SUBLAYER THICKNESS DEPENDENCE OF THE OPTICAL PROPERTIES OF Ni/Ti AND Fe/Zr MULTILAYERS

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ABSTRACT

The study of the thickness dependence of the electron energy structure of Fe, Ni, Ti and Zr sublayers in Ni/Ti and Fe/Zr multilayers by using the experimental and computer simulated optical spectroscopy has been performed. A series of Ni/Ti and Fe/Zr multilayered films (MLF) with a bilayer period of 0.5 - 30 nm and constant (Ni/Ti) / different (Fe/Zr) sublayer thickness ratios were prepared by using computer-controlled double-pair target face-to-face sputtering onto a glass substrate at room temperature (RT). Computer simulation of the resulting optical properties of these MLF was carried out by solving of multireflection problem with a matrix method, assuming either "sharp" interfaces resulting in rectangular depth profiles of the components or "mixed" (alloy-like) interfaces of variable thickness between pure-metal sublayers. Optical constants of pure bulk metals as well as equiatomic alloy interfaces were employed in these simulations. It was shown that the difference between experimental and simulated optical properties of the investigated MLF increases with decrease in sublayer thickness. This result allows to conclude that the electronic structures of sublayers below 4 - 5 nm thickness in MLF differ from the corresponding bulk metals.

1. INTRODUCTION

The study on the properties of ultrathin magnetic films and artificial metal superlattices are useful in testing the limits of band theory, exploring new phenomena, and establishing a technology to control the electronic properties of materials. For these purposes, sensitive techniques must be employed. It is known that the optical spectroscopy of metals is a rather sensitive tool for studying their electronic structure and may be used for a registration of tiny changes in the crystalline structure of metals, which sometimes can not be detected by usual x-ray study. The authenticity of our knowledge about real crystalline structure of the MLF and/or their electron energy structure may be verified by the comparison of the experimental and computer simulated optical data, based on different models for the MLF. Their coincidence suggests that the real crystalline structure and optical properties of the MLF are approximated by the model.

Ni/Ti and Fe/Zr MLF are the typical examples of the 3d transition metal based MLF which exhibit interesting physical properties. For example, Ni/Ti multilayers are of interest

for special applications like neutron optics [1]. Both Ni/Ti and Fe/Zr MLF show a tendency for amorphization through a solid state reaction [2,3] and can be considered as model objects for the investigation of this process.

In this article we present the experimental and computer simulated study of the sublayer thickness dependence of the optical properties of the Ni/Ti and Fe/Zr MLF, aiming at the investigation on the thickness dependence of the electron energy structure of the sublayers in the MLF.

2. EXPERIMENT

A series of the Ni/Ti MLF with a bilayer period of 0.5 - 30 nm, a constant sublayer thickness ratio (1:1) and a total thickness of 150 nm as well as a wedge-shape Fe/Zr MLF were prepared by using computer-controlled double-pair target face-to-face sputtering onto glass substrates at RT. All the investigated Ni/Ti MLF have a Ti sublayer at the top. In wedge-shape MLF the Zr sublayers were kept at a nearly constant thickness ($h_{Zr} \sim 1.8$ nm), while h_{Fe} varied from 1.4 to 4.2 nm. The number of bilayers was 15. Details of the sample preparation can be found elsewhere [3]. The sublayer thicknesses in the MLF were estimated by x-ray fluorescence analysis. The x-ray fluorescence data were additionally confirmed by low-angle x-ray diffraction. The crystalline structures of the Ni/Ti and Fe/Zr MLF were analyzed by x-ray diffraction.

The optical properties of the Ni/Ti and Fe/Zr MLF were investigated both experimentally and theoretically. The theoretical simulations were carried out by solving exactly a multireflection problem with a matrix method, assuming either "sharp" interfaces resulting in rectangular depth profiles of the components or "mixed" (alloy-like) interfaces of variable thickness between pure-metal sublayers. Optical parameters of pure metals (bulk Ni and Ti [4,5], and Fe and Zr films of 150 nm thickness) as well as an amorphous FeZr alloy [6] were employed in these simulations.

The experimental study of the optical properties of the Ni/Ti and Fe/Zr MLF have been carried out at RT by using an automatical spectroscopic ellipsometer (I.S.A. Jobin Yvon UVISEL) in an energy range of 1.4 - 4.5 eV. The optical properties of the Fe/Zr MLF were investigated at 5 spots along the wedge with mean Fe sublayer thicknesses of 1.4, 2.1, 2.8, 3.7 and 4.2 nm.

3. RESULTS AND DISCUSSION

According to the results of x-ray diffraction study for the Ni/Ti MLF, the crystalline structures typical for bulk Ni and Ti are conserved in case of the sublayer thickness above 5 nm. For $h_{Ni,Ti}$ below 2.5 nm the measurements reveal an appearance of a highly disordered or amorphous structure in the Ni/Ti MLF (see Fig. 1(a)). It should also be mentioned that x-ray diffraction study of the "thicker" side of the wedge-shape Fe/Zr MLF also reveals typical bulk *bcc* Fe and *hcp* Zr structures, on the other hand, for the "thin" side of the film only Zr peak has been detected. The layered structures of the investigated Fe/Zr and Ni/Ti MLF was confirmed with the low-angle diffraction data (see, for example, Fig. 1(b)).

The computer simulated and experimental optical OC spectra for the Fe/Zr MLF are shown in Fig. 2. It is seen in Fig. 2(a) that the OC of Fe exhibits a pronounced interband absorption peak near 2.3 - 2.4 eV, while the OC spectrum of Zr has only tiny peculiarity near 1.9 eV. The nature of these absorption peaks is well known by *ab initio* band structure and optical property calculations. For example, the interband absorption peak of Fe at 2.4 eV

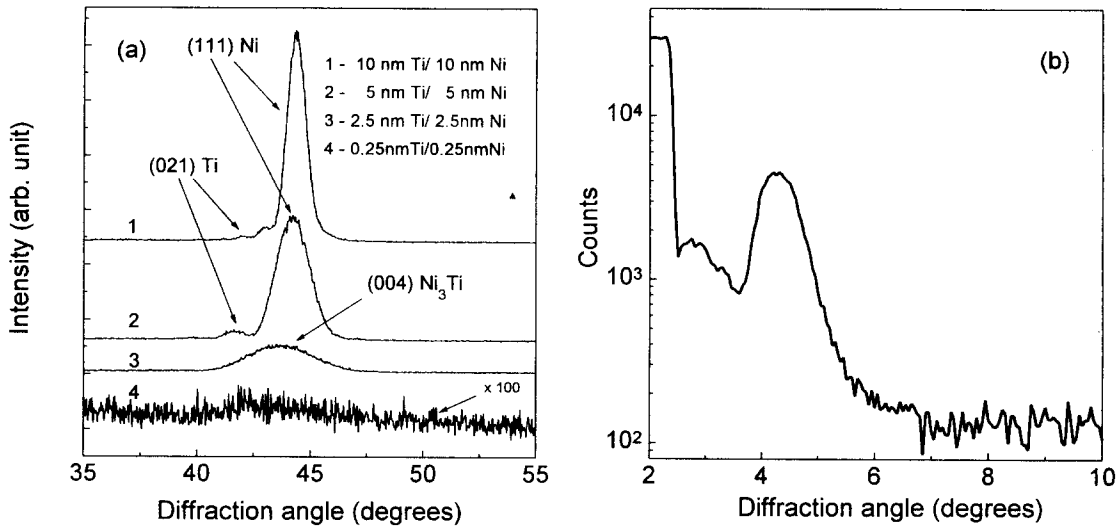

Fig. 1. (a) x-ray diffraction patterns of the Ni/Ti MLF with sublayer thicknesses of 10, 5, 2.5 and 0.25 nm and (b) low-angle diffraction patterns for "thick" side of the Fe/Zr MLF, where the film structure is nearly 15 x (3.5 nm Fe/ 1.9 nm Zr).

arises from the electron transitions in a spin-down subband between nearly parallel *d*-like bands mainly in *H* - *P* - *N* high symmetry directions [7]. It is seen that the resulting OC spectra for the Fe/Zr MLF predicted by the simulations either for a sharp or for a mixed interfaces of 1 nm thickness exhibit an absorption peak near 2 eV of Fe in origin. It should be emphasized that even though the x-ray diffraction data confirm the existence of *bcc* Fe in "thick" side of the Fe/Zr MLF, the tiny traces of the Fe absorption peak in the experimental OC spectra can be observed only for the wedge-shape Fe/Zr MLF with the Fe sublayers of 4.2 nm thickness (see Fig. 2(b)). For a parts of the wedge-shape Fe/Zr MLF with a thinner Fe

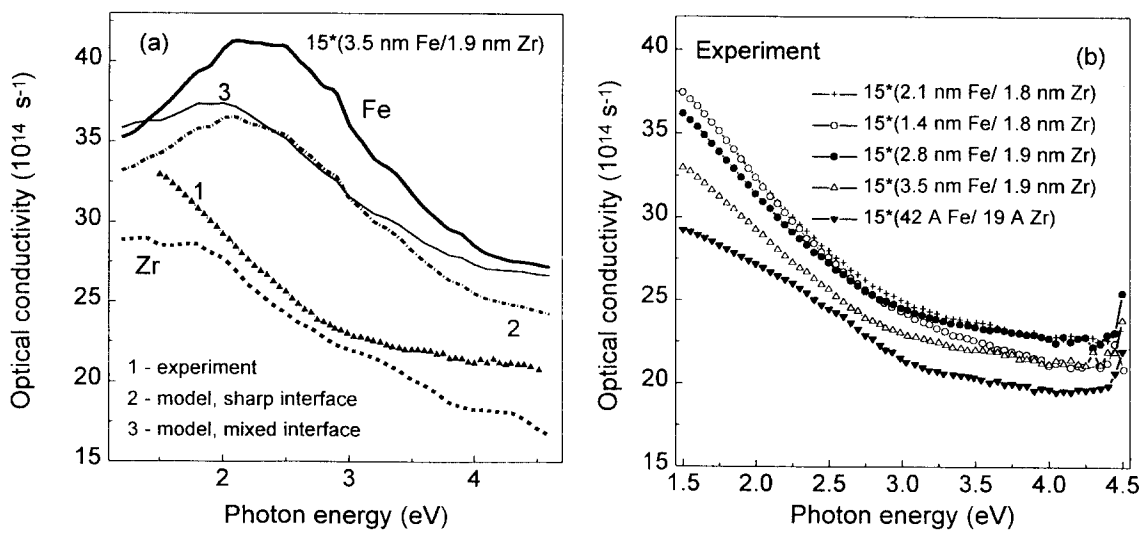

Fig. 2. (a) Simulated and (b) experimental OC spectra for the Fe/Zr MLF.

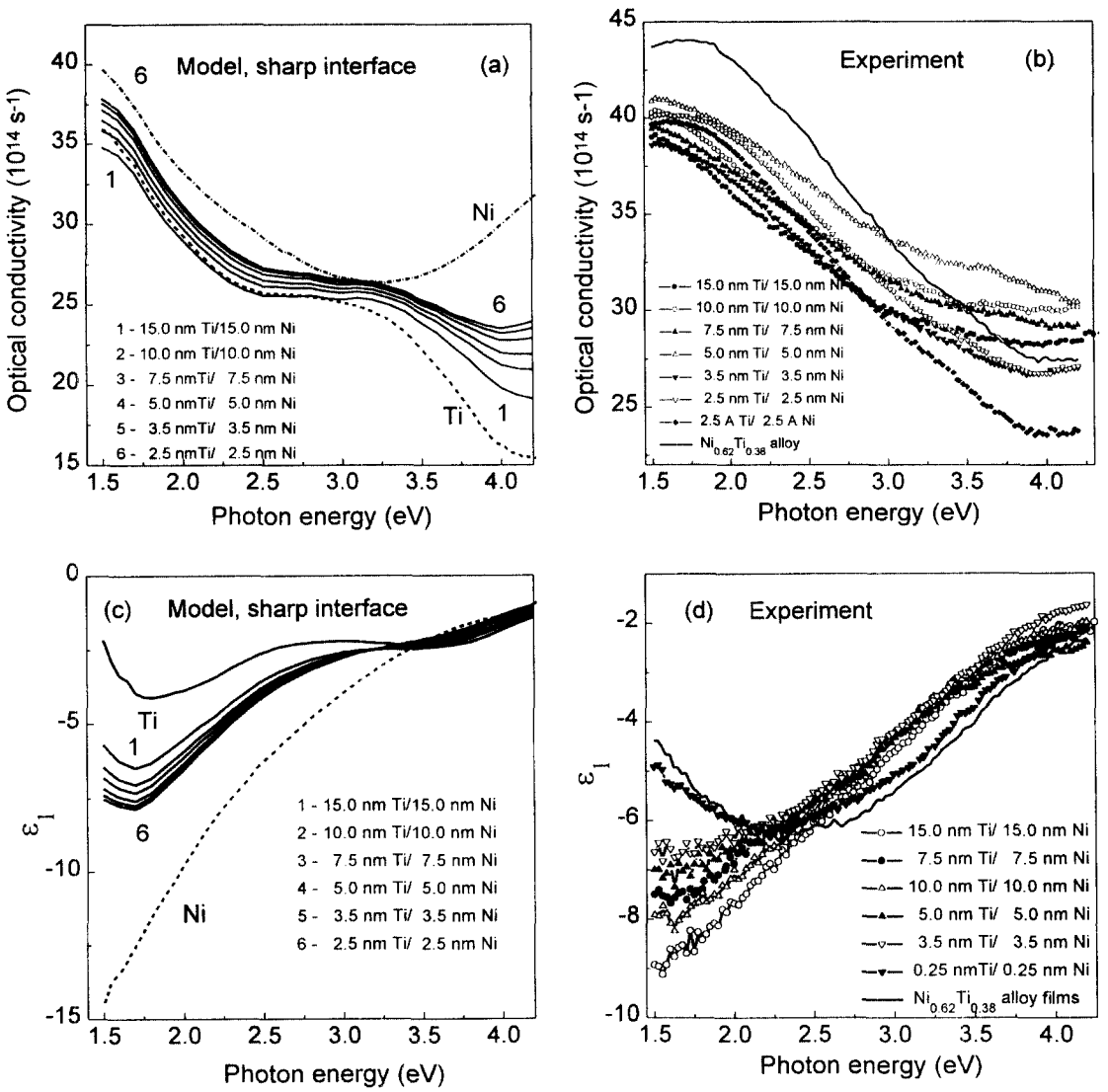

Fig. 3. (a,c) Simulated and (b,d) experimental (a,b) OC and (c,d) ε_1 spectra for the Ni/Ti MLF.

sublayers any manifestation of the this peak can be found. This result allows to conclude that below of 4.2 nm thickness the electronic structure of Fe sublayers in the Fe/Zr MLF differs from the bulk one.

Figures 3(a) and 3(c) present the simulated optical properties of the Ni/Ti MLF. It is clearly seen that the resulting properties of the Ni/Ti MLF manifest the features of both constituent metals. It is also seen, that the model spectra based on bulk optical properties of constituent metals show an insignificant thickness dependence. But the experimental optical properties of all the investigated Ni/Ti MLF differ from predictions of the simulation. First of all, it is necessary to emphasize that both experimental OC and ε_1 do not exhibit the shoulder in the visible range of the spectra. The decrease in sublayer thickness in the Ni/Ti MLF leads to a gradual change of both experimental spectra mainly in the near IR region: the thinner the

nominal thickness of the sublayers, the lower the magnitude of ε_1, and the more clear an absorption peak near 1.75 eV in the OC spectra is appeared. On the basis of x-ray diffraction data for the Ni/Ti MLF and results of the experimental study of the optical properties for bulk Ni-Ti alloy [8] it can be supposed that these changes are caused by solid state amorphization of the MLF during the film deposition. Indeed, the chosen sublayer ratio (1:1) leads to the nearly equiatomic volume composition (62 at. % of Ni and 38 at. % of Ti). According to Rhee *et al.* [8], the OC spectrum for bulk crystalline equiatomic NiTi alloy is characterized by an intense absorption peak at 2.25 eV. The significant difference in the peak location in the OC spectra can be referred to an amorphization of the MLF.

4. CONCLUSIONS

The comparison between the experimental and computer simulated optical properties of the Fe/Zr and Ni/Ti MLF allows to conclude that in the investigated MLF the optical properties of metal sublayers of a few nanometers of thickness (and hence their electronic structures) differ from the bulk ones. The appearance of the interface region with mixed components or amorphization of sublaeyrs near interface region caused by solid state reaction during the deposition can not be considered as an only origin of this discrepancy.

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