron devices of the nanometre size based on the films of high-melting metals.

INTRODUCTION

A novel interesting area of electronics is connected with the potential use of metal nanostructures as electron devices. There are two important length scales for mesoscopic structures, *i. e.* the length of electron phase interruption which is about 1 μm for metals at low temperature and about 10 nm at room temperature, and electron free path length. The ratio of the latter to the size of nanostructure determines which type of electron transport (diffusion or ballistic) takes place in the structure. Major attention has been paid to the investigation of metal nanostructures with diffusion-type electron transport that were prepared from irregular polycrystal metal films. Till the present moment, ballistic transport has been realized only in heteroepitaxial semiconductor nanostructures on the basis of two-dimensional electron gas with high mobility of charge carriers (for example, [2, 3]). Ballistic metal nanostructures has become the subject of investigation only recently [4].

It is urgent to investigate ballistic electron transport in monocrystalline metal nanostructures as a new class with electron density five orders of magnitude higher than that of two-dimensional electron gas of semiconductor heterostructures. These structures could become the basis for new electron devices such as a spin transistor [5] with ballistic electron transport, ultrafast ballistic electron devices on the basis of metal-dielectric heterostructures [6]. Besides, high charge carrier density in metals allows to decrease the size of a device down to 10 nm and even less [6–8]. The task arises to carry out epitaxial growth of perfect monocrystalline metal films in which the conductivity electrons have long free paths, in order to prepare nanometre-sized ballistic electron devices on this basis. Possible decrease of lateral size of the nanostructures down to 10 nm puts forward definite demands to the morphology of films. Their mean roughness should be at least an order of magnitude less than the expected lateral sizes.

In the present paper we report the investigations of epitaxial growth of thin (10–100 nm) monocrystalline(100) films of high-melting metals (W, Mo, Nb, Ta) with perfect volume structure, and the morphology of their interface (with the mean roughness amplitude of about 0.1–0.4 nm), as well as structural and

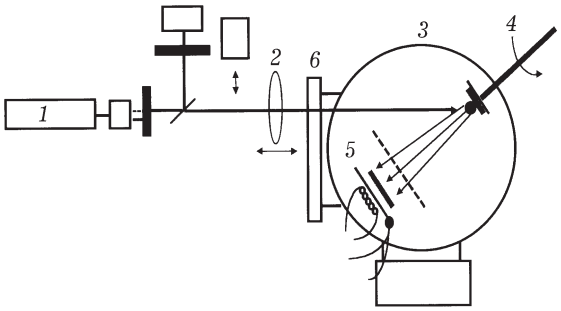

Fig. 1. Growth set-up: *1* – solid-state pulsed laser, *2* – focusing lens (∅ = 0.9 mm), *3* – high-vacuum chamber, *4* – rotating target, *5* – heated table with thermocouple for the samples, *6* – window.

electron transport characteristics. It is demonstrated that, as a result of optimizing the epitaxy process, free paths of electrons in films can be more than an order of magnitude longer than the film thickness.

EPITAXIAL GROWTH OF FILMS

Epitaxial growth of films was carried out in a bulk metal heated ultrahigh vacuum chamber (3) with the basic vacuum of $5 \cdot 10^{-10}$ mbar (Fig. 1). Solid pulsed laser (1) with a wavelength of 1.08 μm and power of 0.2–0.4 J per pulse and pulse duration of 15 ns was focused by a long-focus lens (2) through the sight windows of the chamber onto a rotating target (4) made of high-purity metal. The pulse frequency was 20 Hz. Pulse density of the power at the target was several gigawatts per square centimetre. As a result, pulse vaporization of the target material occurred which resulted in the formation of metal atomic flow directed towards the substrate (5) which was fixed at the temperature table with temperature controlled within the range of 100–800 °C. Metal film grew at a mean rate of about 10 nm/min.

The method of pulsed laser vaporization in ultrahigh vacuum has definite advantages connected with the substantially non-equilibrium character of epitaxial growth: atomic flow is characterized by some effective temperature which is sufficient for the migration of atoms along the surface and building them into the film lattice. The equilibrium temperature of the substrate can be rather low; high density of atomic flow in the pulse leads to high concentration of film growth centres which has a positive effect on the interface morphology. The method allows to conduct vaporization of hardly melting targets. Since mean power released at the target is low (several watts), its equilibrium temperature remains close to room temperature. This does not cause unfavourable processes of vacuum worsening due to the desorption of an impurity.

We used this method for the epitaxial growth of both individual and heteroepitaxial films of high-melting metals (W, Mo, Nb, Ta) on the surface of $(\bar{1}012)$ face of monocrystalline sapphire used as a substrate. It was stated that at substrate temperature above 400 °C the growth of perect monocrystalline (100) films occurs.

STRUCTURAL CHARACTERISTICS OF FILMS

The volume structure of films was characterized by means of X-ray diffraction patterns. Figure 2 shows the results for a (100) tungsten

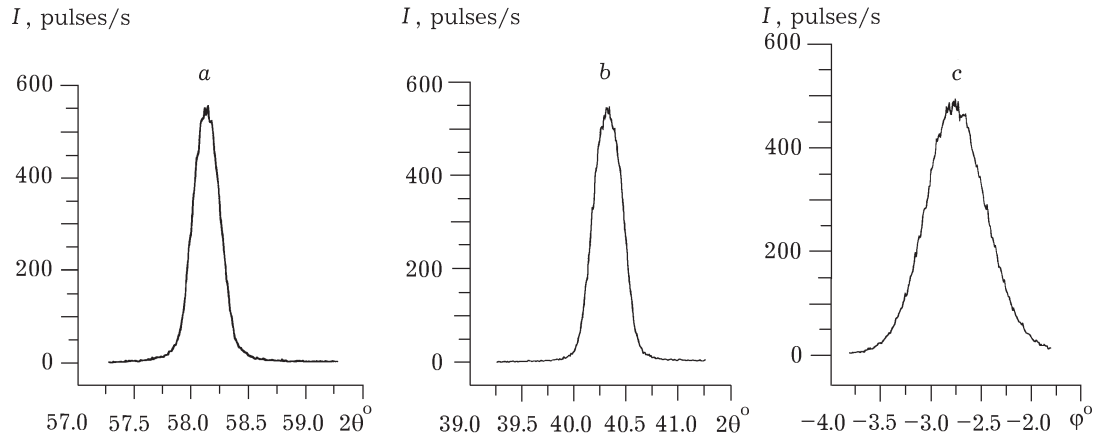

Fig. 2. X-ray diffraction of (100) tungsten films. The data are presented for the symmetrical (200) (*a*) and asymmetrical (110) (*b* and *c*) reflections.

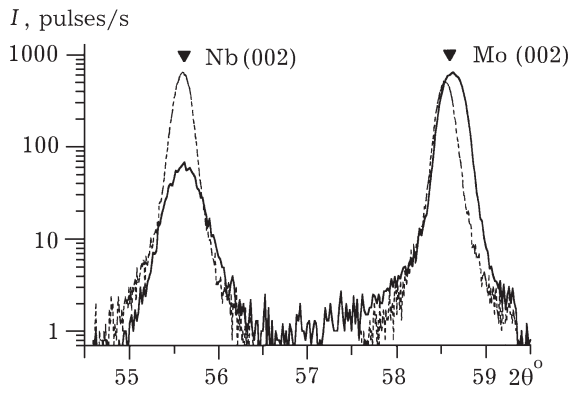

Fig. 3. X-ray diffraction of (100) films of Nb and Mo (dashed line) and heteroepitaxial Mo/Nb film (continuous line) for the (200) reflection. Triangles indicate the position of the reflection from bulk crystal.

film 100 nm thick for the symmetrical (200) and asymmetrical (110) reflections. Half-width of the lines is 0.2, 0.25 and 0.5°, respectively. These data can serve as an estimate of angular disordering of single crystal blocks comprising the tungsten film. Similar results are obtained for individual molybdenum and niobium films, as well as for the heteroepitaxial Mo – Nb film (Fig. 3). The results obtained show that the films possess perfect volume structure with the (100) axis nearly coincident with the perpendicular to the substrate. The comparison of film lattice parameters with the characteristics of the voluminous single crystal (see triangles in Fig. 3) revealed the former and the latter were close to each other.

Structural properties of the films are confirmed by the data on rapid electron diffraction on reflection (Fig. 4). For comparison, the diffraction along the [110] direction is shown for the individual Mo film (100) and for Mo/Nb(100), as well as along [010] for a two-layer film. The reflections are prolonged along the perpendicular to the film surface which is the evidence of two-dimensional diffraction.

SURFACE MORPHOLOGY

The substrates used for the epitaxy of metal films exhibited the mean amplitude of roughness within the range of 0.1–0.2 nm. By optimizing the growth process we succeeded in obtaining the films with the roughness of the outer boundary at the level of 0.2 nm. The morphology of films is substantially dependent on the growth conditions. Both prolonged morphological structures and the structures with no indices of anisotropy along the surface can be observed. Figure 5, *a* shows a typical film surface profile obtained by means of an atomic force microscope (the contrast determined the amplitude of the relief). In order to characterize the film morphology, it is convenient to determine autocorrelation function using experimental data on the surface topography. Figure 5, *b* shows a two-dimensional (x and y are the shifts of the coordinates over the surface) correlation function obtained by means of numerical treatment of the surface profile shown in Fig. 5, *a*. For convenience, the function is presented in a linear scale; the contrast determines the function at a given point. The amplitude of the maximum in the centre is a square mean deviation of the surface profile. One can see that its mean amplitude is about 0.15 nm. The function over the $x - y$ coordinates forms a drawing. This is due to the exhibited different correlation lengths that characterize the morphology. The main maximum in the centre corresponds to small-scale (along the surface) fluctuations of the surface profile with a mean half-width of about 100 nm. This is a mean size of monocrystalline films comprising the film.

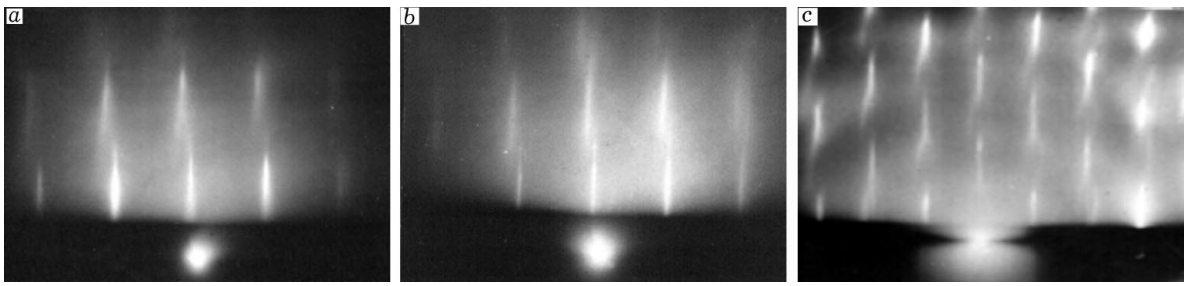

Fig. 4. Diffraction of fast electrons along the [110] direction for the Mo (*a*) and Mo/Nb (*b*) films; *c* – diffraction along the [010] direction for the Mo/Nb film.

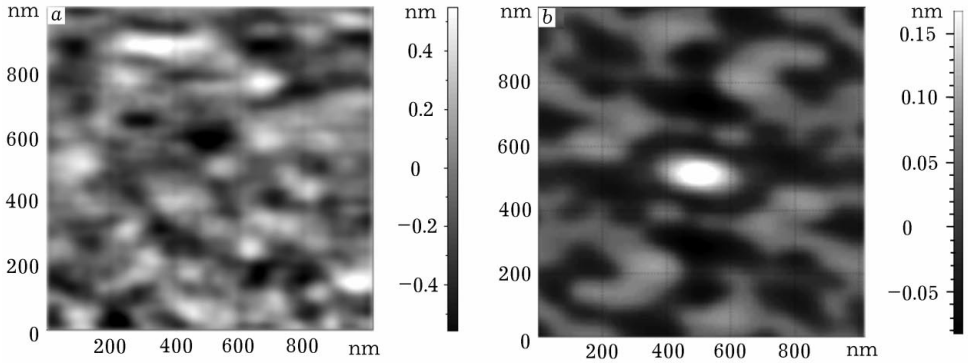

Fig. 5. The surface profile of Mo (100) film recorded with the help of an atomic force microscope (*a*); a two-dimensional autocorrelation function (*b*) obtained by means of numerical processing of *a* (for convenience, it is shown in linear scale over *z*-axis).

ELECTRON TRANSPORT STUDIES

Temperature dependencies of specific electrical resistance of monocrystalline films (of high-melting metals) 10–100 nm thick within the temperature range of 4.2–295 K were studied. At room temperature the specific resistance of films well coincides with that of bulk monocrystals. When temperature is decreased the dependence of specific resistance is determined by electron-phonon interaction till low temperatures when the dependence gets saturated and becomes determined by the scattering of the electrons at static defects. For Nb and Ta the temperatures of superconducting transition are close to those of bulk crystals at film densities above 20 nm.

It was found that the scattering of electrons becomes substantial at the film interfaces. Because of this, quasilinear dimensional dependence on film thickness (d) within the range of 30–100 nm is observed. In metals the perfection of films is characterized by the ratio of resistance at room temperature to that at liquid helium temperature (RRR) (for superconductors, resistance was determined above the point of superconducting transition). This ratio is approximately equal to the residual free path length divided by free path length at room temperature. Because of this, it is convenient to determine experimentally the ratio RRR/d in order to characterize the perfection of the film with respect to both the bulk structure and the interface for the given film material. This dependence as the function of the thickness of grown films is shown in Fig. 6 for W, Mo, Ta and Nb. The analysis of literature data shows that the values obtained in the present study are somewhat record-breaking. The estimated residual free path of electrons, for example in tungsten or molybdenum, exceeds the thickness of the films under investigation by an order of magnitude.

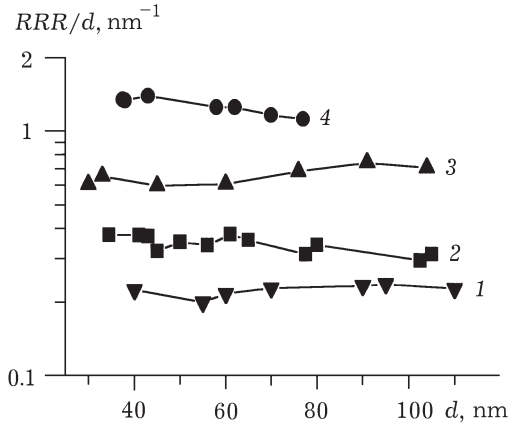

Fig. 6. The dependence of RRR/d on film thickness: W (*1*), Mo (*2*), Ta (*3*), and Nb (*4*). RRR is determined as the ratio of film resistance at room temperature to that at liquid helium temperature.

CONCLUSION

Monocrystalline (100) films of high-melting metals obtained by means of pulsed laser vaporization in ultrahigh vacuum possess extremely long residual free path of electrons. This result is achieved by optimizing the growth process when perfect bulk structure of the film is combined with small roughness amplitude at the interfaces which minimizes electron scattering both in volume and at the interfaces. This optimization is possible under sharply nonequilibrium film growth conditions provided by

pulsed laser vaporization technique. The energy of atomic flow is sufficient for epitaxy in short-range order with the formation of monocrystalline blocks with a size of about 100 nm at rather low substrate temperature. Due to epitaxial growth, monocrystalline blocks are weakly disoriented with respect to each other. High density of atoms in the flow within the pulse and low substrate temperature lead to improved morphology of the film surface.

The results obtained open the possibility to use the studied films of high-melting metals as the basis for the production of nanometre-sized ballistic electron devices.

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