Magnetic Critical Behavior of γ -Fe/Cu Multilayer

Y. Yamada, B. Sadeh, C. Lee, M. Doi, and M. Matsui

Dept. of Mat. Sci. and Engin., Nagoya Univ., Nagoya, 464-8603, Japan

(Received ; June 8, 1998 Accepted ; August 26, 1998)

Ferromagnetic moment, a magnetic critical exponent β and Curie temperature (T_C) of γ -Fe/Cu multilayer (0.8 to 7.2 ML Fe thickness) prepared by the molecular beam epitaxy (MBE) method have been investigated. The crystal structure of γ -Fe layer is distorted tetragonally ($c/a > 1$) for the Fe thickness less than 4.5 monolayer (ML) and cubic for thicker films. Magnetization (M) of γ -Fe is increased with decreasing Fe layer thickness because of the increase of the ratio of high spin state γ -Fe atoms to total Fe atoms at interfaces. A critical exponent in a relationship $M/M_s = (1 - T/T_C)^\beta$, is estimated to be 0.30 ± 0.02 . T_C is decreased with decreasing Fe layer thickness. The behavior is explained by taking into account of the number of nearest neighbor Fe atoms around an Fe atom.

Key words : γ -Fe, multilayer, epitaxial growth, magnetic critical exponent, Curie temperature, MBE method.

1. Introduction

With the development of the thin film fabrication or microprocessing techniques, low dimensional magnetic materials such as ultra thin films, micro-wires, or magnetic micro-dots have been recently investigated. These materials are considered to be new materials for breaking through the ultra high-density magnetic recording technique. In such a practical use, the magnetic phase transition temperature of the low dimensional magnetic material becomes very important. Therefore, in this paper, magnetic phase transition of γ -Fe/Cu multilayers with ~ 1 to 7 ML (monolayer) in Fe layer thickness has been investigated as a low dimensional magnetic material.

Many investigations on the critical phenomenon near the Curie temperature (T_C) have been reported from both sides of theoretical and experimental standpoints so far. The magnetization (M) as a function of temperature near T_C is generally expressed by

$$\frac{M}{M_s} \propto \left(1 - \frac{T}{T_C}\right)^\beta, \quad (1)$$

where β is a critical exponents which depends on the dimensionality of the material, and M_s is the saturation magnetization at 0 K. Representative theoretical values of β are, $\beta = 0.125$ in the two dimensional Ising model¹⁾ and $\beta = 0.5$ in the three dimensional molecular field approximation. On the other hand, experimentally, the β values reported previously for two dimensional systems are 0.81 for the surface of Ni thin film grown on GaAs(100)²⁾, 0.127 for the surface of Fe thin film on Pd(100)³⁾, 0.25 $\sim$ 0.43 for the Ni/Cu(100) multilayer⁴⁾ and so on⁵⁻¹⁰⁾.

In the present γ -Fe/Cu multilayer, Fe layer with 0.8 to 7.2 ML in thickness is considered to be two dimensional magnetic layer. The multilayer is, however, also considered to be three dimensional system, because the Fe layer is sandwiched by Cu layers. Consequently, from the viewpoint of dimensionality, the critical exponent of the γ -Fe/Cu multilayer is very interesting, since it does not belong to the category studied until now.

2. Experimental Procedure

Multilayer films were made using the MBE method in a

pressure of lower than 5×10^{-11} Torr. Substrate was a MgO (001) single crystal. After annealing the MgO at 773 K, Cu was deposited 200 Å at 673 K as a seed layer and 800 Å at 423 K as a buffer layer. On the Cu buffer layer, [t ML Fe/ 14.2 ML Cu], ($t=0.8-7.2$, $n=10,30,50$) multilayer was prepared, where 1 ML = 1.8 Å. The substrate temperature during film deposition was 330 K. Fe was deposited by EB-gun and Cu by K-cell. Typical deposition rates were ~ 1.5 Å/min for Fe and ~ 3.0 Å/min for Cu. The film thickness and deposition rate of Fe were controlled using a quartz thickness monitor, and those of Cu were controlled by deposition time using experimental results for temperature vs. film growth rate.

Reflection high-energy electron diffraction (RHEED) was utilized to confirm the epitaxial growth and to estimate the lattice spacing in the film plane (a). Artificial periodicity and the lattice spacing normal to the film plane (c) were evaluated by X-ray diffraction.

Magnetization was measured by a vibrating sample magnetometer (VSM) and a superconducting quantum interference device (SQUID). The magnetization in this paper is expressed by the magnetization of Fe per gram, emu/g. The weight of Fe was evaluated by the volume of Fe layers and the density of the γ -Fe estimated from lattice constants (Fig.1, Fig.2), where the Fe layer thickness was evaluated by the positions of satellites in the X-ray diffraction pattern.

3. Results and Discussion

3.1 Structural Characterization

RHEED pattern of Cu buffer layer was observed as a "streaky" one with face centered symmetry. The same pattern was observed for Fe layer deposited on Cu. The results indicate the epitaxial layer-by-layer growth of γ -Fe on Cu (001). Now, the artificial periodicity and sharp interfaces of multilayer were confirmed by the sharp low angle peaks and middle angle satellites in the X-ray diffraction pattern.

Fig. 1 shows the lattice spacing (a) of Fe (200) plane as a function of Fe thickness. The spacing a is independent of Fe thickness and equal to a Cu bulk value within error limits. The lattice spacing normal to

Fig. 1 Lattice spacing in the film plane (a) as a function of Fe thickness.

Fig. 2 Lattice spacing normal to the film plane (c) as a function of Fe thickness. The results of samples on Cu (001) substrates are also shown.

Fig. 3 Saturation magnetization at 4.4 K and 77 K as a function of Fe thickness.

the film (c) evaluated by a conventional step model¹¹⁾ is shown in Fig. 2 as a function of Fe thickness. In this figure, the results for films deposited on a Cu (001) bulk single crystal substrate are also shown. Since the spacing c is increased with decreasing Fe layer thickness, the crystal structure of γ -Fe layer is distorted tetragonally ($c/a > 1$) for the Fe thickness less than 4.5 ML and cubic for thicker films.

3.2 Magnetic Critical Behavior

Fig. 3 shows the saturation magnetization (M_s) as a function of Fe thickness. The magnetization of γ -Fe is increased with decreasing Fe layer thickness. It should be noted that the magnetization of 0.8 ML Fe is estimated to be 221 emu/g at 4.4 K, which means no magnetic dead layer. The thickness dependence of magnetization in Fig. 3 is explained by considering the ratio of high spin state Fe atoms to the total Fe atoms, where the magnetization of the high spin state remains unchanged. The number of high spin state Fe atoms is increased with decreasing Fe thickness because of the increase of the ratio of Fe atoms at interfaces, where the Fe atoms near Cu atoms at interfaces are easy to take the high spin state. The explanation is consistent with previous works¹²⁾⁻¹⁷⁾ and theoretical calculation¹⁸⁾. Now, the rapid increase of magnetization over 7 ML in Fig. 3 is due to the transformation of γ -Fe to bcc α -Fe.

Fig. 4 shows the magnetization M as a function of temperature (M - T curve). The temperature region measured for thinner Fe layer films than 1.2 ML and that for thicker films were different. Before the

Fig. 4 Magnetization as a function of temperature. Magnetization was measured from 4.4 K to RT for about 1 ML thickness ((a)). Magnetization of thicker films, was measured from 77 K to 873 K ((b)).

estimation of T_C , the attention should be paid to the high temperature behavior in Fig 4(b). The magnetization increases at about 650 K and it decreases rapidly over 700 K for all samples measured. The X-ray diffraction experiment for samples annealed at several temperatures for two hours around the peak of M - T curve at high temperatures revealed that the increase of M at about 650 K is due to the transformation of γ to α phase of Fe layer and the rapid decrease of M over 700 K is come from the oxidation of the film. It is furthermore revealed that the two hours annealing below 600 K make the film structure including the artificial periodicity unchanged. Accordingly, the magnetization below about 600 K was used to estimate the Curie temperature T_C and the critical exponent β in equation (1).

In order to determine T_C and β , the $\log M$ vs. $\log (1 - T/T_C)$ plot was used. The exponent β is deduced by choosing a T_C that maximizes the range of a straight-line fit in the plot. Fig. 5 shows the log-log plot for the indicated data from Fig. 4. The critical exponent β was 0.30 ± 0.02 which was almost independent of Fe thickness. It seems that the independence is due to the small change of Fe thickness in the present samples, 0.8 to 5.9 ML, while β generally depends on the dimensionality. It should be emphasized that β obtained for the present samples is an intermediate value between values of two and three dimensional systems, because the present samples include not only very thin two dimensional magnetic layers but also Cu layers between Fe layers, which make the film approach to the three dimensional system.

Fig. 6 shows T_C as a function of Fe thickness. The Curie temperature T_C of γ -Fe decreases with decreasing Fe thickness. In order to explain the tendency, we adopted a brief expression for T_C by Weiss model. According to the Weiss theory, T_C is given by the following equation, $T_C = 2zJS(S+1)/3k_B$, where z is the

Fig. 5 Log-log plot of magnetization data as a function of temperature from Fig. 4.

number of nearest neighbor Fe atoms around an Fe atom, J is the exchange integral, S is a quantum number of total spin and k_B is the Boltzmann constant. Accordingly, T_C is proportional to z directly, if J and S do not depend on Fe thickness. We assumed that the spin S of the high spin state, this is magnetization, is not changed by the Fe thickness as described above. The number z in the γ -Fe/Cu(001) multilayer is expressed by $z = 12 - 8/n$, where n is Fe thickness (ML). The dashed line in Fig.6 indicates the calculated T_C using the z value, where the least mean square fitting was made. The line is in good agreement with experimental data. Therefore, it is concluded that T_C depends on z and T_C of high spin state of γ -Fe bulk can be estimated to be about 650 K by means of an extrapolation of T_C to $z=12$.

4. Conclusion

In this paper the crystal structure and magnetic properties of epitaxial γ -Fe/Cu multilayers have been demonstrated. The crystal structure of γ -Fe layer was distorted tetragonally ($c/a > 1$) for the Fe thickness less than 4.5 ML and cubic for thicker films. Magnetization of γ -Fe was increased with decreasing Fe layer thickness because of the increase of the ratio of high spin state γ -Fe atoms at interfaces. The critical exponent β was estimated to be 0.30 ± 0.02 , which is intermediate value between values of two and three dimensional systems. The Curie temperature T_C was decreased with decreasing Fe layer thickness. The behavior is explained by taking into account of the

Fig. 6 The Curie temperature as a function of Fe thickness. The dotted line is the relationship between T_C and Fe thickness supposing that T_C was proportional to the number of nearest neighbor Fe atoms (in detail, see text).

number of nearest neighbor Fe atoms around an Fe atom. The Curie temperature T_C of high spin state of γ -Fe was estimated to be about 650 K

Acknowledgements

A part of the present study was supported by Ministry of Education, Science, Sports and Culture under Grant-in-Aid for Scientific Research on Priority Areas, Japan (No. 09NP1201, No. 6499223).

Reference

- 1) J. Kohlhepp, H. J. Elmers, S. Cordes, and U. Gradmann : *Phys. Rev. B* **45**, 12287 (1992).
- 2) S. F. Alvarado, M. Campagna, F. Ciccacci, and H. Hopster : *J. Appl. Phys.* **53**, 7920 (1982).
- 3) C. Liu and S. D. Bader : *J. Appl. Phys.* **67**, 5758 (1990).
- 4) F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis : *Phys. Rev. B* **49**, 3962 (1994).
- 5) J. A. Davis : *J. Appl. Phys.* **36**, 3520 (1965).
- 6) E. Kisker, K. Schröder, W. Gudat, and M. Campagna : *Phys. Rev. B* **31**, 329 (1985).
- 7) C. Rau, K. Kuffner : *J. Magn. Magn. Mat.* **54-57**, 767 (1986).
- 8) W. Dürr, M. Taborelli, O. Paul, R. Germar, W. Gudat, D. Pescia, and M. Landolt : *Phys. Rev. Lett.* **62**, 206 (1989).
- 9) U. Gradmann : *J. Magn. Magn. Mat.* **100**, 481 (1991).
- 10) Ch. V. Mohan and H. Kronmüller : *J. Magn. Magn. Mat.* **182**, 287 (1998).
- 11) Y. Fujii, T. Ohnishi, T. Ishihara, Y. Yamada, K. Kawaguchi and N. Nakayama : *J. Phys. Soc. Jpn.* **55**, 251 (1986).
- 12) M. Doyama, M. Matsui, H. Matsuoka, S. Mitani, and K. Doi : *J. Magn. Magn. Mat.* **93**, 374 (1991).
- 13) S. Mitani, A. Kida, and M. Matsui : *J. Magn. Magn. Mat.* **126**, 76 (1993).
- 14) M. Matsui, S. Mitani, and A. Kida : *New Functionality Materials Vol. C 125* Elsevier Sci. Pub, (1993).
- 15) M. Matsui, A. Kida, Y. Kamada, and Y. Yamada : *J. Magn. Soc. Jpn.* **19**, 767, (1995).
- 16) A. Kida, and M. Matsui : *J. Phys. Soc. Jpn.* **65**, 1409, (1996).
- 17) Y. Yamada, K. Ohtani, and M. Matsui : *to be published*.
- 18) T. Asada and S. Blügel : *Phys. Rev. Lett.* **79**, 507 (1997).