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Abstract. Fe atoms in Fe (16 Å)/MgO (60 Å) multilayered films, prepared in UHV conditions, have been identified as having the same BCC structure as for the bulk specimens, using x-ray diffraction. The lattice parameter for the Fe atoms was found to be larger than the bulk value by 1.0%. Such a lattice dilation is related to the enhancements in magnetization and hyperfine field of Fe atoms in Fe/MgO multilayered films.

1. Introduction

Multilayered films whose thickness is in a range from several to several tens of ångströms have attracted much attention as new synthetic materials in recent years. The most striking feature of multilayered films is an artificial periodicity produced by the sequence of an alternate layered structure, which is referred to as an artificial superlattice. In metallic systems, there is an opportunity to design the electronic state of the materials through such an artifical periodicity. On the other hand, the layer thickness of multi-layered films is so extremely thin that this leads to two peculiar crystallographic features. One of them causes an anomalous elastic constant (Wang *et al* 1977, McWhan *et al* 1983, Ohishi *et al* 1989) and a lattice deformation (Ohishi *et al* 1989) due to the size effect. The other gives a magnetic anomaly due to the concentration gradient of the interfaces. It is, therefore, of great interest to obtain the magnetic moment at interfaces in theoretical and experimental studies (Ohnishi *et al* 1984, Oguchi and Freeman 1986, Krishnan and Tessier 1990, Xiao *et al* 1990).

There are many reports on the interfacial magnetism of Fe, because Fe is a most popular element for Mössbauer spectroscopy (Shinjo and Takada 1987). The magnetic properties at interfaces have been studied by ^{57}Fe Mössbauer spectroscopy, and it has been reported that the hyperfine field is enhanced. The materials studied are as follows: Ag (Keune *et al* 1979, Droste *et al* 1986, Koon *et al* 1987, Volkeing *et al* 1988), MnF$_2$ (Droste *et al* 1986), Pd (Hosoito *et al* 1981), MgF$_2$ (Droste *et al* 1986, Hine *et al* 1979), MgO (Hine *et al* 1979, Shinjo *et al* 1979), γ-Fe$_2$O$_3$ (Shinjo *et al* 1982) and GdO$_x$ (Koyano *et al* 1987). Hine suggested further that the magnetic moment of Fe atoms in the Fe/MgO system is enhanced at such interfaces, the origin being a lattice dilation of Fe layers (Hine *et al* 1979). Recently, we reported that enhancements in both the hyperfine field

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and the magnetic moment were observed in Fe/MgO multilayered films (Koyano *et al* 1988, 1989). However, there are no reports which discuss the relation between the lattice dilation and enhancements of the magnetic moment experimentally.

X-ray diffraction experiments have been performed to obtain the average structure and lattice spacings of Fe atoms in Fe/MgO multilayered films prepared in UHV conditions. The relation between the lattice dilation and enhancements of the magnetic moment of Fe atoms is discussed in the final section.

2. Experimental procedures

Fe/MgO multilayered films were prepared with an alternate deposition technique in a vacuum of better than 5×10^{-9} Torr. Natural Fe and MgO were evaporated from two electron-beam-heating apparatuses, which are independently controlled by an Inficon XTC quartz thickness monitor system. The layers were deposited at the rate of about 0.3 Å s^{-1}. The substrate, a Si(111) wafer, was water cooled during deposition. The deposition of an Fe/MgO bilayer, which consists of an Fe layer of 16 Å and a MgO layer of 60 Å (referred to as Fe(16 Å)/MgO(60 Å) hereafter), was repeated 100 times.

The x-ray diffraction measurements with synchrotron radiation were performed on BL-4C at the Photon Factory, National Laboratory for High Energy Physics (KEK), Tsukuba, Japan, where a four-circle diffractometer (Huber 5020.4) equipped with a graphite (00.2) crystal analyser was used (Iwasaki *et al* 1989). A wavelength of 1.00 Å was selected by the double-crystal Si(111) monochromator with sagittal focusing. The positions of diffraction lines were calibrated with the Si(111) and Si(333) Bragg reflections from the substrate. The diffraction profiles in the small-angle region were also measured with a conventional x-ray diffractometer with a Cu target. All the x-ray diffraction patterns were taken at room temperature.

3. Results and analysis

Figure 1 shows the x-ray diffraction pattern of Fe(16 Å)/MgO(60 Å) multilayered film measured with the reflection geometry ranging from $2\theta = 22°$ to $2\theta = 45°$. The full and broken lines at the top of the figure indicate the positions of Bragg reflections of bulk Fe with the BCC structure and of MgO with the NaCl structure, respectively. Profiles of Bragg reflections are asymmetric at around 28° and 39° and seem to be composed of two lines. Rocking curves collected at the positions corresponding to diffraction lines showed no peaks, and so we believe that the film has little preferred orientation. In an x-ray small-angle region, Bragg peaks have been observed only up to third order. It is thought that this is due to the rough interface between Fe and MgO layers.

To obtain the average structure and lattice spacings of Fe atoms in the multilayered film, the profile analysis was performed for the data as shown in figure 1. The diffraction profile of multilayered films is described by the following equation:

$$I_{\text{obs}}(Q) = KA(Q)P(Q)L(Q)|F_h(Q)|^2 \tag{1}$$

where K, $A(Q)$, $P(Q)$, $L(Q)$, $F_h(Q)$ and Q are a scale factor, an absorption factor, a polarization factor, the Lorentz factor, a structure factor and $(4\pi \sin \theta)/\lambda$, respectively. The absorption factor for a thin film placed in the symmetrical reflection geometry is given as $A(\theta) = [1 - \exp(-2\mu t/\sin \theta)]/2\mu$ where μ is an average linear absorption
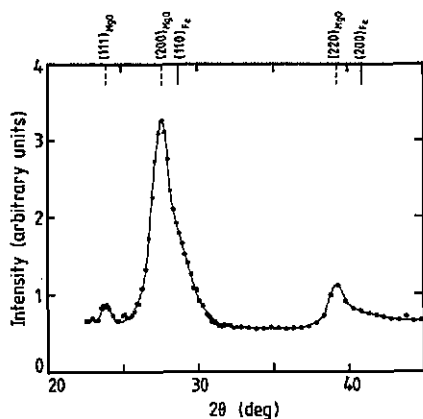
Figure 1. X-ray diffraction pattern of Fe(16 Å)/MgO(60 Å) multilayered film measured along the surface normal direction. The full and broken lines at the top of the figure indicate the positions of Bragg reflections from bulk Fe and MgO, respectively.
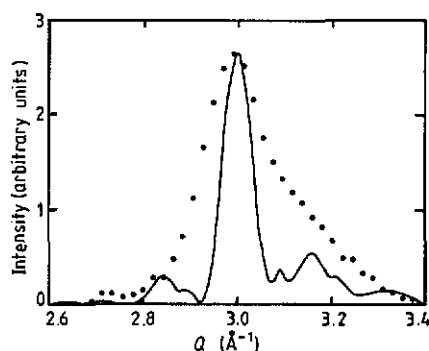
Figure 2. The calculated intensity profile at around $2\theta = 28°$ with the use of equation (2), in which the coherent length is given as $(d_{110}^{Fe} \times 8 + d_{200}^{MgO} \times 28)$ Å. The layer spacings d_{110}^{Fe} and d_{200}^{MgO} are 2.027 Å and 2.106 Å, respectively. The observed intensity profile is also shown (●).

coefficient. $P(Q)$ is unity as the synchrotron radiation is utilized in this experiment. As the specimen is regarded as a powder sample, $L(\theta)$ is $1/\sin^2 \theta \cdot \cos \theta$. For the one-dimensional step model (Fujii *et al* 1986), the structure factor $F_h(Q)$ is expressed as

$$F_h(Q)^2 = L_n(Q)|F(Q)|^2 \tag{2}$$

$$L_n(Q) = \sin^2(n\Lambda Q/2)/\sin^2(\Lambda Q/2) \tag{3}$$

$$F(Q)^2 = f_{Fe}^2 L_{Fe}(Q) + f_{MgO}^2 L_{MgO}(Q) + 2f_{Fe}f_{MgO}[L_{Fe}(Q)L_{MgO}(Q)]^{1/2} \cos(\Lambda Q/2). \tag{4}$$

Here $L_n(Q)$ is the Laue function due to an artificial periodicity Λ, $F(Q)$ is the structure factor within one pair of constituent layers and f_X is an atomic scattering factor for species X ($\equiv$ Fe, MgO). In these equations, n represents the number of pairs of constituent layers. According to equation (2), diffraction linewidths are determined dominantly by the linewidth of $L_n(Q)$, since an artificial period is much longer than inter-atomic spacings. With the use of equation (2), we calculated the intensity profile in which the coherent length Λ is given as $(d_{110}^{Fe} \times 8 + d_{200}^{MgO} \times 28)$Å. The layer spacings d_{110}^{Fe} and d_{200}^{MgO} are 2.027 Å and 2.106 Å, corresponding to the bulk values, respectively. As shown in figure 2, the agreement between the observed and calculated diffraction profiles is not so good. It is thought that the one-dimensional coherent model is not suitable for analysing the observed intensity profile. The reason might be due to the rough interface between the Fe and MgO layers. We, therefore, introduce the incoherent model whose diffraction profile is given as follows:

$$I_{obs}(Q) = KA(Q)P(Q)L(Q)[M_{Fe}f_{Fe}^2 L_f(Q) + M_{MgO}f_{MgO}^2 L_m(Q)] \tag{5}$$

where M_X and $L_X(Q)$ represent the multiplicity of atomic planes and the Laue function of atomic layers for X, respectively. The terms $L_f(Q)$ and $L_m(Q)$ can be expressed in

the same way as equation (3), except that Λ and n correspond to the spacing d of each of the atomic planes and the number of coherent atomic layers of corresponding diffraction planes of Fe and MgO, respectively. It can be said that the profile corresponds to an incoherent addition of diffraction profiles of Fe and MgO layers. Therefore, equation (5) is reduced to the following equation:

$$I_{calc}(Q) = KA(Q)L(Q)[CL_f(Q) + L_m(Q)]. \tag{6}$$

In this equation, C is an intensity ratio of the Bragg reflection of Fe to that of MgO. The parameters K, C, n_{Fe}, n_{MgO}, d_{110} and d_{200} are fitting parameters, where n_{Fe} and n_{MgO} represent coherent atomic layer number of Fe and MgO, respectively and are integers for the present model. Thee parameters were refined to minimize the reliability factor R:

$$R = \left(\sum |I_{obs}(Q) - I_{calc}(Q)|\right)\Big/\sum I_{calc}(Q). \tag{7}$$

The results of least-squares fitting and experimental data around $(200)_{MgO}$ and $(110)_{Fe}$ lines are presented in figure 3(a), where no fitting parameters were constrained. The d-values of the Fe(110) and MgO(200) planes thus obtained are 2.047 Å and 2.107 Å, respectively, where the R-factor is 4.2%. The d-value of Fe is larger than that of the bulk value, 2.0268 Å, and the d-value of MgO is the same as the bulk value, 2.106 Å. The fitting result, where d-values of Fe and MgO were fixed to corresponding bulk values, is also shown in figure 3(b), and the resulting R-factor is 9.6%. It is clear that the profile in figure 3(a) gives better coincidence between experimental data and calculation.

The profile analysis around $(200)_{Fe}$ and $(220)_{MgO}$ was performed using the same procedure as for figure 3. Figure 4(a) shows the result where d-values are fixed to the values determined by the fitting presented in figure 3(a). The R-factor determined is 8.9%. The result where d-values are fixed to the corresponding bulk values is also presented in figure 4(b), and the R-value is 14.6%. The diffraction profile around the $(200)_{Fe}$ line also provides clear evidence of the lattice expansion of the Fe lattice.

4. Remarks

Fe atoms in the Fe(16 Å)/MgO(60 Å) multilayered film, prepared in UHV conditions, have been identified as having the same structure as the bulk specimen, and lattice dilation of Fe atoms in the film was observed from analysing the observed diffraction profiles of $(110)_{Fe}$ and $(200)_{MgO}$ lines. The enhancements in magnetization and hyperfine field of the same films as in this experiment have been found to be 234 emu g^{-1} and 345 kOe at 4.2 K, respectively; compare the bulk values of 222 emu g^{-1} and 338.5 kOe at 4.2 K. Details of this experiment will be reported elsewhere (Koyano *et al* 1991). The lattice dilation is, therefore, the principal origin of the enhancements in magnetization and hyperfine field. On the other hand, it is known that the magnetization of α-Fe depends on its atomic volume. The pressure experiments showed that the magnetization (Kondorskii and Seldov 1960) and hyperfine field (Williamson *et al* 1972) decrease with increase in pressure. The energy band calculation also predicted that the magnetization is enhanced when the atomic volume is enlarged (Anderson *et al* 1977, Moruzzi *et al* 1986). The lattice parameter corresponding to the observed magnetization can be calculated (Moruzzi *et al* 1986) and corresponds to a 2% expansion at normal pressure.
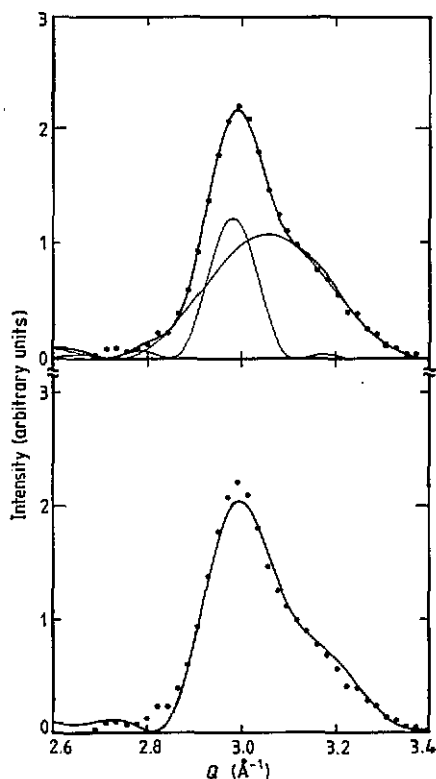
Figure 3. The results of least-squares fitting (——) and the experimental data (●) for the $(110)_{Fe}$ and $(200)_{MgO}$ diffraction profiles. (*a*) The result of least-squares fitting where *d*-values of Fe(110) and MgO(200) are treated as free parameters. The broader subspectrum is the $(110)_{Fe}$ line and the narrower one is the $(200)_{MgO}$ line. (*b*) Least-squares fitting profile, where the *d*-values are fixed to those corresponding to bulk values.

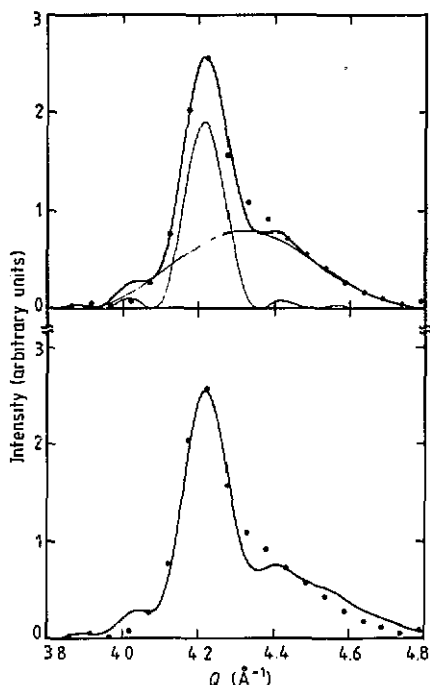**Figure 4.** The fitted (——) and measured (●) profiles for the $(200)_{Fe}$ and $(220)_{MgO}$ diffraction profiles. (*a*) The fitted profile where the *d*-values are fixed to the values obtained from the fitting shown in figure 3(*a*). The broader subspectrum is the $(200)_{Fe}$ line and the narrower one is the $(220)_{MgO}$ line. (*b*) The fitted spectrum under the assumption that the lattice spacings were equal to the corresponding bulk values.

The observed dilation of 1% is considered to correspond to the half-value of that calculation for bulk α-Fe.

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