Theoretical Calculation of Magnetic Properties of Ultrathin Fe Films on Cu(100)

R. E. Camley

Department of Physics, University of Colorado at Colorado Springs, Colorado Springs, Colorado 80933-7150

Dongqi Li

Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

(Received 13 October 1999)

The temperature dependence of the magnetization in fcc Fe on Cu(100) is calculated using a self-consistent local mean-field theory. The model reproduces an experimental magnetization oscillation as a function of film thickness and supports a picture where the top two layers are ferromagnetically coupled, and the remaining layers are antiferromagnetically coupled. The origin of the puzzling linear temperature dependence in oscillation amplitude is understood as a "surface phenomena" of the antiferromagnetic layer at the Fe/Cu interface. Proximity effects between a thin antiferromagnet with a low Néel temperature and a neighboring ferromagnet with a higher Curie temperature are discussed.

PACS numbers: 75.70.Ak, 75.25.+z, 75.50.Ee

Ultrathin face-centered Fe film on Cu(100) is the single most challenging and rich magnetic film system to date due to its structural and magnetic instabilities [1–18]. While fcc Fe is normally a high-temperature phase, face-centered phases of Fe can be stabilized via epitaxial growth on a fcc substrate like Cu(100). These structures are inevitably influenced by many factors such as lattice mismatch, growth energetics, film morphology, interdiffusion, etc., to form subtly different, metastable face-centered phases [1–5]. Meanwhile, fcc Fe is predicted by first-principle theories as magnetically unstable [6–12]. Several metastable phases of high-spin and low-spin ferromagnetic (FM) phases, antiferromagnetic (AF) phases, and nonmagnetic phases exist with a total energy difference in the order of meV. a 1%–5% variation in atomic volume and/or tetragonal distortion, which is compatible with the subtle structural differences of the Fe phases on Cu(100), is enough to switch among the various magnetic phases.

With an extensive amount of experimental work, the various descriptions for the behavior of Fe on Cu(100) are converging. Most of the recent work agrees on the following for thermally deposited films: (a) The face-centered phases exist below a critical thickness of 10–12 monolayers (ML), above which the structure relaxes into the regular bcc phase [1,4,13]. (b) The low-temperature grown films have face-centered tetragonal (fct) structure and are FM [13,14]. (c) For the films grown above ~250 K, the same fct, FM phase is observed below 5 ML [2,13]. (d) Between 6–11 ML, only the top two layers have the tetragonal expansion [1]. Ferromagnetic ordering is observed at the surface below 260 K [1,13]. The inner layers have an isotropic fcc structure [15].

The magnetic properties of the inner layers of the 6–11 ML of fcc Fe, however, remain controversial. Mössbauer studies observed an AF phase with low moment ($\sim 0.6\mu_B$) and low Néel temperature T_N (~67 K). This is the same magnetic phase observed in fcc bulk Fe precipitates in the Cu matrix. In contrast, *ab initio* calculations for fcc Fe/Cu(100) consistently suggest ferromagnetic coupling between the top two layers only, and a type-l AF phase with a sizable magnetic moment ($\sim 1.2\mu_B$ to $1.8\mu_B$) for the inner layers [8,9,11]. An experimental study [13] agrees with this picture by finding that between 6–11 ML, the Kerr signal oscillates as a function of film thickness, which is interpreted as a type-l antiferromagnet with alternating layers of spins and a T_N of ~200 K. The existence of this AF phase was later supported by a spin-polarized low-energy electron diffraction study [17] and AF coupling in Fe/Ni/Cu(100) [18]. So far almost all the theoretical efforts have concentrated on ground state properties. The current work discusses the temperature dependence of the magnetic structure and accounts for the unique behavior of the data. Our theory reproduces all the major experimental results, giving strong support to a picture where the first two layers are coupled ferromagnetically, and the remaining layers are all antiferromagnetically coupled.

A number of unusual features were noted in the data of Ref. [13]. The size of the Kerr oscillation was the largest at low temperature and decreased as the temperature increased. If one plotted the Kerr oscillation amplitude $\Delta M(T)$, which is proportional to the difference in the total magnetic moment between even- and odd-layer systems, it showed a distinct linear behavior with temperature. Such a linear behavior is usually associated with a surface effect, and yet this measurement involved the magnetic moment of the entire sample. Furthermore the total magnetic moment of the sample $M(T)$, thought to be primarily from the surface ferromagnetic phase, showed a temperature dependence which looked much like a bulk material. In other words, surface features show a bulklike temperature dependence, and conversely bulk features show an apparent surfacelike temperature dependence.

The experiments reported a number of other interesting features. The transition temperature associated with the

0031-9007/00/84(20)/4709(4)$15.00

antiferromagnet, T_N, was found to be about 200 K, a value larger than that of bulk γ Fe precipitates in Cu. The transition temperature for the ferromagnet, T_c, occurs at a different temperature of 260 K.

In this paper we present a theoretical calculation for the magnetic structure of Fe on Cu(100). Our theoretical results agree very well with the experiments and give insight into the origin of these unexplained behaviors. In particular, we find the following: (1) The difference in the total magnetic moment, i.e., the oscillation amplitude, has a linear temperature dependence in agreement with the experiments. This linear dependence is, indeed, associated with a surface effect, and we identify it as essentially measuring the magnetic moment of the outermost antiferromagnetic layer at the Fe/Cu interface. (2) The total magnetic moment as a function of temperature has a behavior close to that of a bulk material, in agreement with the experimental data. (3) The Néel temperature of the antiferromagnet can be enhanced by coupling to ferromagnetic surface layers.

Our theoretical calculations are based on a self-consistent local mean-field theory [19]. This method has been applied in a number of systems and has given a good description of both the temperature and field behavior of magnetic layered structures [20–22]. We use a Heisenberg-type Hamiltonian for the system that uses an effective exchange coupling between spins S_i in layer i and S_{i+1} in the neighboring layer. Only couplings between nearest neighbor layers are included. A small external field H_0 is also included for stability. Thus

$$H = -\sum_{\text{layer } i} A_{i,i+1}\mathbf{S}_i \cdot \mathbf{S}_{i+1} - \sum_{\text{layer } i} g\mu_B \mathbf{H}_0 \cdot \mathbf{S}_i .$$

The parameters used are $S = 1$ and $g = 2.2$. The exchange coupling constants are given in terms of temperature. The ferromagnetic coupling between the first two layers is found to be $A_{1,2}/k_B = 360$ K, while the antiferromagnetic coupling between all other layers is taken as $A_{i.i+1}/k_B = -130$ K. To put these values into perspective, the ferromagnetic exchange coupling for bulk (bcc) Fe is about 530 K.

The exchange parameters in this system are not well known. From the hyperfine field found in Mössbauer experiments [23] on Fe/Cu multilayers, we can estimate that a spin in the ferromagnetic phase of fcc Fe ($T_c = 150$ K) has an exchange energy in the range of $E/k_B = 100$–200 K. First-principle calculations, again for superlattices [24], show antiferromagnet exchange energies on the order of $E/k_B = 300$ K. Our choice of parameters was based initially on the experimentally observed values of T_N and T_c, but these were modified by finite size effects. For example, for the ferromagnet the Curie temperature would indicate an exchange constant of $A/k_B = 193$ K for a bulk sample. Because of finite size effects we need to use an increased value of $A/k_B = 360$ K.

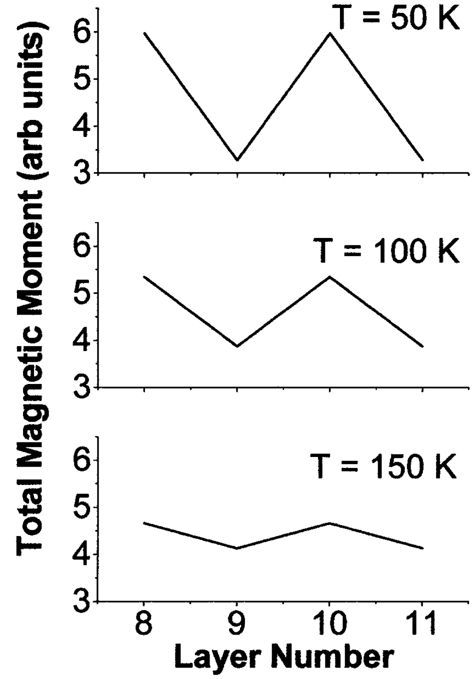

FIG. 1. Total magnetic moment of structure as a function of the number of layers for different temperatures. Note that, in agreement with experiment, the oscillation size remains the same, independent of the layer number.

An iterative method is used to find the ground state for the Hamiltonian as a function of temperature and applied field. In Fig. 1(a) we show the results for the magnetic moment as a function of thickness for different temperatures. All the key features of the experiment are reproduced. First we note that the oscillation amplitude is independent of the number of layers, i.e., $\Delta M(T)$ is the same if the difference is taken between M at 8 and 9 monolayers or between 10 and 9 monolayers.

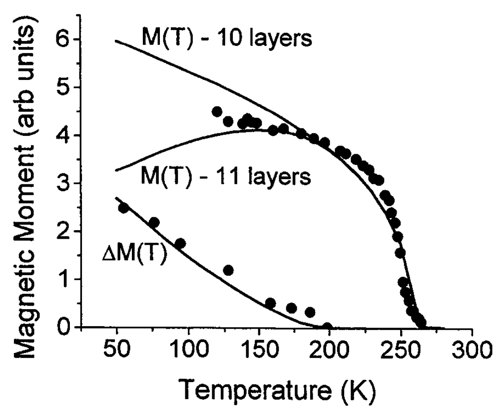

FIG. 2. Temperature dependence of $M(T)$ and $\Delta M(T)$. Note that $\Delta M(T)$ is nearly linear in temperature as is also found in the experiment. The dots represent the experimental data from Ref. [12]. For $M(T)$ the data for odd and even numbers of layers are averaged. The vertical scales for $\Delta M(T)$ and $M(T)$ are slightly different.

Second, the oscillation amplitude clearly decreases as temperature is increased. In Fig. 2 we show the temperature dependence of $M(T)$ and of $\Delta M(T)$. Again, the theoretical results agree very well with the experimental ones. In particular, $\Delta M(T)$ shows a linear decrease with temperature, and $M(T)$ shows a bulklike temperature dependence. This is strong supporting evidence that the correct magnetic structure is one where the first two Fe layers are ferromagnetic, and the remaining layers are antiferromagnetically coupled. We note that mean-field theory, of course, provides only an approximate description of $M(T)$, and that it is not expected to give the correct form near phase transitions.

There is an unusual temperature dependence found in the theoretical calculation for the 11-layer structure seen in Fig. 2. This structure has an odd number of layers for the antiferromagnet, and at low temperature the antiferromagnet provides a net moment which is opposite to the ferromagnetic layers. As the temperature increases the antiferromagnet reaches its transition temperature and no longer provides a moment opposite to the ferromagnet. As a result the total magnetization initially increases with T, only to decrease at even higher temperatures as the ferromagnet reaches its transition temperature. The experimental temperature dependence of the valley in the Kerr oscillation with film thickness (not shown) agrees with the theoretical results.

We now explore what additional insight the theory can provide. Perhaps the most interesting question is—can we explain the linear dependence on temperature for $\Delta M(T)$? In Fig. 3 we plot the magnetic moment of each layer for a 10-layer system and an 11-layer system at several fixed temperatures. For low temperatures the moments are essentially the same *except for the outermost layer.* Thus the difference in magnetic moments for the two structures simply measures the temperature dependent magnetic moment of the last layer in the antiferromagnet. Since this is a surface spin, it has the typical linear reduction in moment as temperature is increased as can be seen in Fig. 4. At higher temperatures finite size effects start to play a larger role, but even for temperatures as high as $T = 150$ K, the primary contribution to $\Delta M(T)$ still is from the outermost layer.

Figure 4 shows why the total magnetic moment as a function of temperature appears to have bulk-like characteristics for $T > 200$. In this region, the total magnetization is basically a function of the magnetic moment of the first two ferromagnetically coupled layers. These are the spins that see the largest effective fields and therefore have the highest transition temperatures. As we can see in Fig. 4, both layer 1 and layer 2 show bulklike behavior for magnetization as a function of temperature. At higher temperatures where the contribution of the antiferromagnet is small this then means the entire sample also shows a bulklike behavior.

To complete our study of the ferromagnet/antiferromagnet structure we examine how the order parameter depends on site index and temperature in Fig. 5. At low temperatures the moments are all at their maximum value. As the temperature is increased, the ferromagnetically coupled spins (layers 1 and 2) are more resistant to thermal fluctuations and retain most of their magnetic moment. A bulklike antiferromagnetic state is evident in the flat regions for temperatures below 150 K. (The Néel temperature for a bulk antiferromagnet with our parameters is about 180 K. For an uncoupled 10-layer antiferromagnet we find $T_N = 165$ K.) Finite size effects, however, influence this state, and the thermal moments are substantially reduced at the outer edge. This becomes progressively more important as temperature is increased.

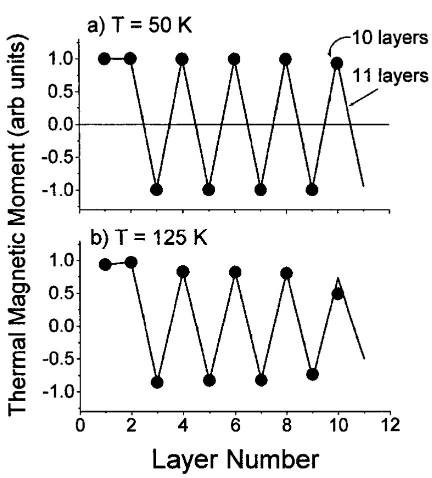

FIG. 3. Thermal averaged magnetic moment of each layer for two different temperatures. The dots show the results for a 10-layer system, while the solid line represents the results for an 11-layer system. At low temperatures the difference in the magnetization comes from the outermost layer only. Even at higher temperatures (125 K), the measured value of $\Delta M(T)$ comes primarily from the outer spin.

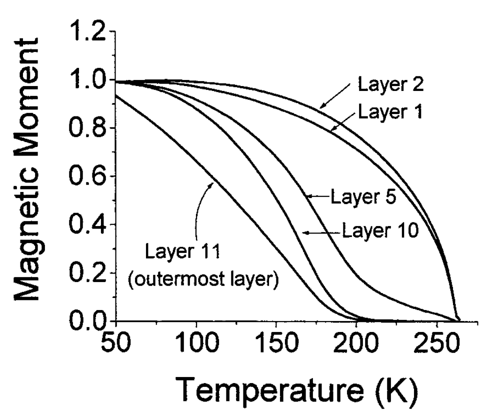

FIG. 4. Thermal averaged magnetic moment for different layers as a function of temperature. Note that the outermost layer has a nearly linear temperature dependence. The ferromagnetic layers (1 and 2) show a much more bulklike magnetization curve.

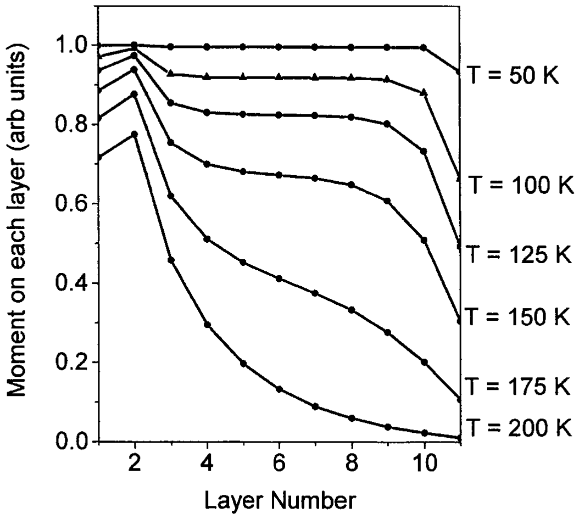

FIG. 5. Magnitude of the thermal averaged magnetic moment for each layer for several different temperatures. The ferromagnetic layers are layer numbers 1 and 2, and all the remaining layers represent the antiferromagnet. Note that above the Néel temperature (about 175 K) the antiferromagnet spins are stabilized only by the coupling to the ferromagnet.

Finally at higher temperatures the existence of an ordered state in the antiferromagnet region is dependent on the coupling to the stable ferromagnetic layers. Here we see a simple exponential-like decay of the magnetic moments with distance. We note that the thermal stabilization of a thin antiferromagnet with a low Néel temperature by a neighboring antiferromagnet with a high Néel temperature has been reported for antiferromagnetic superlattices of NiO/CoO [25] and for FeF_2/CoF_2 [26]. Here we have a case of an antiferromagnet being stabilized by a ferromagnet.

Based on first-principle calculations [6–12,24], magnetic moments should be layer dependent, especially at the Fe-Cu interface. Experimentally one also finds that the moments in the AF phase are generally smaller [27]. Changing the AF moment to $S = 1/2$ for the AF layers and increasing the exchange coupling (so that the net effective field acting on the AF spins remains about the same), however, does not alter the general features of our results. Magnetic coupling should also be layer dependent. A weaker coupling or moment at the Fe-Cu interface should make this outer AF layer even softer. Nonetheless, we obtain the general features with the two parameters we currently use, and there is not enough experimental input to uniquely determine more parameters.

In summary, we have examined theoretically the magnetic properties of a model structure for fcc Fe on Cu(100). The model—the first two layers are ferromagnetically coupled and the remaining ones are antiferromagnetically coupled—agrees well with all the experimental results. Furthermore, the theoretical calculations have shown the origin of the puzzling linear temperature dependence of $\Delta M(T)$ found originally in the experimental results.

This work was supported by ARO Grant No. DAAG55-98-0294 and DOE BES-Materials Sciences No. W-31-109-ENG-38.

[1] J. Thomassen *et al.*, Phys. Rev. Lett. **69**, 3831 (1992).
[2] S. Müller *et al.*, Phys. Rev. Lett. **74**, 765 (1995).
[3] M. Zharnikov *et al.*, Phys. Rev. Lett. **76**, 4620 (1996).
[4] J. Shen *et al.*, Europhys. Lett. **43**, 349 (1998).
[5] H. Magnan *et al.*, Phys. Rev. Lett. **67**, 859 (1991).
[6] V. L. Moruzzi, P. M. Marcus, and J. Kübler, Phys. Rev. B **39**, 6957 (1989).
[7] S. S. Peng and H. J. F. Jansen, Phys. Rev. B **43**, 3518 (1991).
[8] C. L. Fu and A. J. Freeman, Phys. Rev. B **35**, 925 (1987).
[9] T. Kraft, P. M. Marcus, and M. Scheffler, Phys. Rev. B **49**, 11 511 (1994).
[10] T. Asada and S. Blügel, Phys. Rev. Lett. **79**, 507 (1997).
[11] G. W. Fernando and B. R. Cooper, Phys. Rev. B **38**, 3016 (1988).
[12] R. Lorenz and J. Hafner, Phys. Rev. B **54**, 15 937 (1996); **58**, 5197 (1998).
[13] Dongqi Li *et al.*, Phys. Rev. Lett. **72**, 3112 (1994).
[14] D. P. Pappas, K. P. Kämper, and H. Hopster, Phys. Rev. Lett. **64**, 3179 (1990).
[15] W. A. A. Macedo and W. Keune, Phys. Rev. Lett. **61**, 475 (1988); R. D. Ellerbrock *et al.*, *ibid.* **74**, 3053 (1995).
[16] J. F. Cochran *et al.*, Phys. Rev. B **45**, 4676 (1992).
[17] Ch. Würsch *et al.*, Phys. Rev. B **55**, 5643 (1997).
[18] B. Schirmer and M. Wuttig, Phys. Rev. B **60**, 12 945 (1999).
[19] R. E. Camley and D. R. Tilley, Phys. Rev. B **37**, 3413 (1988).
[20] R. E. Camley, J. Kwo, M. Hong, and C. L. Chien, Phys. Rev. Lett. **64**, 2703 (1990).
[21] A. S. Carriço and R. E. Camley, Phys. Rev. B **45**, 13 117 (1992).
[22] M. Sajieddine *et al.*, Phys. Rev. B **49**, 8815 (1994).
[23] D. J. Keavney *et al.*, Phys. Rev. Lett. **74**, 4531 (1995).
[24] Yu-mei Zhou, Wen-tong Geng, and Ding-sheng Wang, Phys. Rev. B **57**, 5029 (1998).
[25] E. N. Abarra *et al.*, Phys. Rev. Lett. **77**, 3451 (1996).
[26] C. A. Ramos *et al.*, Phys. Rev. Lett. **65**, 2913 (1990).
[27] D. Schmitz *et al.*, Phys. Rev. B **59**, 4327 (1999).