

# RHEED and AES Study of Metal - Insulator - Metal Thin Film Systems: Nb-Al<sub>2</sub>O<sub>3</sub>-Nb

K. Mašek, R. Kapsa, B. Gruzza\* and V. Matolín<sup>1</sup>

*Department of Electronics and Vacuum Physics, Charles University, V Holesovickach 2, 180 00 Prague 8, Czech Republic*

*\* LASMEA, Universite Blaise Pascal, Les Cezeaux, 63177 Aubiere, France*

*<sup>1</sup> matolin@mbox.troja.mff.cuni.cz*

(Received October 5 1998; accepted January 11 1999)

The layered Nb-Al<sub>2</sub>O<sub>3</sub>-Nb structure was investigated by Reflection High Energy Electron Diffraction (RHEED). Several nanometers thick  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> 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  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> films.

The Nb(111)|Al<sub>2</sub>O<sub>3</sub>(210)|Nb(111) and Nb(211)|Al<sub>2</sub>O<sub>3</sub>(110)|Nb(211) epitaxial systems were obtained by consecutive molecular beam deposition of Nb on epitaxial  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/Nb layers. These systems are very promising for the fabrication of very high quality Josephson tunnel junctions.

## 1. Introduction

Niobium-Al<sub>2</sub>O<sub>3</sub> is an often investigated metal - ceramic interface due to its excellent thermal stability and its use in different superconducting applications such as Josephson junctions. It has been shown that the structural properties of Nb and alumina films determine behavior of multilayer Nb-AlO<sub>x</sub>-Nb devices. For this reason during the last ten years the epitaxial growth of niobium films on Al<sub>2</sub>O<sub>3</sub> using electron-beam 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<sub>x</sub> 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-Al<sub>2</sub>O<sub>3</sub>-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 Al<sub>2</sub>O<sub>3</sub> 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  $3 \times 10^{-6}$  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<sup>-2</sup>, 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<sub>23</sub>VV transitions of Al<sup>0</sup> at 64 eV and L<sub>23</sub>VV transitions given by Al<sup>3+</sup> species, shifted from the ideal value of 64 eV to ~ 50 eV.

### 3. Results and discussion

The results of the study of the Nb-Al<sub>2</sub>O<sub>3</sub>-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. Pronounced 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].

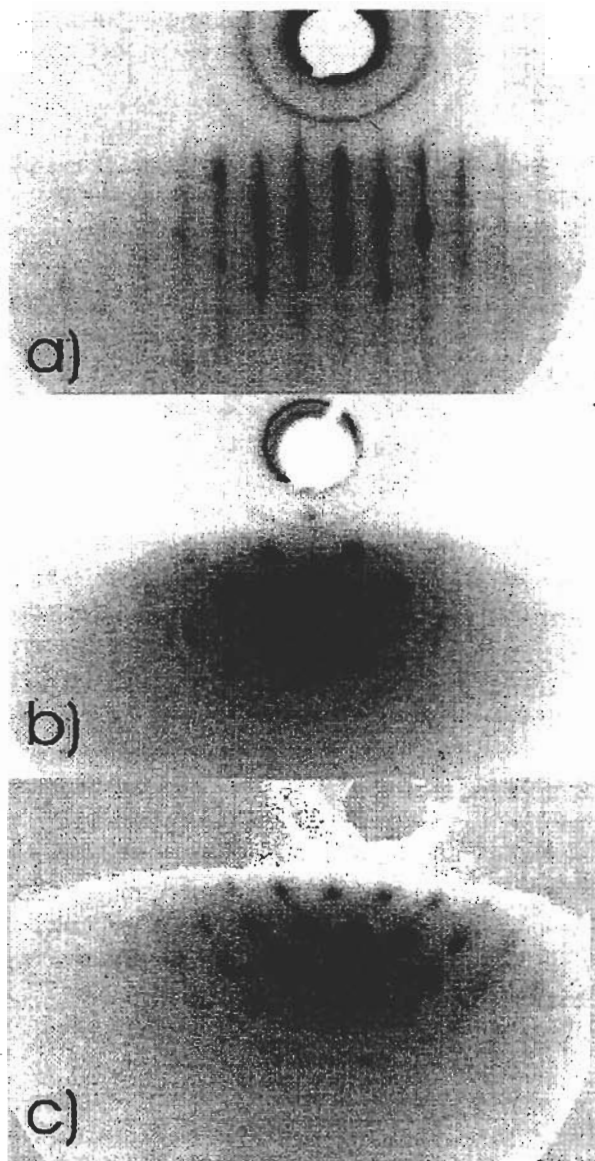


Figure 1: RHEED diffraction patterns. a) Nb(211) substrate, Nb(110) reciprocal lattice plane, b)  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/Nb(211) layer,  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>(100) reciprocal lattice plane, c) Nb/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/Nb(211) layer, Nb(111) reciprocal lattice plane.



crystallographical directions of the Nb and  $\gamma$ - $\text{Al}_2\text{O}_3$  crystal lattices.

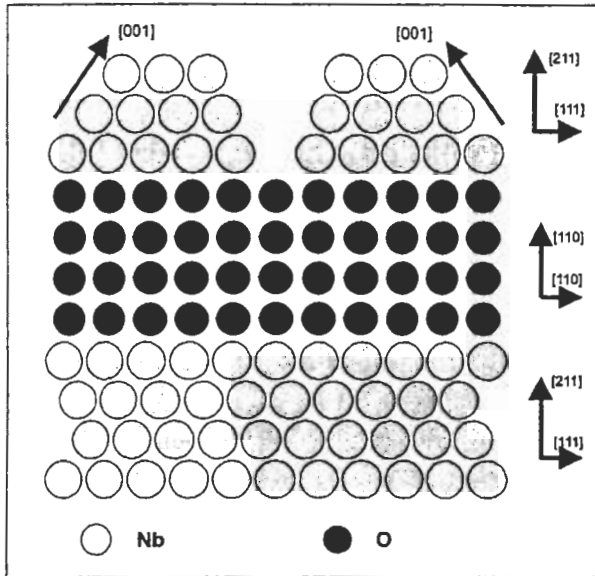


Figure 3: Hard ball model of the Nb/ $\text{Al}_2\text{O}_3$ /Nb(211) multilayer system.

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- $\text{Al}_2\text{O}_3$ -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  $\gamma$ - $\text{Al}_2\text{O}_3$  (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  $\gamma$  modification. The

$\gamma$ - $\text{Al}_2\text{O}_3$ (210) // Nb(111) and  $\gamma$ - $\text{Al}_2\text{O}_3$ (110) // Nb(211) epitaxial orientations were found.

Consecutive molecular beam deposition of Nb on epitaxial  $\gamma$ - $\text{Al}_2\text{O}_3$ /Nb layer permitted to obtain a fully single-crystalline Nb- $\text{Al}_2\text{O}_3$ -Nb system that might be favorable for fabrication of high quality Josephson tunnel junctions. The well-defined epitaxial parameters of the Nb- $\text{Al}_2\text{O}_3$ -Nb multilayer systems can be explained by means of good accommodation along the main crystallographic directions between the Nb and  $\gamma$ - $\text{Al}_2\text{O}_3$  crystal lattices.

#### Acknowledgment

This work was supported by the Grant No. 202/97/P077 of Czech Grant Agency.

#### References

- [1] J. Mayer, C.P. Flynn and M. Rühle, *Ultramicroscopy* 33 (1990) 51
- [2] J. Mayer, G. Gutenkunst, G. Möbus, J. Dura, C.P. Flynn and M. Rühle, *Acta Metall. Mater.* 40 (1992) S217
- [3] A. Gibaud, D.F. McMorrow and P.P. Swaddling, *J. Phys.: Condens. Matter* 7 (1995) 2645
- [4] C.P. Flynn and S. Yadavalli, *Acta Metall. Mater.* 40 (1992) S45
- [5] K. Masek and V. Matolin, *Thin Sol. Films*, 1996, in press
- [6] Gin-ichiro Oya, Masanori Koishi and Yasuji Sawada, *J. Appl. Phys.*, 60 (1986) 1440
- [7] R. Dolata, M. Neuhaus and W. Jutzi, *Physica C*, 241 (1995) 25
- [8] L. Capogna, G. Costabile, P. Barbara, N. Martucciello and R. Monaco, *Cryogenics*, 34 (1994) 895
- [9] R. Kapsa, I. Stara, D. Zeze, B. Gruzza and V. Matolin, *Thin Sol. Films*, 1996, in press
- [10] K. Masek, R. Kapsa, V. Matolin, *Surf. Sci.*, 1998, in press
- [11] G. P. Lopinski, J. A. Prybyla and P. J. Estrup, *Surf. Sci.* 296 (1993) 9
- [12] S. Titmuss, A. Wander, and D. A. King, *Chem. Rev.* 96 (1996) 1291
- [13] S. Speller, J. Bomermann, S. Molitor, T. Rauch, W. Heiland, *Surf. Sci.* 331-333 (1995) 1070
- [14] R. Koch, M. Borbonus, O. Haase, and K. H. Rieder, *Phys. Rev. Lett.* 67 (1991) 3416