Enhancement of Reflectivity of Short D-Spacing Multilayer Mirrors by Ion Polishing in Combination with Ion Beam Sputtering Deposition

Kazuhiko SOYAMA
Japan Atomic Energy Research Institute
Tokai, Naka, Ibaraki, 319-11 JAPAN

Wakana ISHIYAMA and Katsuhiko MURAKAMI
Nikon Corporation
Nishi-ohi, Shinagawa, Tokyo 140 JAPAN

ABSTRACT

The reflectivity of multilayer mirrors depend on the interface roughness created during deposition of the layers. We have investigated the influence of argon ion bombardment after deposition by ion beam sputtering of nickel or titanium on the interface roughness of an Ni/Ti multilayer. Roughness changes have been measured by X-ray and neutron reflection. The process is applied to Ni/Ti multilayers with a d-spacing of 26.6 Å and 50 bilayers, and the interfacial roughness of 3.0 Årms has been evaluated.

1. Introduction

Multilayer neutron monochromators(1) and supermirrors(2) are now widely used at many neutron sources for neutron optical devices. A decrease of the d-spacing of multilayer is desirable to lead grater angles of reflection for most applications such as neutron guide with higher transportation efficiency.

One of the most important problem in the case of producing the small d-spacing multilayers is the interface roughness which becomes large with an increase in the number of bilayers deposited. If the interface roughness can be kept small compared to the bilayer spacing for a number of bilayers, a large reflectivity can be achieved by making a sufficient number of layers.

When Ni/Ti multilayers, which are widely used for these neutron optical devices, are fabricated, the above mention problem is also pointed out. The Ni layers are very highly oriented with the (111) planes parallel to the substrate plane, and the large grained Ni layers with a structure of columnar growth lead to rough interfaces and a gradual degradation in layer structure with an increase in the number of layers(3). The decrease of crystallization of Ni has been carried out successfully by Ni-C alloying and it also increases the scattering length density(4),(5). Nowadays NiC/Ti multilayers are used most commonly, and have been fabricated with d=40Å and a roughness $\sigma \sim 6$Å.

If we are getting toward the coating of multilayer with less surface roughness and toward coating of the smaller d-spacing multilayer with d= ~ 20Å, which would be utilized for a multilayer ultracold neutron turbine source(6) and focusing system(7), a new technique to control the surface roughness with few atoms layer is required.

Recently ion polishing (also called as ion bombardment) in combination with electron beam deposition has been shown to be a promising tool in the production of X-ray reflecting multilayer structures(8)-(9). The concept of ion polishing is mainly explained as follows, particles with the lowest binding energy, which are in a valley of the surface, will be removed first by recoil effects after ion bombardment. But the roughening and intermixing of interface might take place in the case of higher energy ion bombardment. In this study, we have applied ion polishing with a combination with ion beam sputtering deposition for the fabrication of Ni/Ti multilayers. Because of an advantage of sputtering that the sputtered atoms arrive with higher energy (10~30 eV) than vacuum evaporation (~0.1eV), it can produce good quality layers with higher density and small grain size. In this paper we have investigated that the influence of argon ion polishing in the interface roughness of the ion beam sputtered Ni/Ti system over a wide range in order to optimize the Ar^+ ion beam parameters.

2. Experimental and characterization

Ni/Ti multilayers were deposited and ion-polished using the ion beam sputtering system which is shown in Fig.1(10). The system is equipped with dual bucket sources which are used to generate Ar^+ ions. One is used for sputtering deposition of Ni/Ti multilayers. The other is used for ion polishing. The base pressure during the operation was 1×10^{-7} mbar. Multilayers were deposited on Si(111) substrates of 75mmϕ with each surface roughness less than 3 Årms. The ion treatment was applied immediately after deposition of each of the nickel or titanium layers to smoothen the layers.

The d-spacing and effect of interfacial roughness of multilayers were measured with X-ray grazing angle reflectivity measurements, which were performed in a θ-2θ mode between 0° and 10° using Cu K_α radiation (λ=1.54Å). Neutron reflectivity measurements were conducted on the C3-1-2 reflectometer of the JRR-3M reactor at

JAERI(11). A well collimated ($\Delta\theta=1.5\times10^{-4}$ rad) beam with a wavelength of 12.6Å ($\Delta\lambda/\lambda=3.2\%$) was used in θ-2θ mode experiments.

In this study, the time dependence of ion bombardment as well as the energy and incidence angle, was studied within the energy range 100eV - 600eV, an angular range of 10° and 45° to optimize the parameters of Ar^+ ion bombardment. Ni/Ti multilayers (d=100 Å) were mainly deposited and characterized to optimize the ion parameters. Ni/Ti multilayer with a very small period (d=20Å, N=50bilayers) has been fabricated and characterized after obtainment of the optimum smoothening parameters.

3. Results and discussion

3.1 Polishing time

In order to apply ion polishing to each layer in a Ni/Ti multilayer, the optimum polishing time of nickel or titanium layer has been investigated. Ni/Ti multilayers (d= 120Å, N=10bilayer) with all Ni layers or all Ti layers ion-polished were coated as a function of Ar^+ ion polishing time. The multilayers were characterized by X-ray reflectivity measurements which is sensitive to the properties of the interfaces and multilayer structure. The ion energy and incidence angles were constant at 100eV and 10° during this experiment.

Figure 2 shows the results of Cu Kα measurements of multilayers with all Ni layers ion-polished for various ion polishing times. The diffraction patterns show Bragg peaks indicating a well defined composition modulation in the growing direction of the multilayers. It is obvious that Bragg peaks up to the 10th order are observed at an ion polishing time of 69 sec from Fig.2. The optimal ion polishing time seems to be in this time range.

Figure 3 shows the results of Cu Kα measurements of multilayers with all Ti layers ion-polished for various polishing times. Bragg peaks up to the 7th order are observed at the sample without ion polishing. On the other hand, Bragg peaks up to the 9th order are observed at polishing time of 127sec and 1310sec from Fig.3. The optimal polishing time seems to be in the time range of 127sec, since a maximum number of Bragg peaks up to a maximum scattering angle is observed in this case.

It is concluded that the quality of multilayers are obviously improved using ion polishing by reducing the interface roughness.

Table 1 shows the measured multilayer periods of these samples as a function of ion polish time. For the samples with all Ni layers ion-polished, the periods are almost constant with an increase in the ion polishing time. On the other hand, as for the samples with all Ti layers ion-polished, they increase with an increase in the ion polishing time. It may be caused by the density reduction of Ti layers or the limitation of inter-diffusions between Ni and Ti layers.

3.2 Ar^+ ion energy

The optimum Ar^+ ion energy has been investigated. Ni/Ti multilayers (d=100Å, N=10bilayer) with all Ni layers ion-polished or all Ti layers ion-polished were coated as a function of ion energy (100eV, 300eV and 600eV). The incidence angle was constant at 10° and each ion polishing time was constant at 69sec for Ni layers and 127sec for Ti layers during these experiments. Figure 4 shows the results of Cu Kα measurements of multilayers with all Ni layers ion-polished for various ion energies. It is obvious that Bragg peaks up to the 8th order are observed at each case, so the clear ion energy dependence is not confirmed in this range. The results of multilayers with all Ti layers ion-polished are shown in Fig.5, and the clear ion energy dependence is not also observed in this range.

3.3 Incidence angle

The optimum incidence angle of Ar^+ ion energy has been investigated. Ni/Ti multilayers (d=100Å, N=10bilayer) with all Ni layers ion-polished or all Ti layers ion-polished were coated as a function of incidence angles (10° and 45°). The ion energy was constant at 100eV and each ion polishing time was constant at 69sec for Ni layers and 127sec for Ti layers during these experiments. Figure 6 shows the results of Cu Kα measurements of multilayers with all Ni layers ion-polished for two incidence angles. Bragg peaks up to the 8th order are observed at the sample with an incidence angle of 10°, on the other hand, it is reduced to the 6th order at the sample with an incidence angle of 45°. From Fig.7, same tendency is observed at the sample with all Ti layers ion-polished. Bragg peaks up to the 8th order are observed at the sample with an incidence angle of 10°, on the other hand, it is reduced to the 7th order at the sample with an incidence angle of 45°.

3.4 Performance of very small d-spacing multilayer

We have prepared two Ni/Ti multilayers with a period of about 20Å and 50 bilayers. One has been fabricated with all Ni layers ion-polished based on the optimized polishing condition with an ion polishing time of 69sec, ion energy of 100eV, and incidence angle of 10°. The other has been fabricated without ion polishing. They were coated on Si(111) substrates of 75mmϕ with each surface roughness less than 3

Årms.

X-ray reflectivity measurements were performed with Cu Kα radiation using a low angle diffractometer. It was observed that a maximum of reflectivity of the first order peak was 11.6% and a d-spacing was 26.5Å at the sample having ion-polished Ni layers, while they were 0.37% and 22.5Å at the sample without ion polishing. On the other hand, calculated reflectivity using the optical constants of the bulk was 24.4% at the sample with a d-spacing of 26.5Å and 50 bilayers, while that of the sample with a d-spacing of 22.5Å and 50 bilayers was 15.0%. Then it is derived that the Debye-Waller factor which define the interfacial roughness is 3.0 Årms at the sample having ion-polished Ni layers and that of non ion-polished sample is 7.5 Årms.

Neutron reflectivity measurements for the same samples were performed using the neutron reflectometer at fixed wavelength (λ=12.6Å). From Fig.8, it is observed that a maximum of reflectivity is 0.48% and a d-spacing is 26.6Å at the sample with ion-polished Ni layers, while they are 0.11% and 22.7Å at the sample without ion polishing. On the other hand, calculated reflectivity based on the dynamical theory(12) assuming stoichiometric nickel and titanium at theoretical density is 6.24% for the sample with a d-spacing of 26.6Å and 50 bilayers, and that of the sample with a d-spacing of 22.7Å and 50 bilayers is 3.38%. In consideration of the difference of periods, the reflectivity of ion-polished sample is increased by a factor of 2.4 in comparison with the sample without ion-polishing. For the reliable evaluation of the interfacial roughness, the wavelength resolution of 3.2% of this machine may be not adequate, so the higher resolution experiment is being planned. Measured multilayer period agrees with that was measured by x-ray reflectivity measurement very well.

From above results, we can also concluded that the quality of multilayer is obviously improved using ion polishing by reducing the interface roughness even a very small d-spacing multilayer with a period of 20 Å.

4. Conclusion

In this paper, in order to apply ion polishing technique to producing of Ni/Ti multilayer in combination with ion beam sputtering deposition, the optimum ion beam parameters of ion polishing time, ion energy and incident angle have been investigated. The process is applied to Ni/Ti multilayer with a very short period of 26.6 Å and N=50bilayers, based on the optimized polishing condition with an ion polishing time of 69sec, ion energy of 100eV, and incident angle of 10°. The interfacial roughness of 3.0 Å rms has been evaluated.

Acknowledgements

The authors wish to express their thanks to Dr. T. Ebisawa, for his technical supports in the neutron reflectivity experiment.

References

(1) B. P. Schoenborn, D. L. D. Caspar and O. F. Kammerer, J. Appl. Cryst. 7 (1974) 508-510
(2) F. Mezei, Commun. Phys. 1 (1976) 81-85
(3) J. E. Keem, J. Wood, N. Grupido, K. Hart, S. Nutt, D. G. Reichel and W. B. Yelon, SPIE Vol. 983 (1988) 38-52
(4) J. L. Wood, SPIE Vol.1738 (1992) 22-29
(5) B. Vidal, Z. Jiang, F. Samuel, SPIE Vol.1738 (1992) 30-31
(6) M. L. Crow, L. Esibov, A. Steyerl, Nucl. Instr. and Meth. in Phys. Res. A 386 (1997) 474-482
(7) F. Mezei, SPIE Vol.983 (1988) 10-17
(8) E. J. Puik, M. J. van der Wiel, J. Verhoeven, H. Zeijlemaker, Thin Soild Films, 193/194 (1990) 782-787
(9) E. Spiller, SPIE Vol. 1160 (1989) 271-279
(10) K. Murakami, H. Nakamura, T. Oshino and H. Nikaido, OSA Proceedings on Soft-X-Ray Projection Lithography, Vol 12 (1991) 101-105
(11) T. Ebisawa, S. Tasaki, Y. Otake, H. Funahashi, K. Soyama, N. Torikai, Y. Matsushita, Physica B 213&214 (1995) 901-903
(12) V. F. Sears, Acta Crys., A39, 601 (1983)

Table 1 Multilayer d-spacings as a function of ion polish time

Ion polished material	Ion polish time	d [Å]	Δd [Å]
Ni layers	0	117.3	0.0
〃	9	117.4	0.1
〃	69	117.0	- 0.3
〃	320	117.6	0.3
〃	714	116.5	- 0.8
Ti layers	0	117.3	0.0
〃	127	124.8	7.5
〃	420	127.8	10.5
〃	1310	131.7	14.4

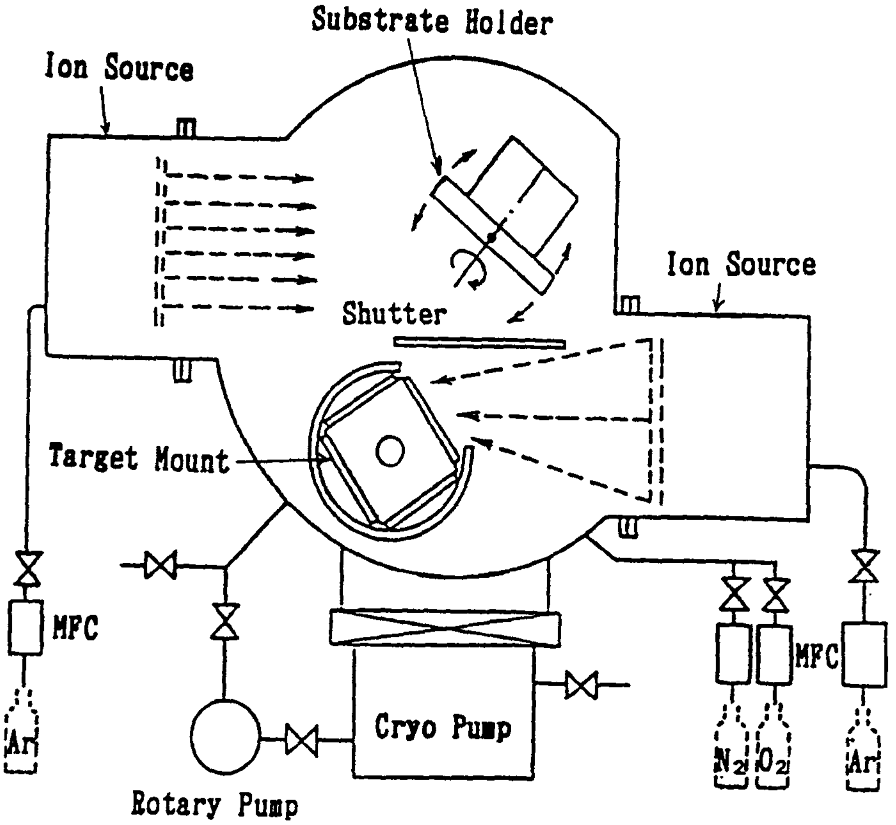

Fig.1 Schematic diagram of the ion beam sputtering deposition system

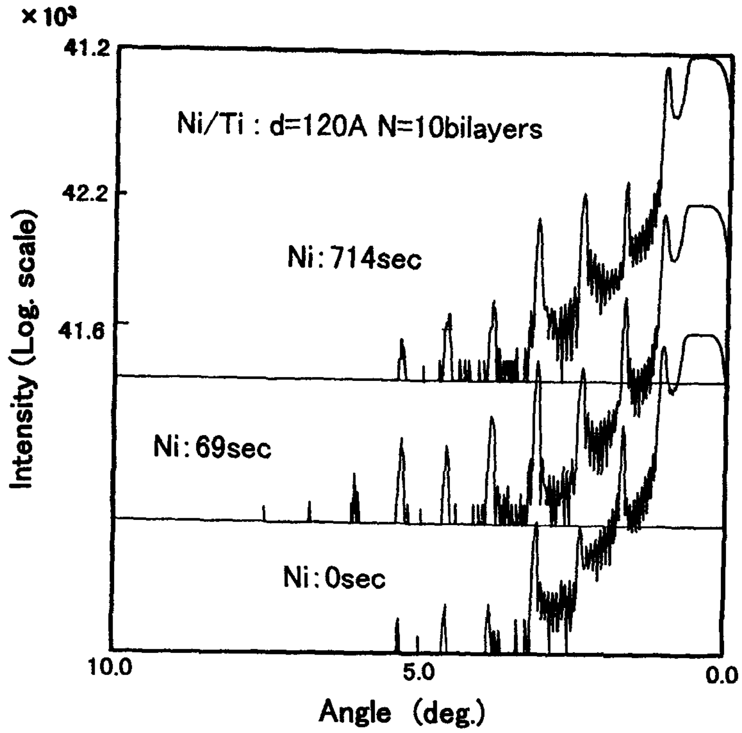

Fig.2 Cu Kα reflection profiles of Ni/Ti multilayers having ion polished Ni layers with variable ion polishing times

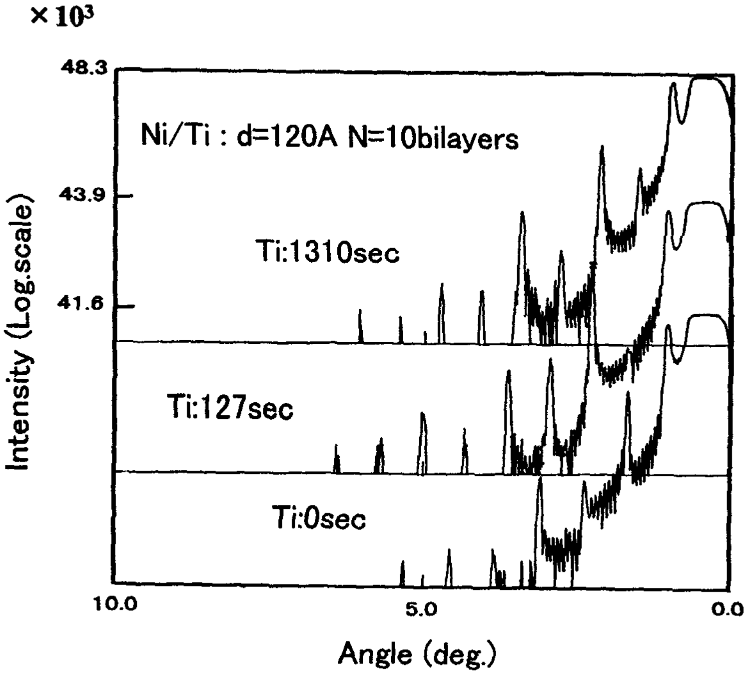

Fig.3 Cu Kα reflection profiles of Ni/Ti multilayers having Ti layers ion polished with variable polishing times

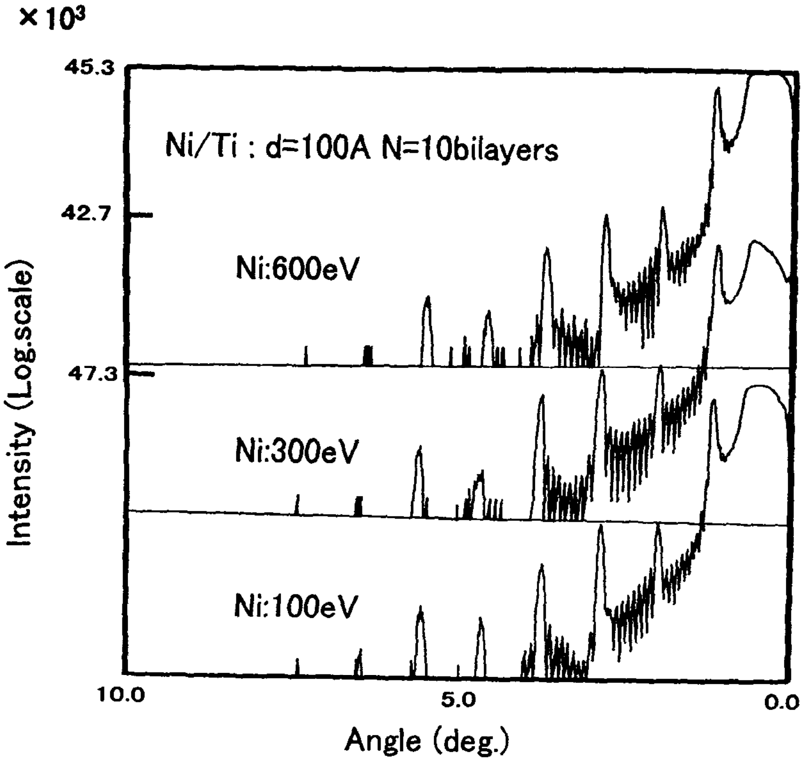

Fig.4 Cu Kα reflection profiles of Ni/Ti multilayers having Ni layers ion polished with variable ion energies

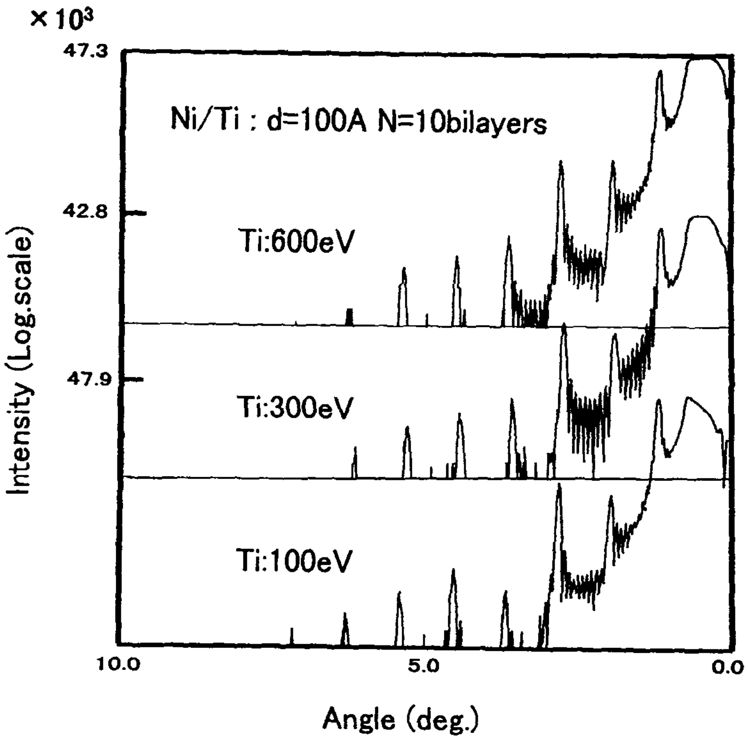

Fig.5 Cu Kα reflection profiles of Ni/Ti multilayers having Ti layers ion polished with variable ion energies

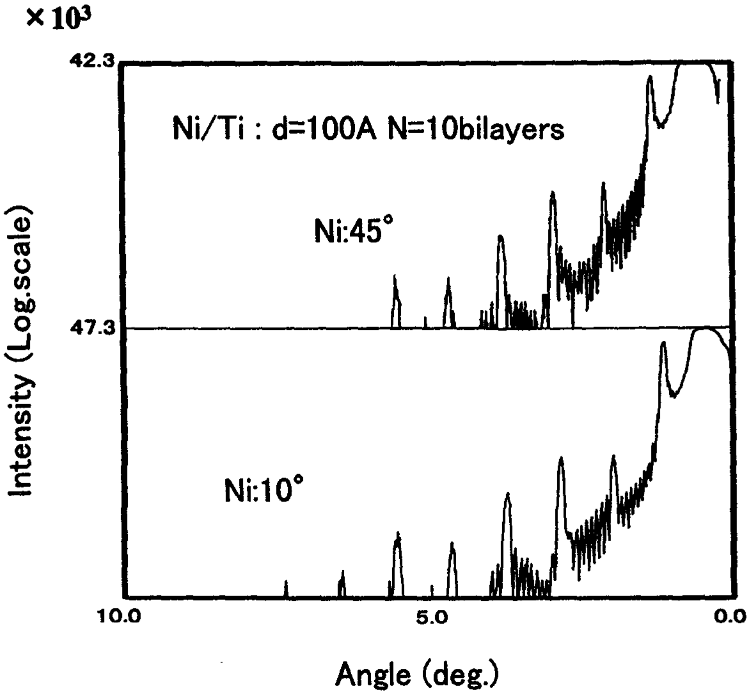

Fig.6 Cu Kα reflection profiles of Ni/Ti multilayers having Ni layers ion polished with variable incident angles

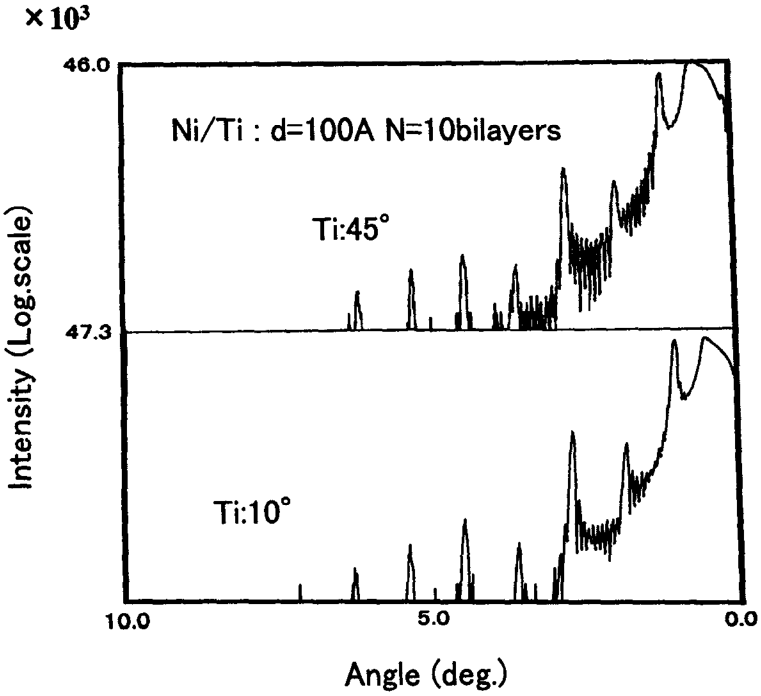

Fig.7 Cu Kα reflection profiles of Ni/Ti multilayers having Ti layers ion polished with variable incident angles

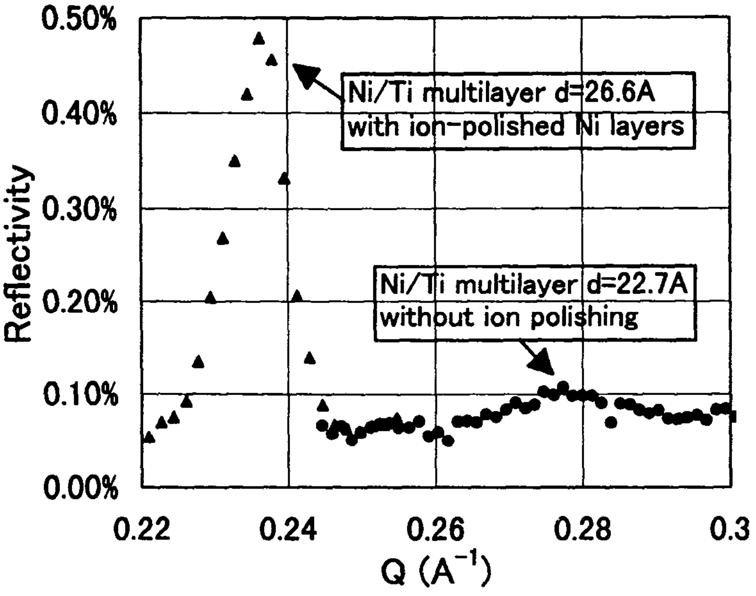

Fig.8 Neutron reflectivity profiles of Ni/Ti multilayers,
▲ with ion-polished Ni layers, ● without ion polishing