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Measurement of Elastic Stiffness of Fe, Cr and Fe/Cr-Multilayer Films by Picosecond Ultrasound

ピコ秒超音波による Fe、Cr および Fe/Cr 多層膜の弾性率測定

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1. Introduction

Elastic stiffness of multilayer films has been measured by several methods, and unexpected elastic properties have been observed. In continuum mechanics, macroscopic elastic stiffness of layered structure is obtained from the elastic stiffness and volume fraction of the constituent materials using the rule of mixture. However, in multilayer films, measured stiffness tended to be smaller than the calculated values, and in some films, the stiffness showed a minimum when thickness of each layer was changed [1-3]. Some causes, the interfacial strain, electronic contribution, et al., are proposed to explain the results, but we consider that it is still unclear if the models can explain stiffness of several kinds of multilayer films. Stiffness measurement for several multilayer films and comprehensive analysis on the results are indispensable for developing a comprehensive model.

For establishing a robust model, we have investigated elastic stiffness of multilayer films using the picosecond ultrasound. Picosecond ultrasound is the noncontact method that can measure round-trip time of a longitudinal acoustic pulse propagating in thin film [4]. In acoustic methods, substrate stiffness is required to determine the film stiffness, but by using the picosecond ultrasound, elastic stiffness is determined only from the round-trip time, mass density, and film thickness [5]. Regarding multilayer films, we have studied Co/Pt and Fe/Pt systems, and observed elastic softening [6,7]; stiffness becomes smaller than the value calculated using the rule of mixture. These systems possesses relatively large lattice misfit, and contributions of interfacial strain and dislocation were supposed as dominant factors. In contrast, we recently studied Fe/Cr films whose lattice misfit is ~0.6 % [8], and observed that the measured stiffness is somewhat close to the bulk value, which is different behavior from the above systems. This implies that degree of softening changes depending on the lattice misfit, and dominant mechanism may also change. In monolayer films, elastic stiffness becomes comparable to or smaller than that of the corresponding bulk, and it recoveries to the bulk stiffness by annealing treatments [9]. This implies that constituent layers in multilayer films are softer than the bulk, and it becomes a dominant cause when the lattice misfit is small; unusual elastic property does not appear at the interfaces. In this study, to confirm the above interpretation, Fe/Cr multilayer films are prepared, and relationship between the thickness ratio of Fe and Cr and elastic stiffness is evaluated in more detail. Elastic stiffness of Fe and Cr monolayer films is also measured, and relationship between monolayer stiffness and multilayer stiffness is discussed.

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2. Specimens

Films were prepared by RF magnetron sputtering on monocrystal silicon substrates at room temperature. Surface of the substrate is covered with native oxide. Total thickness of multilayer films is about 60 nm. Thickness of each layer is controlled by the deposition time.

3. Picosecond ultrasound

The longitudinal elastic stiffness $C_\perp$ in the thickness direction is measured using the picosecond ultrasound. By irradiating the film surface with a femtosecond pulse laser, an acoustic pulse is excited. It propagates in the thickness direction in the film, and reflection and transmission occur at the interface with the substrate. The reflected pulse reflects at the film surface, and multiple reflection occurs in the film. By measuring the reflectivity change near the film surface using another femtosecond laser, multiple reflection echoes are observed in time-reflectivity relationship. From the round-trip traveling time Δt, mass density ρ, and the film thickness d, $C_\perp$ is determined as $C_\perp=\rho(2d/\Delta t)^2$. In some previous studies, multilayer films are assumed to be elastically isotropic, although they show anisotropy between the in-plane and out-of-plane directions. By using the picosecond ultrasound, elastic stiffness is determined with considering the elastic anisotropy. The total film thickness is determined by the x-ray reflectivity measurement and the bulk mass density is used to determine the stiffness.

4. Results and discussion

Figure 1 shows an example of the x-ray reflectivity spectrum. A large peak at 2θ=2° originates from the interference of x-rays reflected at Fe-Cr interfaces, and appearance of the peak represents that the multilayer film possesses the periodic structure. The peak angle is related to the bilayer thickness (periodicity). The other peaks originate from the interference of x-rays reflected at the top surface and interface between the film and substrate. By fitting the spectrum with the theoretical function, total film thickness is determined.

Figure 2 shows an example of the reflectivity change measured for the Fe/Cr multilayer film. A couple of echoes are observed, and Δt is determined from the echo intervals.

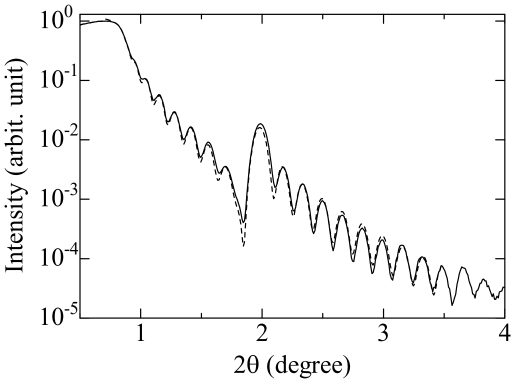

Fig.1 X-ray reflectivity spectrum for an Fe/Cr multilayer film (d_{Cr}=3nm, d_{Fe}=3nm, and 10 periods). The solid and dashed curves are the measured spectrum and the fitted function, respectively.

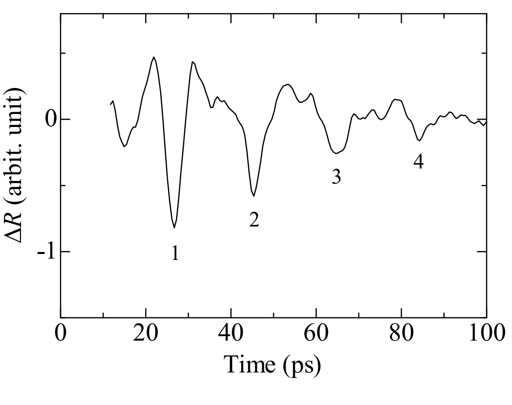

Fig.2 Reflectivity change measured by the picosecond ultrasound for an Fe/Cr multilayer film (d_{Cr}=3nm, d_{Fe}=3nm, and 10 periods). The numbers indicate the echoes.

First, elastic stiffness of Fe and Cr monolayer films was measured, and they were compared to the bulk values. The measured stiffness was smaller than those of the corresponding bulks by 2-9 %. Such softening is observed in other metallic films, and we confirm that softening is a general behavior in monolayer films.

Then, several Fe/Cr multilayer films were prepared by changing the thickness of Fe and Cr layers, and elastic stiffness was measured. For comparison, the macroscopic stiffness was calculated from the bulk stiffness using the rule of mixture. According to the rule of mixture, elastic stiffness changes monotonically, when the thickness ratio of Fe and Cr changes monotonically. In the measured stiffness, the monotonic change is observed, but the values were smaller than the calculated values. So, assuming that the Fe and Cr layers are softer than the bulks like Fe and Cr monolayer films, the macroscopic stiffness from the elastic stiffness of monolayer films was calculated. As the result, the measured stiffness showed agreement with the calculated stiffness.

When the thickness ratio of the Fe and Cr layers are changed, the number of the Fe-Cr interfaces changes. However, without considering the stiffening nor softening at the interfaces, the measured stiffness is reproduced only by considering the softening of Fe and Cr layers. These results indicate unusual elasticity does not appear at the interfaces when the lattice misfit is not so large, and the macroscopic stiffness of multilayer films can be estimated from the stiffness of monolayer films.

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