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Investigation of Surface Metal Oxide Formed in 10^{-4} Torr Oxygen by Neutron Reflectometry

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Neutron reflectometry is a developing method and is applied for the investigation of the surface distribution of various materials including polymers and metals. In the present study, we investigated the surface distribution of metal oxide formed in 10^{-4} Torr oxygen. The sample was evaporated on float-glass or Si wafer substrates by vacuum evaporation. Since the sample was fabricated by 20 times repetition of evaporation and oxidization, oxidized metal distributes periodically. In the neutron reflectivity curve, Bragg peaks owing to the periodicity of the oxidized metal are obviously found. Analyzing the reflectivity curve gives us the distribution of the oxidized metal.

KEYWORDS: neutron reflectometry, surface metal oxide, Titanium oxide,

§.1. Introduction

It is reported that neutron reflectivity of a Ni/Ti multilayer rises if we introduce oxygen into the vacuum chamber during evaporation of neutron multilayer mirrors.[1),2)] This might be because the oxide suppresses interface roughness or mixing of materials through the interface. In the present study, we intend to investigate the metal oxide at interfaces by neutron reflectometry.

We choose Ti as a target material since 1) Ti is a common material used for multilayer neutron mirrors, and 2) Ti can be oxidized in very low O_2 pressure.[3)]

Surface titanium oxide has been investigated by several researchers.[3-6)] They, however, did not investigate oxidation in very low pressure ($\sim 10^{-5}$ Torr) for a short time (several minutes), which occurs during evaporation process of multilayer mirrors. In addition, they did not take into account the oxide distribution, i.e., they assumed that the surface material was perfectly oxidized to definite thickness and was perfectly un-oxidized beneath the oxide layer.

In the present research, the distribution of the oxide formed at interfaces in multilayers is estimated via neutron reflectivity and it is shown that oxygen penetrates deeply.

§.2. Sample preparation and experimental details

The sample is fabricated on a Si wafer with a vacuum evaporation system.[2)] The sequence of the sample fabrication is: 1) evaporate Ti for 160Å in less than 1×10^{-6} Torr pressure; 2) leave the sample for 120sec.; 3) introduce O_2 up to the pressure of 1×10^{-4} Torr and leave the sample in the oxygen environment for 600sec.; 4) evacuate O_2 till less than 1×10^{-6} Torr for next evaporation. This sequence was repeated twenty times.

In the sample, oxidized layers are periodically distributed, as shown in Fig.1. The oxidized layers cause Bragg reflections in the neutron reflectivity, which enables to estimate the distribution of the oxidized titanium.

It is shown in the figure that adsorbed oxygen is not saturated in 600 sec. However, we choose this oxidation time in order to estimate oxidized Ti during multilayer evaporation.

The neutron reflectivity was measured at E-3 port of Research Reactor Institute of Kyoto University.[2)]

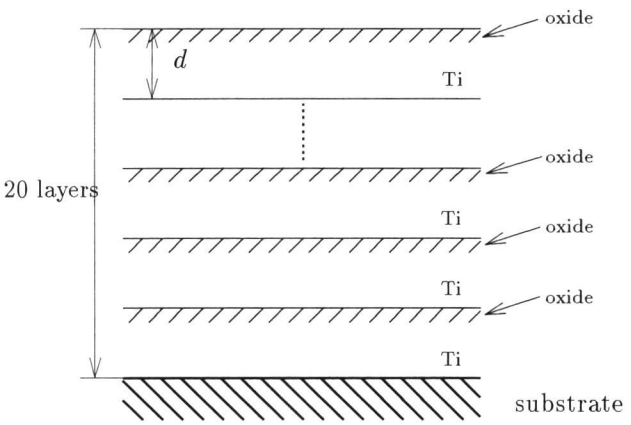

Fig. 1. Schematic view of the oxidized layers distributed periodically in the sample.

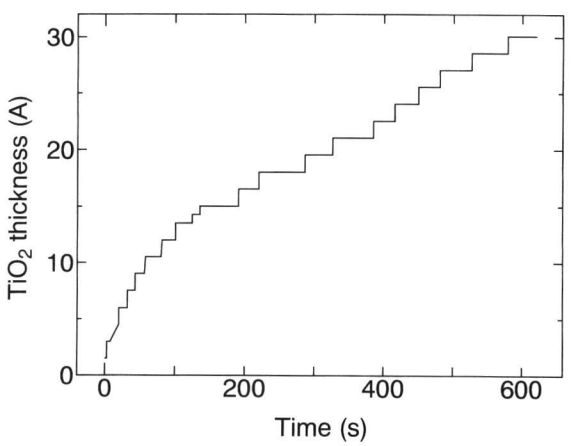

Fig. 2. The estimated thickness of oxidized Ti layer estimated from the indication of quartz crystal thickness monitor.

The amount of absorbed oxygen is estimated through the increase indicated, after evaporation, by the quartz crystal thickness monitor. A typical example is shown in Fig.2.

§.3. Results and discussions

The measured neutron reflectivity of the sample is shown in Fig.3. In the figure, the broken and the solid lines represent the result calculated using the optical potentials shown as the broken and the solid lines in Fig.4,

respectively. In the calculation for the broken line, it is assumed that titanium is per-oxidized for thickness of 12Å. The calculation reproduces well the first Bragg peak but not the second one.

In the calculation for the solid line, 60% of the titanium is oxidized in the 22Å-thick oxide layer; the thickness of the intermediate layer between the oxide and the titanium layer is 38Å. These parameters are chosen to reproduce the experimental result. This result indicates that oxygen penetrates into the titanium layer for more than 60Å, and that the surface titanium is not fully oxidized.

In the present analysis, the optical potential for Ti-oxide is assumed to be symmetrical, since when we assume asymmetric optical potential, χ^2 of the experimental and the calculated data increased.

This method is not suitable for the estimation of surface oxide distribution on the outmost surface layer, since the layer is not oxidized uniformly. However, this method is suitable for interface oxide layer in a multilayer since 1) fabrication of the sample is similar to that of multilayers; 2) surface oxide caused by atmosphere is ineffective for the estimation of internal oxide layers as the latter is estimated through the Bragg peaks.

The same measurements are planned for other materials (Ni, Si, Ge, Fe, etc.) commonly used for multilayer neutron mirrors.

§.4. Conclusions

We estimate distribution of titanium oxide, which is formed at the interfaces of a multilayer, by the neutron reflectometry. The sample is fabricated by the repetition of evaporation-oxidization cycle, simulating the multilayer evaporation process. In this way, titanium oxide is periodically distributed within the sample, and it causes Bragg reflection of neutrons. Distribution of the titanium oxide within one oxide layer can be estimated from the Bragg reflection peaks. In the present study, evaporation thickness for a single cycle is 157Å, and oxygen pressure and time for oxidization is 10^{-4} Torr and 600sec., respectively. Resultant oxide distribution in an oxide layer is trapezoidal, and plateau concentration of oxide is 60%; the plateau thickness is 22Å; and the oblique side thickness is 38Å. This result indicates that oxygen atoms penetration into the titanium is more than 60Å, and that neutron reflectometry is very effective for investigation of the oxide distribution.

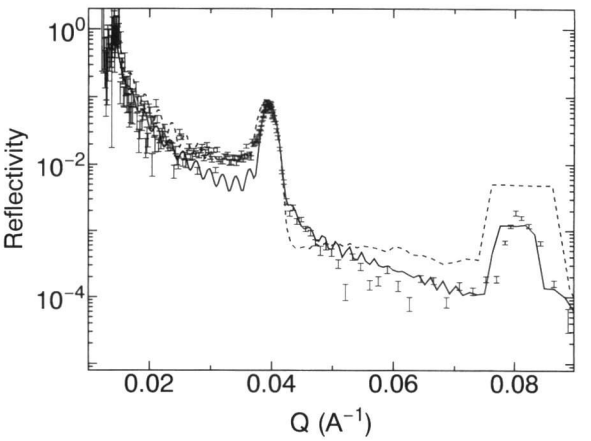

Fig. 3. The neutron reflectivity of titanium oxide sample. The solid and broken lines indicate calculated reflectivity with optical potential shown in Fig.4.

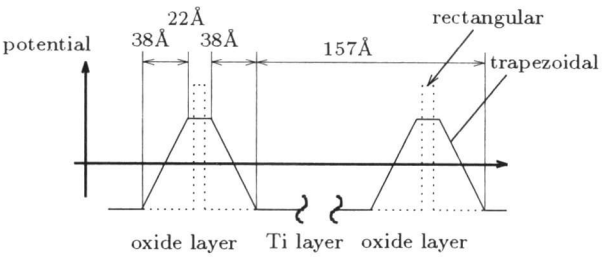

Fig. 4. Schematic representation of optical potentials used in the calculation of Fig.3. The broken and solid lines represent square well and deformed trapezoidal potential.

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