RHEED and AES Study of Metal - Insulator - Metal Thin Film Systems: Nb-Al₂O₃-Nb

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The layered Nb-Al₂O₃-Nb structure was investigated by Reflection High Energy Electron Diffraction (RHEED). Several nanometers thick γ -Al₂O₃ epitaxial layers were prepared by molecular beam epitaxy on Nb(111) and Nb(211) substrates. Alumina was deposited by vacuum evaporation method using a graphite Knudsen cell heated by electron bombardment. Auger spectroscopy analysis showed a good stoichiometry of the γ -Al₂O₃ films.

The Nb(111) $|Al_2O_3(210)|$ Nb(111) and Nb(211) $|Al_2O_3(110)|$ Nb(211) epitaxial systems were obtained by consecutive molecular beam deposition of Nb on epitaxial γ -Al₂O₃/Nb layers. These systems are very promising for the fabrication of very high quality Josephson tunnel junctions.

1. Introduction

Niobium-Al₂O₃ is an often investigated metal - ceramic interface due to its excellent thermal stability and use in different superconducting applications such Josephson junctions. It has been shown that the structural properties of Nb and alumina films determine behavior of multilayer Nb-AlO,-Nb devices. For this reason during the last ten years the epitaxial growth of niobium electron-beam Al_2O_3 using evaporation techniques was investigated [1-6] in order to understand their atomic structure and that of the interface.

Large attention has been paid to the preparation of tunnel oxide barriers – vital part of the tunnel junction. The oxide layer was prepared generally by oxidation of Al films 2-7 nm thick, resulting in formation of an AlO_x layer [7,8]. Its stoichiometry, continuity and thickness have been found to be the most important parameters.

Regarding to the above mentioned works there was an exciting idea to prepare fully single-crystal trilayer Nb-Al₂O₃-Nb structures with well stoichiometric oxide tunnel barrier. The epitaxial growth of Nb on alumina was relatively easily made possible as soon as appropriate evaporation sources had been available [1-6], however the key problem to prepare an epitaxial alumina layer remained. For this reason we started to investigate the growth of alumina layers on orientated Nb

substrates and vice versa in order to be able to prepare heteroepitaxial M-I-M structures by depositing successively the aluminum oxide on Nb crystal and the Nb film on the oxide.

The crucial step of this study was to develop reliable sources of Al₂O₃ and Nb atomic flux required for the epitaxial growth. Unlike in the Al oxidation methods, our evaporation source permitted the direct deposition of alumina. This source provided us with amorphous well stoichiometric layers in the case of deposition on a polycrystalline substrate at room temperature [9]. During the Nb source construction it was necessary to overcome some technical difficulties due to the high evaporation temperature of Nb and its relatively low vapor pressure at temperatures above the melting point. This NbEBES cell (Niobium Electron Beam Evaporation Source) was used successfully in our previous study of Nb epitaxial growth on the sapphire (0001) surface [6].

The crystallographic structure of the deposits was checked by reflection high energy electron diffraction (RHEED) during the growth and temperature annealing, purity and stoichiometry of the layers were checked by AES.

2. Experimental

The studies reported in this paper were performed in a specially designed UHV system. Alumina was deposited by means of its evaporation from a graphite crucible heated by electron bombardment. The NbEBES evaporation cell was operated on the principle of direct electron bombardment of a water cooled Nb tip. Both sources, alumina and Nb cell, provided molecular beam sufficient for the epitaxial growth without contamination by metals used for the source construction.

The UHV chamber was equipped with RHEED facility permitting simultaneous observation of the substrate surface during the metal deposition. The RHEED instrument was operated at the acceleration voltage of 40 kV. The diffraction pattern was recorded by a RHEED-Vision Computer System consisting of a CCD camera, video recorder and a computer. It performed both image processing and a real time analysis of the diffraction pattern. A movable sample holder made it possible to change the polar as well as the azimuthal angle of incidence of electrons onto the sample surface. The evaporation sources were surrounded with a liquid nitrogen cooled cryo-panel. The chamber was also equipped with a fast entry air lock for sample exchange.

Nb and alumina were deposited at a background pressure of about $3x10^{-6}$ Pa, at different substrate temperatures. Nb single-crystalline substrates were cleaned by cycles of Ar ion bombardment at low ion energy (500 eV, 15 mA cm⁻², 30 min) and annealing at 900 K (30 min) till a clean and well ordered surface was obtained.

The AES spectra were taken at the primary energy of 2000 eV in the direct (i.e. non derivative) mode using the Staib Ins DPCMA analyzer. Stoichiometry of alumina was checked by comparing the peak intensities of $L_{23}VV$ transitions of Al^0 at 64 eV and $L_{23}VV$ transitions given by Al^{3+} species, shifted from the ideal value of 64 eV to ~ 50 eV.

3. Results and discussion

The results of the study of the Nb-Al₂O₃-Nb multilayer system prepared on Nb(111) were published elsewhere [10]. In this paper we present the results concerning of Nb(211) substrate.

Aluminum oxide growth

The substrate surface quality was checked by the RHEED method. An example of the RHEED diffraction pattern corresponding to the Nb(211) surface with the [110] surface direction parallel to the primary electron beam is represented in Fig. 1. a. The diffraction pattern corresponds to the (110) reciprocal lattice plane having rhomboedric symmetry typical for BCC metals. Pronounce elongation of diffraction spots showed high quality substrate surface. In the [111] direction the well-known 1 x 2 surface reconstruction was observed. Similar reconstruction was found for Mo(211) surface [11] and also for the (110) FCC metal surfaces having analogous surface geometry (Au, Cu, Ir ...) [12,13,14].

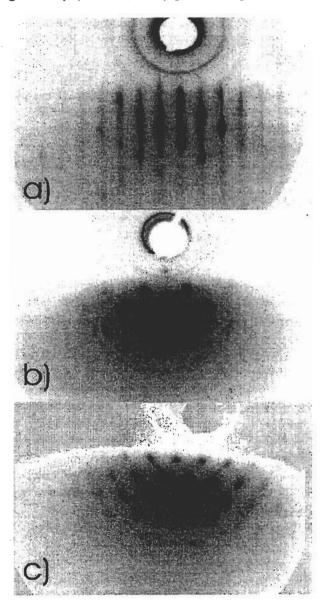


Figure 1: RHEED diffraction patterns. a) Nb(211) substrate, Nb(110) reciprocal lattice plane, b) γ -Al₂O₃/Nb(211) layer, γ -Al₂O₃(100) reciprocal lattice plane, c) Nb/ γ -Al₂O₃/Nb(211) layer, Nb(111) reciprocal lattice plane.

Alumina was deposited at the substrate temperature 950 K. AES analysis of the alumina layers gave the same results for both Nb(111) and Nb(211) substrates, respectively [10]. The obtained thin layers exhibited oxide character and zero signal of the Nb substrate indicated their continuity. The RHEED diffraction pattern of the alumina deposit on the Nb(211) substrate is presented in fig. 1.b. The diffraction pattern taken at the incident electron beam parallel to the Nb[110] surface direction was identified with the cubic γ -Al₂O₃ spinel structure of the following epitaxial parameters:

$$Al_2O_3(110) \parallel Nb(211)$$
 and $Al_2O_3[100] \parallel Nb[110]$

The spot-like character of the diffraction spots showed certain surface roughness however complete disappearance of the Nb substrate pattern confirmed the AES analysis results showing the continuous character of the oxide layer.

In the case of the Nb(111) substrate a well ordered alumina layer was obtained after recrystalization of the deposit at a high temperature [10].

Nb film growth

The upper Nb layer of Nb-Al₂O₃-Nb systems were prepared at the substrate temperature of 830 K. Similar AES analysis results were obtained for both Nb(111) [10] and Nb(211) substrates. The AES spectra indicated presence of the Nb peaks and complete disappearance of the Al³⁺ and O signal confirming the continuous character of the Nb deposit.

The RHEED diffraction pattern observed after the Nb deposition on Al₂O₃-Nb(211) is presented in fig. 1.c. The pattern corresponds to the (111) Nb reciprocal plane. There are no diffraction spots corresponding to the Al₂O₃ layer. It is in agreement with the results of the AES analysis mentioned earlier. In the [110] Nb direction the diffraction pattern indicated a presence of two populations of crystal grains having the same (211) epitaxial plane and rotated by 180° with respect to each other. It permits of twinned crystals to arise during the layer growth. In the case of the Nb(111) substrate presence of twinned crystals has not been observed.

The epitaxial parameters of the complete Nb-Al₂O₃-Nb systems have been found as follows:

where suffixes c and f stand for the crystal and the film, respectively. The circular form of the diffraction spots in Fig. 1.c indicated a rough Nb surface. This roughness was partially induced by roughness of the alumina layer support.

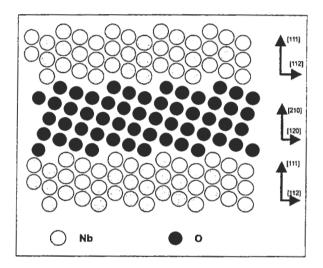


Figure 2: Hard ball model of the $Nb/Al_2O_3/Nb(111)$ multilayer systém.

The ball models for both Nb-Al₂O₃-Nb(111) and Nb-Al₂O₃-Nb(211) systems in the cut perpendicular to the substrate surface are presented in Fig. 2 and 3, respectively. The relatively good accommodation of Nb and γ -Al₂O₃ crystal lattices in the direction parallel to the interface may be seen. It can be expressed also by means of accommodation parameters which are equal to (it is defined as a ratio of the atom distances between the O and Nb atoms in the Nb and γ -Al₂O₃ crystal lattices in the corresponding directions):

1.
$$p_{Nb(111)} = d_{O[120]} / d_{Nb[112]} = 1.09$$

2. $p_{Nb(211)} = d_{O[110]} / d_{Nb[111]} = 0.98$

One can conclude that the growth is probably driven by good accommodation along the main

crystallographical directions of the Nb and γ -Al₂O₃ crystal lattices.

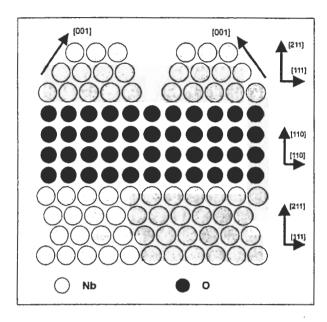


Figure 3: Hard ball model of the Nb/Al₂O₃/Nb(211) multilayer systém.

The presented results show that the aluminum oxide layers can be prepared by means of molecular beam epitaxy and that it is possible to obtain fully epitaxial Nb-Al₂O₃-Nb multilayer systems. The actual epitaxial characteristics depend on the deposition conditions and the chosen substrate structure. We have observed different alumina epitaxial structures also on Nb(110) and Nb(211). These results will be published in the near future.

The Nb growth on the γ -Al₂O₃ (210) and (110) surfaces represents a new result that confirmed the already known fact of a good heteroepitaxy of Nb on alumina.

In future investigations detail characterization of the morphological parameters of prepared single-crystalline structures, specially thickness and roughness, will be necessary.

4. Summary

Alumina layers were grown on the (111) and (211) Nb single-crystals substrates by means of molecular beam epitaxy. The layer structure and composition were investigated in situ by Reflection High Energy Electron Diffraction (RHEED) and Auger Electron Spectroscopy (AES). It was shown that well stoichiometric alumina layers grew in the γ modification. The

γ-Al₂O₃(210) // Nb(111) and γ-Al₂O₃(110) // Nb(211) epitaxial orientations were found. Consecutive molecular beam deposition of Nb on epitaxial γ-Al₂O₃/Nb layer permitted to obtain a fully single-crystalline Nb-Al₂O₃-Nb system that might be favorable for fabrication of high quality Josephson tunnel junctions. The well-defined epitaxial parameters of the Nb-Al₂O₃-Nb multilayer systems can be explained by means of good accommodation along the main crystallographic directions between the Nb and γ-Al₂O₃ crystal lattices.

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