

# Effect of Bi surfactant on the heteroepitaxial growth in Fe/Cr(100) multilayers

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## Abstract

This study was to investigate the surfactant effect of Bi on the heteroepitaxial growth of Fe/Cr(100) multilayers by reflection high-energy electron diffraction (RHEED) measurements. With predeposition of submonolayer Bi on Fe(100) prior to evaporation of Fe/Cr multilayer, more long-lasting RHEED intensity oscillations were observed. This implies that the layer-by-layer growth of Fe/Cr multilayer is enhanced. The observations of grazing incidence X-ray reflectivity confirmed that the interface structures of Fe/Cr multilayer with Bi were sharper than that of multilayer without Bi. The study was also to investigate the magnetotransport properties between Bi surfactant-mediated multilayers and normal ones. The magnetoresistance (MR) ratio of the multilayers was enhanced by predeposition of Bi.

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## 1. Introduction

Understanding the epitaxial growth of thin metal films at an atomic scale is essential for the manipulation of the thin structure and interfaces to improve the quality of thin-film devices. The correlation between the interface structure and magnetic or electrical properties of metallic multilayers has not yet been fully understood. It is, however, proved that growth behavior of thin films greatly affects the properties of metallic multilayers. Consequently, knowledge of controlling the growth mode is necessary to understand the basic

properties of metallic multilayer. Recent investigations on the fabrication of self-organized low-dimensional film structures suggested that the control of surface free energy and growth kinetics were crucial to obtain well-defined structures. This is because the surface processes during epitaxy are dominated by these two factors. During epitaxial growth of thin films it is, however, often the case that the desired film structure is not thermodynamically favorable.

To produce step edges of uniform height and sustain their production through the growth process, 2D layer growth following a Frank-van der Merwe model (layer-by-layer and step-flow mechanism) has to be achieved, leading to the uniformity and interface abruptness needed for the high quality of the epitaxial films. Surfactant epitaxy is a useful method for changing the thin film growth mode from 3D-island formation to layer-by-layer growth [1,2].

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Employing surfactant-mediated epitaxy, in which surfactant atoms are predeposited onto a substrate surface prior to growth, can control the surface energy and growth kinetics. A study by van der Vegt et al. reported that considerable surfactant effects in the growth of metals on [3]. Camarero et al. reported that a small amount of Pb deposited before the deposition of Co/Cu bilayers on Cu(111) reduced the amount of fcc twin formation, thereby improving the structural quality [4,5]. Previously, using Bi as a surfactant, we found experimentally that the surfactant atoms could induce both the layer-by-layer growth of Fe and Cr films onto Fe(100)- $c(2\times 2)O$  reconstruction surface as well as a strong surface segregation effect of Bi during growth [6,7]. Heteroepitaxial systems with surfactants are of greater interest for electronic devices, such as the giant magnetoresistance (GMR). There have been, however, only few examples of application for the metallic multilayers [8–11].

The GMR effect was found in metallic multilayers consisting of alternating ferromagnetic and non-magnetic films [12,13]. Numerous observations, especially in the Fe/Cr system, have been reported to indicate that interface roughness has a profound influence on the magnitude of the GMR effect. However, no clear picture has yet emerged and the reported data are contradictory. It was found that the increase of interface roughness might enhance the GMR [14,15]. In contrast to these results, a large GMR has been found in Fe/Cr multilayers with sharp interfaces [16,17]. Therefore, it is important to get detailed information on the growth behavior and its effect on interface morphology.

This paper reports the influence of Bi as a surfactant on the heteroepitaxial growth of Fe/Cr(100) metallic multilayers, combining results from reflection high-energy electron diffraction (RHEED), grazing incidence X-ray reflectivity (XRR) measurements and Auger electron spectroscopy (AES). The research also investigated the magnetotransport properties between Bi surfactant-mediated multilayers and normal ones.

## 2. Experimental procedure

Molecular beam epitaxy (MBE) experiments have been carried out. The base pressure was on  $10^{-10}$  Torr order and the pressure during deposition did not exceed  $5\times 10^{-9}$  Torr. Fe, Cr, and Bi were deposited from electron beam gun evaporators. Fe(100) buffer layers were fabricated by evaporating Fe on MgO(100) single crystals. The substrates of MgO(100) single crystal were cleaned by heating at 850 °C for 10 min. Fe buffer layers of 100 Å thick were deposited on the substrates at a rate of 0.04–0.06 Å/s, and the growth temperature was 200 °C. After the deposition of Fe, the buffer layers were annealed at 850 °C for 30 min to obtain a flat Fe(100) surface. Bi was used as the surfactant. RHEED intensity measurements were performed during deposition of Fe/Cr on an Fe(100) buffer layer with and without predeposited Bi at 100 °C. The Fe, Cr and Bi were

evaporated on Fe buffer layer at the deposition rates of 0.09–0.11, 0.09–0.11 and 0.01–0.02 Å/s, respectively, which were controlled using quartz thickness monitor. Deposition temperature was maintained at 100 °C during the preparation of the Fe/Cr multilayer. In all multilayers, thickness of the Cr layer and Fe layer were fixed at 10 and 20 Å, respectively.

The morphology of the surface during the deposition was determined by RHEED measurements. To analyze the interface structures of the Fe/Cr multilayers, a grazing incidence X-ray reflectivity technique was carried out. XRR measurements were performed by high resolution X-ray diffractometer. The samples were mounted with a vertical sample stage. The scanning rates were 0.01°/min with a step of 0.005° automatically controlled by a computer. The corresponding range of the scattering angle  $2\theta$  was from 0.4 to 4.2°. The measured data were fitted by computer in order to obtain the information of the interface root-mean square (rms) roughness. AES was used to determine the composition of film surfaces. The samples were analyzed using a scanning Auger electron microprobe PHI-680 with electron beam energy of 3 keV, at a probe current of 5 nA and a diameter of approximately 2 μm.

To analyze the magnetic or electrical properties of multilayers, vibrating sample magnetometer (VSM) and four-lead magnetoresistance measurements were carried out at room temperature. The  $\Delta R/R$  data were normalized to the resistance at 20 kOe field. The current and the magnetic field were applied parallel to the film plane.

## 3. Results and discussion

To avoid a shunting effect, which often becomes a problem when applying to the electric device, the thickness of the buffer layer should be thin [18,19]. Therefore, the thickness of Fe buffer layers used on MgO(100) single crystal substrates was 100 Å. The RHEED patterns of the Fe buffer layer surface are shown in Fig. 1. Azimuths of the incident beam are parallel to the [010] direction in (a) and to the [011] direction in (b) for bcc-Fe(100). The epitaxial relationship between Fe(100) and MgO(100) is determined from the RHEED patterns as: Fe[010]||MgO[011] and Fe(100)||MgO(100). As shown in Fig. 1(b), half-order streaks (indicated by arrows) are observed between the first streaks in the RHEED patterns from the Fe[011] direction. Bertacco et al. reported that low energy electron diffraction and X-ray photoemission revealed that annealing promotes oxygen surface segregation giving rise to a  $c(2\times 2)$  reconstruction even in films several hundred Å thick [20]. Therefore, the surface structure of Fe buffer layer is considered to be an Fe(100)- $c(2\times 2)O$  reconstruction surface. This structure disappears gradually with the deposition of Cr and Fe (shown in Fig. 4).

The surfactant effects of Bi in the epitaxial growth of Fe and Cr on Fe(100)- $c(2\times 2)O$  were investigated in our previous works [6,7]. Results of those studies indicated that

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