Temperature dependence of the magnetization in Fe/Au multilayers

1992 J. Phys.: Condens. Matter 4 8163

(http://iopscience.iop.org/0953-8984/4/41/011)

J. Phys.: Condens. Matter 4 (1992) 8163–8168. Printed in the UK

Temperature dependence of the magnetization in Fe/Au multilayers

H Yamazaki and T Shinjo
Institute for Chemical Research, Kyoto University, Uji, Kyoto 611, Japan

Received 19 May 1992, in final form 6 July 1992

Abstract. The temperature dependence of the magnetization of Fe/Au multilayers was investigated using a SQUID magnetometer in the temperature range from 10 to 90 K with an external field of 10 kOe. The samples prepared by vacuum deposition consist of the bilayers [Fe(10 Å)/Au(d)]$_n$, where d = 20, 50 or 100 Å and the number n of bilayer repetitions is from 1 to 12. The temperature dependence closely obeys the relation $1 - M(T)/M(10\ \mathrm{K}) = CT^{\alpha}$ and, as the number n increases, the exponent α increases from 1.10 ($n = 1$) to 1.55 ($n = 10$) for $d = 50$ Å. The crossover behaviour of α as a function of n is qualitatively explained by a simple spin-wave model.

1. Introduction

Recently, the temperature dependence of the magnetization in multilayers composed of magnetic and non-magnetic layers has been investigated to make clear the dimensional-crossover behaviours. Bloemen *et al* [1] (using Co/Pt and Co/Au multilayers) and Gutierrez *et al* [2] (using Fe/Ag multilayers) direct their attention mainly to the dependence of magnetization–temperature (M–T) curves on the thicknesses of the non-magnetic layer(s) and/or the magnetic layer(s). Here we focus our attention on the dependence of M–T curves on the number n of bilayer repetitions of the Fe/Au multilayers. So far, the n-dependence of magnetic properties has not been systematically studied. We investigate the crossover behaviour from the dependence of the M–T curves not on the Fe layer thickness but on the number of bilayer repetitions, in other words the number of Fe layers, with fixed thickness of each Fe layer. It has been widely recognized that an ideal two-dimensional (2D) magnet is hard to prepare. Even when one finds a linear dependence of M on T in thin magnetic layers at low temperatures, one cannot conclude the sample to be a 2D magnet without resolving the problem of clustering. We expect, however, that clustering and inhomogeneities, even if they exist in Fe layers, do not change drastically when multiple Fe/Au bilayers are utilized, so that the crossover behaviour dependence on n is considered to be almost free from imperfections in the samples.

2. Experimental details

A series of Fe/Au multilayers was prepared in an ultra-high-vacuum (10^{-9} Torr) system using electron beam evaporation. A typical evaporation rate was 0.2 Å s^{-1}, and the substrates were kept at room temperature. Polyimide substrates were used for the magnetization measurements and glass substrates for the x-ray diffraction

0953-8984/92/418163+06$04.50

study. For *in-situ* rate and thickness control, a quartz oscillator was used. After a buffer layer of Au with a thickness of 250 Å had been deposited on the substrate, bilayers [Fe(10 Å)/Au(d)]$_n$ of Fe and Au with the repetition number n = 1–12, where d = 20, 50 or 100 Å, were deposited. In order to avoid oxidation at the surface, an overlayer of 100 Å Au was further deposited. The artificial superstructure of the samples was confirmed by x-ray diffraction in the small-angle region. Figure 1 shows the x-ray diffraction patterns for the [Fe(10 Å)/Au(50 Å)]$_{12}$ sample. The diffraction peaks were observed up to the fourth peak and the superstructure period of 60 Å was confirmed. The crystal structure of each component was also studied by x-ray diffraction in the medium-angle region. It was shown that BCC Fe(110) planes and FCC Au(111) planes are preferentially oriented in the film plane, which has also been shown for Fe/Au multilayered films [3] and Au/Fe/Au sandwiches [4]. Marlière *et al* [4] reported that deposited Fe films with a thickness ranging from 10 to 200 Å have the bulk BCC structure with a (110) plane parallel to the (111) surface plane of Au crystals and the free surface roughness is constant when the Fe thickness is thinner than 15 Å. This is the reason that the Fe layer thickness in our samples was set to 10 Å in order to make the thickness as small as possible, keeping the BCC crystal structure. The 10 Å thickness corresponds to a thickness of about 5 monolayers (MLS).

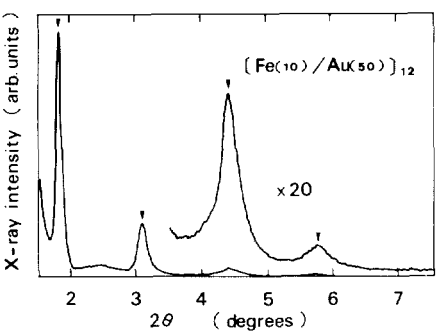

Figure 1. The x-ray diffraction patterns in the small-angle region for the [Fe(10 Å)/Au(50 Å)]$_{12}$ sample.

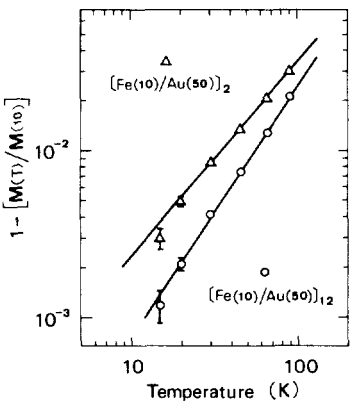

Figure 2. Temperature dependence of $1 - M(T)/M(10\ \mathrm{K})$ for the [Fe(10 Å)/Au(50 Å)]$_2$ (△) and [Fe(10 Å)/Au(50 Å)]$_{12}$ (○) samples plotted on a log–log scale: ——, results of least-squares fitting.

The temperature dependence of the magnetization was measured by the SQUID magnetometer in the temperature range from 10 to 90 K with an external field of 10 kOe applied in the film plane. At each temperature, measurements were repeated ten times to obtain a precise average value with a small standard deviation. The M–H curves for Fe/Au multilayered films indicate that the magnetization is sufficiently saturated under a 10 kOe field [5].

3. Results

The observed data were fitted to the equation

$$1 - M(T)/M(10\ \mathrm{K}) = CT^{\alpha} \tag{1}$$

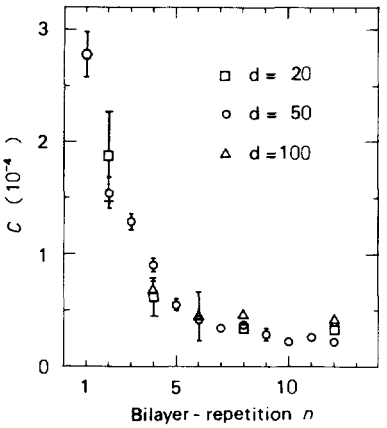

Figure 3. The dependence of the coefficient C on the number n of bilayer repetitions.

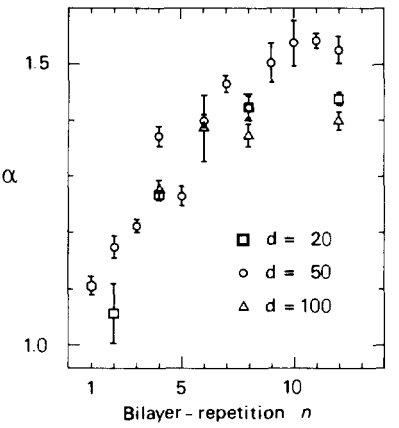

Figure 4. The dependence of the exponent α on the number n of bilayer repetitions.

where $M(T)$ is the saturation magnetization at T K, and C and α are constants characteristic of each sample. A curve of $1 - M(T)/M(10\ \mathrm{K})$ versus temperature was plotted on a log–log scale, and C and α were evaluated from a least-squares fitting. Figure 2 shows the examples $[\mathrm{Fe}(10\ \text{Å})/\mathrm{Au}(50\ \text{Å})]_2$ and $[\mathrm{Fe}(10\ \text{Å})/\mathrm{Au}(50\ \text{Å})]_{12}$. We can see that the temperature dependence closely obeys equation (1) in this temperature range. The evaluated C and α as functions of n are shown in figure 3 and figure 4, respectively.

The significance of the coefficient C is not clear. It is known that, even in the case of bulk samples, the experimental value of C is not in agreement with the theoretically calculated value based on the spin-wave approximation. In figure 3, however, a marked dependence on n can be seen. Roughly speaking, it seems that C is inversely proportional to n. For the variation in α in figure 4, we can see a monotonic increase with increasing n as a whole for the series $d = 20$, 50 and 100 Å. The crossover from 2D-like ($\alpha \simeq 1$) to 3D-like ($\alpha \simeq 1.5$) behaviour can be recognized. It is also seen that the variation in α with n depends on the Au thickness d, particularly for $n \geqslant 8$. For $d = 20$ Å samples the value of α saturates at 1.44 ± 0.02 ($= \alpha_s^{20}$), for $d = 50$ Å it saturates at 1.53 ± 0.06 ($= \alpha_s^{50}$) and for $d = 100$ Å it saturates at 1.40 ± 0.04 ($= \alpha_s^{100}$). This means that the temperature dependence of the magnetization is affected by the thickness of the non-magnetic layer between magnetic layers. In the case of Fe/Au/Fe trilayers, we previously studied the dependence of the interlayer coupling strength between Fe layers on the Au layer thickness by means of ferromagnetic resonance and found that the interlayer coupling extends to a rather long distance [6]. We can say that the variation in α with d is attributed to the variation in the interlayer coupling strength between Fe layers. However, the intermediate value α_s^{20}, i.e. $\alpha_s^{100} < \alpha_s^{20} < \alpha_s^{50}$, is hard to understand because one expects the behaviour of the multilayers to become more 3D-like as d decreases and the coupling strength between Fe layers increases. One explanation is that in fact, for $n \leqslant 12$, the value of α for $d = 20$ Å is not saturated and, for larger n-values, one would find a saturation value of α larger than α_s^{50}. For $d = 20$ Å, we have investigated only four samples ($n = 2$, 4, 8 and 12) which are not enough to confirm the explanation.

The $n = 1$ sample shows a nearly linear temperature dependence of M as if it is a 2D magnet. It has been reported that Ni–Fe alloy films show nearly 3D behaviour even when the thickness is reduced to a thickness of a few MLs [7]. It is hard to distinguish whether superparamagnetism is the origin of the linear dependence in our study or not. For the study of n-dependence, however, we assume that the $n = 1$ sample is a quasi-2D magnetic system.

4. Discussion

By way of a trial, a simplified spin-wave model is introduced to explain the observed n-dependence of α. The essential idea is the same as that previously discussed by Klein and Smith [8] and Glass and Klein [9] for thin ferromagnetic films. In our treatment, however, we introduce two kinds of exchange integral: intralayer J in an Fe layer and interlayer J' between Fe layers. Further, although each Fe layer has a thickness of 5 ML and clustering problems have not been resolved, we assume that each Fe layer behaves as a single 2D layer.

Let us assume a single layer which contains $N \times N \times N_z$ spins, where N and N_z are the x (or y) and z dimensions of the crystal in units of the lattice parameter. We take the z axis to be perpendicular to the film plane hereafter. The spin-wave approximation carried out to the first order shows that $\epsilon_{x,y} \sim SzJ(\pi/N)^2$ and $\epsilon_z \sim SzJ(\pi/N_z)^2$, where ϵ_i is the first excitation energy of spin waves propagating in the i direction, S the spin quantum number, z the number of nearest neighbours and J the exchange integral. When we consider an $N \gg N_z$ (thin-layer) case, we obtain $\epsilon_{x,y}/\epsilon_z \ll 1$. This means that the spin waves normal to the film plane can be hardly excited at low temperatures and the layer behaves like 2D magnetic material.

Next we deal with the case of the multilayer in which the thin magnetic layers are separated by non-magnetic layers. We take J' as the exchange integral between two magnetic layers separated by non-magnetic layers. If $J' \ll J$ holds, we get $\epsilon'_z \ll \epsilon_z$, where ϵ'_z is the excitation energy for which spin waves propagate normal to the film plane through the multilayer and at the same time in each magnetic layer the wavevector component normal to the film plane is zero. This means that we can regard the multilayer as a system composed of 1 ML magnetic layers separated by non-magnetic layers. From this point of view, the excitation energy ϵ'_z can be expressed as $\epsilon'_z \sim Sz'J'(\pi/n)^2$, where z' is the number of nearest neighbours in the nearest layers and n the bilayer repetition or the number of magnetic layers. The above discussion does not hold when J' is comparable with J. In this case, we have to take the thickness of the magnetic layers into consideration.

Now we assume a lattice which has $2N^2n$ spins located on its lattice sites, where we take $N = 10^7$ in the calculation and n corresponds to the number of Fe layers in real samples. It has been confirmed that the size of N does not affect the calculation results when N is larger than 10^4. The planes which contain $2N^2$ spins are assumed to be (110) planes of BCC structure as in the actual samples. We obtain the dispersion relation

$$E_{(K)} = 2SzJ\{1 - \tfrac{1}{3}[\cos(\tfrac{1}{2}K_x)\cos(\tfrac{1}{2}K_y) + \cos(\tfrac{1}{2}K_x) + \cos(\tfrac{1}{2}K_y)]\} + 2Sz'J'(1 - \cos K_z) + g\mu_B H_{\text{eff}} \quad (2)$$

where K_i is the ith component of the spin-wave vector, g the g-factor, μ_B the Bohr magneton and H_{eff} the effective field including both the external and the anisotropy

field. In calculations, the parameters are taken as follows: $S = 1$, $z = 8$, $z' = 2 - 2/n$, $J = 2.76 \times 10^{-21} J$ (bulk value) and $g = 2$. The expression for z' the effective number of nearest neighbours along the z axis, is obtained with the assumption of the end effect from the outer layers. Only J' and H_{eff} are variable in obtaining the experimental results. The reduction in the magnetic moment at low temperatures is

$$1 - \frac{M}{M_0} = \frac{1}{N_A S} \sum_{K} \frac{1}{\exp(E_{(K)}/k_B T) - 1} \tag{3}$$

where N_A is the total number of spins ($N_A = 2N^2 n$) and k_B the Boltzmann factor. The cosines as a function of K_x or K_y in equation (2) are expanded to first order (long-wavelength approximation) and equation (3) is calculated numerically. The calculation is the same as that in the ordinary spin-wave approximation [8, 9].

To compare the calculated results with the experimental results, the values of C and α were evaluated as follows. First, the value of $1 - M/M_0$ is calculated as a function of temperature, and then C and α are evaluated using the least-squares fitting to the relation $1 - M/M_0 = CT^{\alpha}$. Figure 5 shows a representative example of evaluating C and α for $n = 1$ and 12. The full circles represent the results calculated from equations (2) and (3) and the full lines the results of least-squares fitting. We can see that the calculated temperature dependence of $1 - M/M_0$ also closely obeys the relation $1 - M/M_0 = CT^{\alpha}$. The evaluated α as a function of n and J' are shown in figure 6. For C, we can only say that the calculated and the observed values are of nearly the same magnitude. In this calculation, H_{eff} is tentatively kept constant at 0.5 kOe for all the n; this hypothesis may not hold in real multilayers. In addition, one difficulty exists in that, if we set $H_{\text{eff}} = 10$ kOe in accordance with the applied field, the evaluated α for $n = 1$ becomes 1.34 which is very different from the observed value (1.10). The discrepancy in the value used for H_{eff} is the main problem in the theoretical explanation of the data. This may be attributable to interface anisotropy which plays an important role in thin magnetic films and/or to clustering or inhomogeneities in the Fe layer. The variation in α observed in experiments (see figure 4) can be qualitatively explained by $J' \simeq J/50$ for $d = 50$ Å and $J' \simeq J/200$ for $d = 100$ Å. For $d = 20$ Å, however, it is difficult to explain the variation by any J'. This may be due to the inappropriate approximation in which the thickness of the Fe layer is disregarded and is thought to be 1 ML. When J' is comparable with J, this approximation is not appropriate as mentioned before, but $\alpha = 1.05$ for $n = 2$ suggests the case where J' is not very weak (about $J/10$) in figure 6.

5. Conclusion

The Fe/Au multilayers composed of the bilayers [Fe(10 Å)/Au(d)]$_n$, where $n = 1$–12 and $d = 20$, 50 or 100 Å, were prepared by the ultra-high-vacuum evaporation technique. The temperature dependence of the magnetization was measured using a SQUID magnetometer, and the values of C and α were evaluated by least-squares fitting to the relation $1 - M(T)/M(10\text{ K}) = CT^{\alpha}$. It was found that the exponent α increases with increasing n and is also affected by the Au layer thickness. The coefficient C was found to be approximately inversely proportional to n. The simple spin-wave model in which each Fe layer is assumed to be a 2D magnet was able

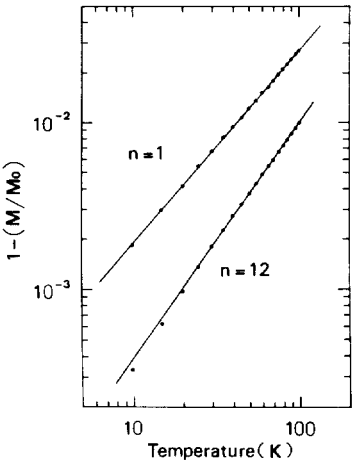

Figure 5. An example of evaluating C and α from the calculated results (●) using equations (2) and (3): ——, results of least-squares fitting. The parameters used in calculation are $J' = J/50$ and $H_{\text{eff}} = 0.5$ kOe.

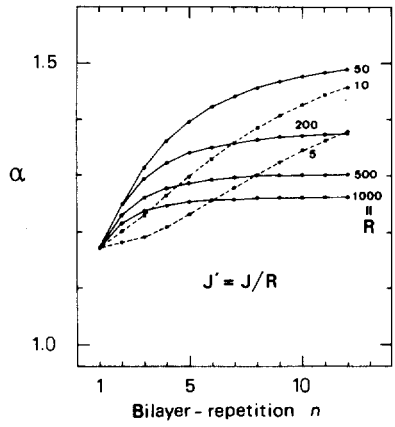

Figure 6. Theoretically evaluated α as a function of n and J' for $H_{\text{eff}} = 0.5$ kOe by a simple spin-wave approximation.

to explain the variations in α qualitatively except for the results for $d = 20$ Å. The exchange integrals J' between Fe layers were estimated to be $J' \simeq J/50$ for $d = 50$ Å and $J' \simeq J/200$ for $d = 100$ Å, where J is the exchange integral in Fe layers. The n-dependence of the magnetic properties in weakly interacting Fe thin layers were partly elucidated although a more elaborate study is required.

Acknowledgments

The authors thank Dr Y Ajiro for a discussion on the whole study. The authors also thank Professor T Tonegawa and Dr M Kaburagi, Kobe University, for helpful discussion on the spin-wave approximation. This work is supported by a Grant-in-Aid for Scientific Research on Priority Areas from the Ministry of Education, Science and Culture.

References

[1] Bloemen P J H, de Jonge W J M and den Broeder F J A 1991 *J. Magn. Magn. Mater.* **93** 105
[2] Gutierrez C J, Qiu Z Q, Wieczorek M D, Tang H and Walker J C 1991 *J. Magn. Magn. Mater.* **93** 326
[3] Araki S, Takahata T, Dohnomae H, Okuyama T and Shinjo T 1989 *Mater. Res. Soc. Symp. Proc. 151* (Pittsburgh, PA: Materials Research Society) p 123
[4] Marlière C, Chauvineau J P and Renard D 1990 *Thin Solid Films* **189** 359
[5] Shintaku K and Shinjo T 1992 *J. Magn. Magn. Mater.* submitted
[6] Yamazaki H, Ajiro Y, Hosoito N and Shinjo T 1991 *J. Phys. Soc. Japan* **60** 764
[7] Gradmann U and Müller J 1968 *Phys. Status Solidi* **27** 313
[8] Klein M J and Smith R S 1951 *Phys. Rev.* **81** 378
[9] Glass S J and Klein M J 1958 *Phys. Rev.* **109** 288