



# Hydrogen ion implantation effect on the magnetic properties of metallic multilayered films

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## Abstract

Metallic multilayers have attracted much interest from their interesting magnetic properties. Very thin magnetic layers separated by other metallic layers show a good perpendicular magnetization anisotropy. Modification of atomic structure at the interfaces is important to study for understanding the perpendicular magnetic anisotropy. Hydrogen can be easily introduced into metals, and be used to modify the interface structure of the multilayers. In this study hydrogen ions were implanted into Co/Pt multilayered films to modify the layered structure, and change of the magnetic properties was studied. The modification of the layered structure by hydrogen ion implantation did not enhance the perpendicular magnetic anisotropy. The perpendicular magnetization anisotropy energy of the multilayered film decreased with the increase in the dose of ions implanted, and changed to an in-plane magnetization film. After evacuation of hydrogen at 100°C, part of the perpendicular anisotropy was recovered. © 1999 Elsevier Science S.A. All rights reserved.

*Keywords:* Metallic multilayer; Co/Pt; Perpendicular magnetization; Magnetic anisotropy; Hydrogen ion-implantation

## 1. Introduction

Metallic multilayers have attracted much interest from their interesting magnetic properties [1]. Since the discovery of perpendicular magnetic anisotropy in Pd/Co multilayers [2], extensive studies have been made on Co/Pd, Co/Pt, Co/Au and others [3–14]. Very thin magnetic layers separated by other metallic layers show a good perpendicular magnetization anisotropy. The origin of the anisotropy has been explained by interfacial anisotropy in terms of Néel-type magnetocrystalline anisotropy [15,4,10], magnetoelastic anisotropy effect [16–18], or spin-orbit coupling effect [19]. Recently, Kim et al. [20] stressed the contribution of magnetoelastic effects at the alloy-like interfaces. Interface geometry and misfit of the lattices at the interface are important to study for understanding the origin of perpendicular magnetic anisotropy. Hydrogen can be easily introduced into metals, and be used to modify the interface structure of multilayers [21,22]. In this study hydrogen ion implantation was used to modify the layered structure and the change of the magnetic properties.

Specimens of Co/Pt multilayered film on Si substrate

with Pt buffer layer, and of a single Co layer with the same amount of Co as the multilayer were prepared by MBE (Molecular Beam Epitaxy). The multilayered film showed a good perpendicular magnetization anisotropy. The single layer Co specimen, which showed in-plane magnetization, was used as a reference.

The change of magnetic properties of the multilayered film by hydrogen ion implantation will be discussed.

## 2. Experimental

Films were prepared by MBE on Si (111) substrate with Pt buffer layer. The pressure during film growth was less than  $5 \times 10^{-9}$  Torr, and substrate temperature was 200°C. Single layered Co film and multilayered Co/Pt films were prepared: they are Pt 10 Å/Co176 Å/Pt250 Å/Si(111), and [Pt10 Å/Co4.8 Å]<sub>30</sub>/Pt250 Å/Si(111), where Pt250 Å are buffer layers. Hydrogen ions were implanted using an ion gun with acceleration voltage of 4 kV or 3 kV; the flux was  $1.5 \times 10^{13}$  ions/cm<sup>2</sup>s. After implantation hydrogen was evacuated by heating the specimens up till 100°C.

The structure was examined by X-ray diffraction, and magnetic properties were measured by VSM (vibrating sample magnetometer). The magnetic domain was ob-

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served by a scanning probe microscope in MFM (Magnetic Force Microscope) mode.

### 3. Results

X-ray diffraction profile of the single layer film shows buffer Pt(111) and film fcc Co(111) peaks. With hydrogen implantation the Pt peak did not change in position and profile, but the Co peak became broad and the intensity decreased, while the peak position shifted to a lower angle (Fig. 1a). In the multilayered film long-period diffraction peaks at low angles and Pt(111) buffer and fcc Pt/Co(111) film peaks at middle angles were observed, as shown in Fig. 1b and c. The intensity of low angle peak decreased by hydrogen implantation; the peak became broader and the peak position shifted slightly to lower angle. The diffraction peak from the film at middle angle also became small and broad and the peak position shifted to lower angle.

The saturation magnetization of the multilayered film first increased and then decreased as the dose of implantation increased, although that of the Co single layer film decreased monotonously with the increase in implantation dose.

The single layered film had in-plane preferential magnetization anisotropy, and this anisotropy did not change after hydrogen implantation. The magnetization curves for layered film are shown in Fig. 2. Fig. 2a shows hysteresis curves of perpendicular (thick line) and in-plane (thin line) magnetization before hydrogen implantation, and Fig. 2b

shows those after hydrogen implantation at 4 kV for 10 h. The hysteresis curves after hydrogen evacuation at 100°C are shown in Fig. 2c. Before hydrogen implantation the film showed a good perpendicular magnetization anisotropy, but it was changed to in-plane magnetization anisotropy by hydrogen implantation. This change of anisotropy by hydrogen implantation recovered partly with evacuation of hydrogen at 100°C. The character of magnetic anisotropy can be expressed by the effective perpendicular magnetic anisotropy energy,  $K_{\text{eff}}$ , which is obtained from magnetization curves measuring the energies to magnetize the film parallel and perpendicular to the film plane. The change of  $K_{\text{eff}}$  with hydrogen implantation time is shown in Fig. 3. Positive values of  $K_{\text{eff}}$  means the preference of perpendicular magnetization. With 4 kV implantation the multilayered film changed from a perpendicular to an in-plane magnetization film. With 3 kV implantation the tendency towards perpendicular preference was reduced by hydrogen implantation but the film remained to perpendicular preference after 10 h implantation. The single layer film retained in-plane magnetization anisotropy after hydrogen implantation.

The magnetic domains observed by MFM are shown in Figs. 4 and 5. The domain magnetized perpendicularly up or down is revealed as bright or dark image. Fig. 4 shows the MFM images before and after hydrogen implantation at 4 kV. Before hydrogen implantation the film shows clear bright and dark images for up or down magnetized domains, but the images become diffuse and unclear after implantation. The images at various stages of hydrogen implantation at 3 kV are shown in Fig. 5. With 3 kV

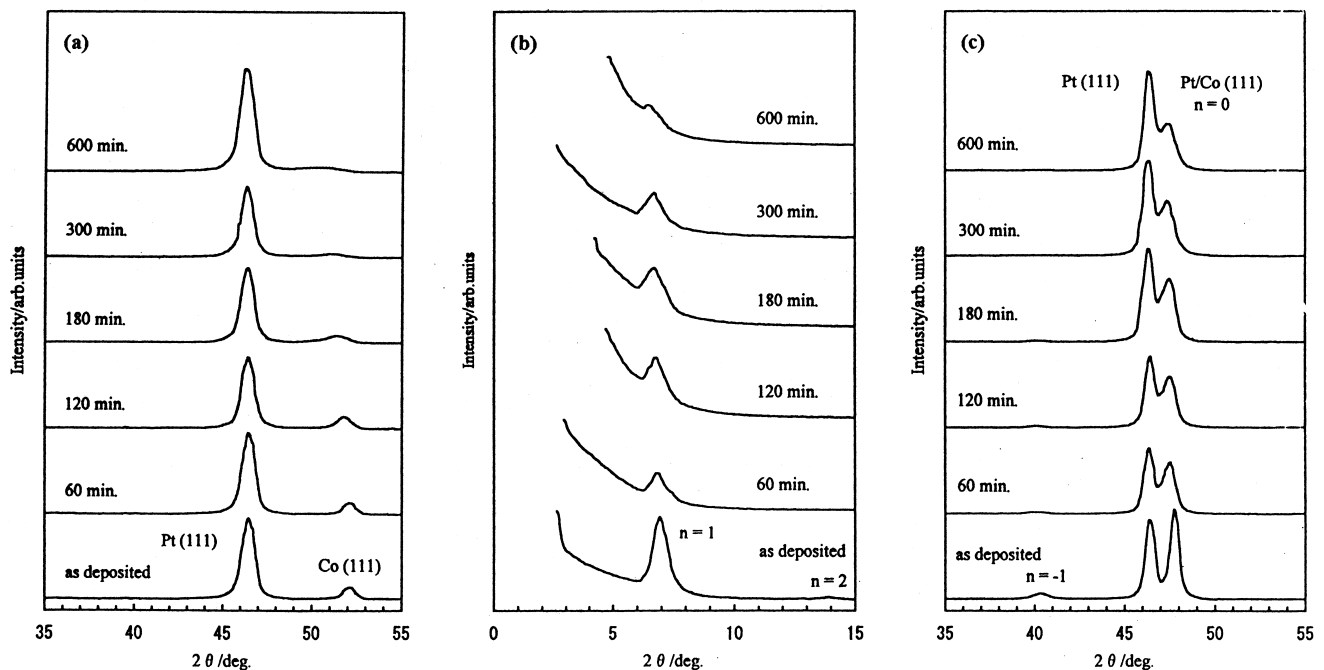


Fig. 1. Change of X-ray diffraction profiles by hydrogen implantation; (a) is for Co single layered film, and (b), (c) are for Co/Pt multilayered film at low diffraction angle and at middle angle, respectively.

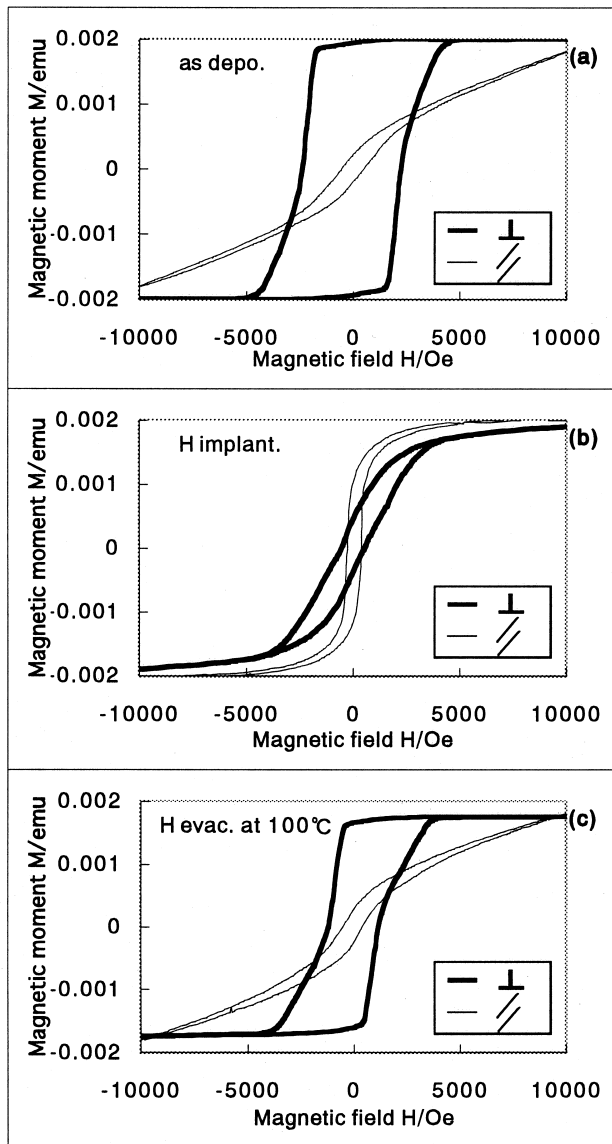


Fig. 2. Magnetic hysteresis curves of multilayered film (a) as prepared, (b) after hydrogen implantation at 4 kV for 10 h, (c) after evacuation of hydrogen at 100°C. Thick lines are for perpendicular magnetization, and thin lines are for in-plane magnetization.

implantation magnetization anisotropy did not change in nature, but the preference for perpendicular anisotropy reduced. The average domain size of perpendicularly magnetized domains decreased with the increase in implantation time, i.e. with the decrease of the value of  $K_{\text{eff}}$ .

#### 4. Discussion

Thin magnetic layer and well defined interface are considered as key factors for perpendicular magnetization anisotropy. Heat treatment disrupts the layered structure by atom mixing, and reduces the preference of perpendicular anisotropy. Modification of the layered structure by hydro-

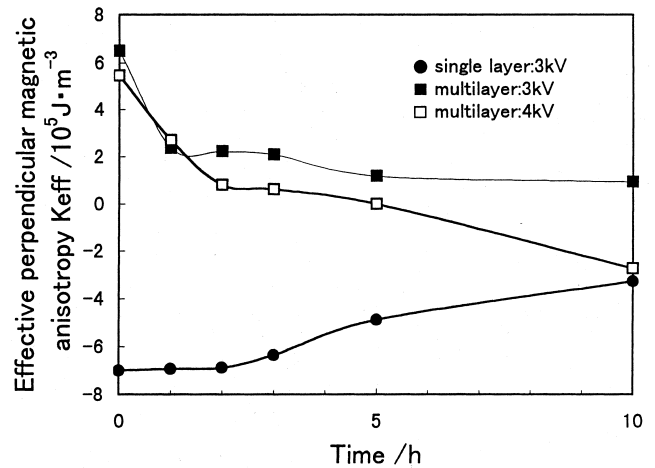


Fig. 3. Change of the effective perpendicular magnetization anisotropy energy,  $K_{\text{eff}}$ , with hydrogen implantation time.

gen introduction may be interesting for understanding the nature of magnetic anisotropy.

Ion implantation method has been employed in this study as a low temperature charging of hydrogen. By this method hydrogen can be introduced into the films irrespective of the solubility limit at low temperature, but lattice defects are inevitably produced during implantation. The acceleration voltage was chosen so that the implanted hydrogen range is in the middle of the film thickness. Simulation by using TRIM (TRansport of Ions in Matter) code revealed the formation of vacancies even at the acceleration voltage of 3 kV. The modification of the multilayer by hydrogen as well as vacancy must be considered in this case. Change of magnetic properties, decrease in saturation magnetization and small change in anisotropic energy, of the single Co layer film by hydrogen implantation may be induced by implanted hydrogen and lattice defects as well as some extent of atomic mixing of cap-layer and substrate Pt into Co layer.

In the multilayer films rearrangement of atoms at the interfaces by implantation sometimes induces an initial increase of saturation magnetization, but further modification of the interfaces makes the saturation magnetization as small as in the case of single layer film. In this experiment the main consequence of the modification of layered structure by hydrogen implantation may be assigned to the atomic mixing at the interfaces and the elastic lattice distortion by retained hydrogen. Ion implantation at 3 kV acceleration produces less lattice vacancies than at 4 kV. Hydrogen can be trapped by vacancies and be well retained in the film when irradiated at 4 kV.

Mixing of atoms and distortion of the lattice obstruct the preference of perpendicular magnetization anisotropy. The decrease of the value of  $K_{\text{eff}}$  means the decrease of domain wall energy in perpendicular magnetization, since the direction of magnetization in the domain wall has a large component in parallel to the plane. It also can be expected

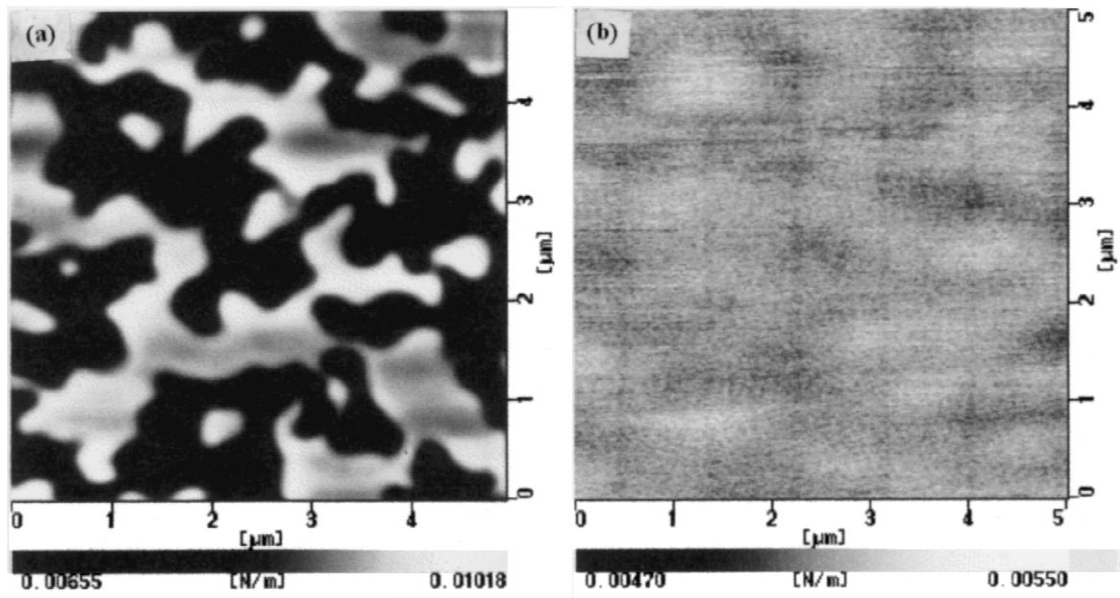


Fig. 4. Images of Magnetic Force Microscopy of multilayered film (a) before and (b) after hydrogen implantation at 4 kV. Note that (a) is of perpendicular anisotropy and (b) is in-plane anisotropy. Bright and dark regions correspond to the domains magnetized up and down directions.

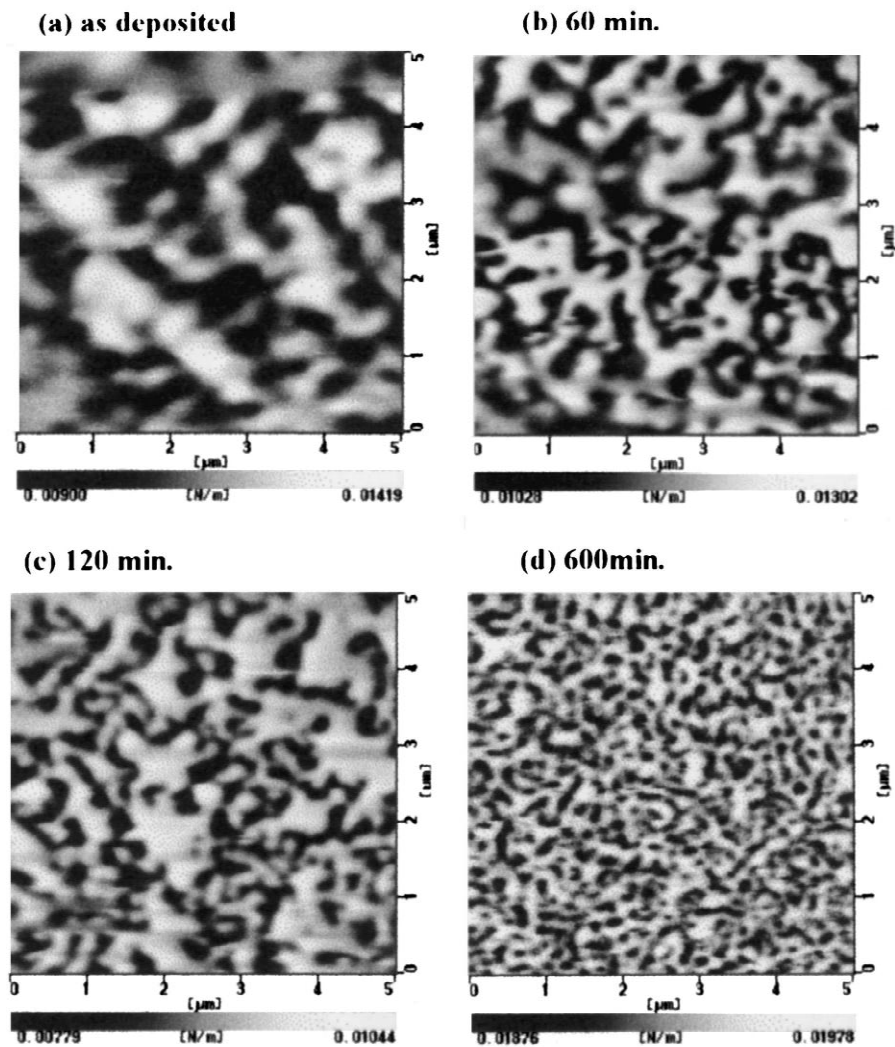


Fig. 5. Images of Magnetic Force Microscopy of multilayered film at various stages of hydrogen implantation at 3 kV. Note that the film retained perpendicular anisotropy after 10 h implantation.

that there is no preferential direction for in-plane magnetization. Therefore, the domain size of perpendicularly magnetized domains decreases and the domain wall can be curvaceous. The perpendicularly magnetized film reveals smaller domain structure by implantation as shown in Fig. 5. If the film changes to in-plane magnetization anisotropy, the characteristic image of perpendicular magnetization disappears. After evacuation of hydrogen at 100°C, perpendicular magnetization anisotropy is recovered to some extent as shown in Fig. 3. This can be explained by the release of elastic distortion induced by retained hydrogen. It can be concluded that atomic mixing at the interfaces and lattice distortion by hydrogen disrupt the preference of perpendicular anisotropy. Though the state and distribution of hydrogen are not clear, elastic distortion by hydrogen has a significant effect to the magnetic anisotropy of the multilayered films.

## 5. Conclusion

Multilayered Co and Pt films with perpendicular magnetization anisotropy were prepared, and change of structure and magnetic properties were examined after the modification of the film by hydrogen ion implantation. Structural changes by atomic mixing and elastic lattice distortion cause the decrease in perpendicular magnetization anisotropic energy. Elastic distortion of the multilayered structure by retained hydrogen plays a significant role in magnetic anisotropy.

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