Session: BP-08, 46th MMM Conference, Seattle, Washington, November 12–16, 2001

Enhanced Fe Moment at Pd/Fe Interfaces Studied by Low-Temperature Conversion Electron Mössbauer Spectroscopy

L. Cheng, Z. Altounian* , D. H. Ryan, and J. O. Ström-Olsen

McGill University, Centre for the Physics of Materials, Department of Physics, Rutherford Physics Building, 3600 University St., Montreal, Quebec, H3A 2T8 Canada

Abstract

Conversion electron Mössbauer spectroscopy (CEMS) has been employed to study the magnetic moment of Fe in proximity to Pd at the interfaces of [Pd 53 Å/Fe t Å]$_{25}$ multilayers grown by DC magnetron sputtering (t was varied from 5 Å to 40 Å). At room temperature, all of the multilayers were ferromagnetically ordered. Analysis gives an interface Fe layer thickness of $\sim$ 2 monolayers. The ground state hyperfine field (B_{hf}), extracted from the temperature dependence of B_{hf}, was found to be as high as 38.9 T, corresponding to a ground state Fe moment of 2.8 μ_B.

PACS numbers: 68.55.Jk 75.70.Cn

Typeset using REVTEX

*Corresponding author: Tel.: 1 514 398 6535; Fax: 1 514 398 6525; e-mail: zaven@physics.mcgill.ca.

I. INTRODUCTION

Theoretical predictions of novel properties for magnetic surfaces and interfaces have stimulated much interest in experimental studies of magnetic multilayers. The commonest systems incorporate Fe, since it has a large magnetic moment and is suitable for Mössbauer studies which provide information on the local environment, magnetic ordering, and the Fe moment. Studies have shown that the magnetic properties of ultra-thin Fe layers are modified by the reduced dimension and are strongly influenced by the material in contact. The magnetism of Fe in proximity to Pd is one of the most intriguing problems because Pd is nearly ferromagnetic. Polarization of Pd by Fe (or Co) due to 3d-4d interactions leads to the well-known giant Fe (or Co) moment in dilute Fe (or Co) alloys with Pd [1,2]. The interaction between Fe and Pd at the 2D interface also plays an important role in the magnetic coupling between Fe across Pd layers [3–6], the distribution of the polarization of the Pd [3,7,8] and the Fe moment. According to calculations on a Fe/Pd(001) system, the enhancement in the Fe moment can be 0.6 $\mu_{\rm B}$ [6] – 1 $\mu_{\rm B}$ [9]. However, only a few attempts have been made to measure the Fe moment in the Pd/Fe system [10–13], among which Mössbauer studies provided contradictory information. Depth-profiling Mössbauer spectroscopy by Hosoito *et al.* [10] showed that about 30 % of the Fe in 3.5 Å probe layers at the Pd interface was paramagnetic at room temperature (RT) and exhibited a reduced B_{hf} of 31.5 T at 4.2 K, while the rest of the Fe had an enhanced B_{hf} of 36.5 T at 4.2 K. In the room temperature converson electron Mössbauer spectroscopy (RT-CEMS) studies by Boufelfel *et al.* [11] and Li *et al.* [12], no paramagnetic component was observed, and the B_{hf} of interface-like Fe was larger than that of bulk Fe. In order to clarify these discrepancies and further investigate the ground state moment of Fe at the interface with Pd, low temperature CEMS has been employed in this study.

II. EXPERIMENTAL TECHNIQUES

The [Pd 53 Å/Fe t Å]$_{25}$ (t = 5, 10, 15, 20, 30, and 40 Å) multilayers, named S_t for simplicity, were deposited on a Si(100) substrate at ambient temperature using DC magnetron sputtering. The base pressure prior to sputtering was 2.0×10^{-7} Torr, and the Ar pressure during sputtering was 7 mTorr. The deposition rates, monitored by quartz-crystal sensors and calibrated using x-ray reflectometry measurements, were 1 Å/s and 2 Å/s for Fe and Pd respectively. Pd and Fe layer thichnesses were obtained by fits to x-ray reflectometry profiles. CEMS spectra were collected using a He+10%CH_4 gas-flow proportional counter installed in a nitrogen-flow cryostat, and analyzed using a nonlinear least-squares fitting routine using Lorentzian lines with a Gaussian distribution of hyperfine fields.

III. RESULTS AND DISCUSSION

The CEMS spectra of the multilayers taken at room temperature (RT) (Fig. 1) show ferromagnetic six-line patterns. The line shape of S_5 is very broad, but no pronounced paramagnetic component is present in the spectrum, which is in agreement with Boufelfel *et al.* [11] and Li *et al.* [12], but in conflict with Hosoito *et al.* [10]. By general consensus, a 5 Å of Fe layer grown by sputtering breaks into an island-like structure. The absence of superparamagnetic components in the S_5 spectrum suggests an indirect exchange interaction of Fe through magnetically polarized Pd atoms. For the S_{15}, S_{20} and S_{30} spectra, lines 1 and 2 are slightly sharper and stronger than lines 5 and 6, so two sextets with different B_{hf}'s and isomer shifts (δ) relative to α-Fe were assumed which when superimposed on each other reproduce the observed asymmetry in the line shape. One sextet with vanishing δ and sharper lines (with a field width of $\sim$ 0.8 T) is attributed to a bulk-like Fe site, while the other with a positive δ and wider lines (with a field width of $\sim$ 1.5 T) is attributed to an interface-like Fe site. The sextets of interface-like Fe, which are depicted by dashed lines in Fig. 1b), have a relative area that decreases with the total Fe thickness. If convert the relative areas of the subspectra to Fe thicknesses by multiplying the relative area of interface-like Fe by the total Fe thicknesses, the interface-like Fe thickness is found to be constant at 8 ± 1 Å per Fe layer. If the interface-like Fe is distributed equally on each side of the Fe layer, then 2.0 ± 0.3 monolayers (ML) of Fe are interface-like at each Pd/Fe interface. The δ's of the interface-like Fe are in the range of 0.08 – 0.16 mm/s, which suggests a lower S electron density at the Fe nucleus at the Pd/Fe interface with respect to bulk Fe. The quadrupole splittings of all samples are negligible. The interface-like B_{hf}'s are ~ 33 T, while the bulk-like B_{hf}'s are slightly reduced. There is no dramatic change in either δ, or B_{hf} as the Fe thickness is reduced, so our study does not show evidence of the bcc to fcc transition claimed by Li *et al.* [12]. Since metastable fcc Fe is very unlikely to be stabilized up to 40 Å, the Fe in our multilayers is most likely bcc in all samples.

For all the multilayers except S_5, the Fe moment is in the sample plane, evident by the intensity ratios of 3:4:1:1:4:3 in all the spectra or subspectra. S_5 has a ratio of 3:3.7:1:1:3.7:3, corresponding to a slight ($12^o \pm 7^o$) out of plane tilt. This strong in-plane anisotropy has been found to persist even in 1 ML of Fe grown on Pd(001), unless the Fe was deposited at low temperatures [14]. This anomalous in-plane anisotropy was attributed to interfacial alloying during RT depositions [15,16]. However, the morphology of the interface has long been a controversial issue, and both non-interdiffused [10,17–19] and interdiffused [11,15,16] interfaces have been reported. In our case, the δ's of the interface-like Fe are generally lower than that of Fe atoms in Pd, which is 0.177 ± 0.002 mm/s [20]. Though the possibility of alloy formation can not be totally ruled out, at least the Pd-Fe alloying effect is insignificant in our Pd/Fe multilayers and the interdiffusion between Pd and Fe must be less than 2 ML.

In order to obtain the ground state hyperfine fields of interface-like Fe and study the evolution of B_{hf} with temperature for both interface and bulk-like Fe, CEMS spectra of S_{10} and S_{15} were taken from 90 K to 296 K. Using the RT spectrum as a guide, one-sextet and two-sextets were used to fit the spectra of S_{10} and S_{15} respectively. As shown in Fig. 2. The errors of the B_{hf}'s of S_{10} do not permit the form of the temperature to be determined. While for S_{15}, the B_{hf}'s of both subspectra follow a $T^{3/2}$ relationship. By extrapolating $B_{hf}(T)$

to 0 K, the ground state hyperfine field ($B_{hf}(0)$) of S_{10} is 38.9 $\pm$ 0.1 T assuming a linear temperature dependence, and 37.8 $\pm$ 0.1 T for a $T^{3/2}$ dependence. For S_{15}, the $B_{hf}(0)$'s are 36.4 $\pm$ 0.1 T and 34.3 $\pm$ 0.2 T for the bulk-like and interface-like Fe respectively.

The final problem is how to relate B_{hf} to the local moment. In the bulk, the major contribution to B_{hf} is the negative polarization field (B_{cp}) of the core electrons due to the d electrons via the Fermi contact interaction. B_{cp} is approximately proportional to the magnetization, so is B_{hf}, which is the basis of the empirical interpretation of B_{hf}. However, at a surface or interface, the contributions from the conduction electron (B_{ce}) and the dipolar field (B_{dip}), which are strongly dependent on the local environment, might become prominent. As a result, B_{hf} might not scale with the magnetization the same way as in the bulk. A detailed calculation of the electronic structure and B_{hf} structure is necessary to evaluate the Fe moment at the Pd/Fe interface. As this type of calculation is not available, we can only present qualitative arguments. First of all, the T or $T^{3/2}$ dependence of B_{hf} resembles the characteristic temperature dependence of the spontaneous magnetization of 2D ferromagnets. In addition, magnetometry and CEMS studies of 1 ML Fe(110) on W(110) [21] have demonstrated that B_{hf} and the magnetization follow the same temperature dependence, though the Fe exhibited an enhanced moment but a reduced B_{hf} with respect to bulk Fe. So it is still plausible to assume that B_{hf} represents the temperature dependence of the magnetization of Fe at the Pd/Fe interface in our multilayers. For S_{15}, the spin stiffness parameters obtained from fits to the $B_{hf}(T)$ are 1 and 2 times larger than that of bulk Fe for the bulk-like and interface-like Fe respectively. Secondly, according to calculations for Fe at a clean surface [22], and interfaces of Fe/Ag(001) [23] and Fe/W(110) [24], B_{cp} scales approximately with the moment regardless of the local environment with a scaling factor of $\sim$ –13 to –14 T per unpaired spin. Assuming the same scaling factor at the Pd/Fe interface, a 2.8 μ_B moment could give rise to a B_{cp} of –36.4 to –39.2 T, which is very close to the interface B_{hf} obtained in our study. The interface B_{hf} of our multilayers could be corresponding to a Fe moment of 2.8 μ_B, if the sum of B_{ce} and B_{dip} is small compared to the B_{cp}.

IV. CONCLUSIONS

Low-temperature CEMS has shown that the Pd/Fe multilayers contain bcc Fe layers with 2.0 $\pm$ 0.3 ML interface-like Fe at each Pd/Fe interface. The ground state B_{hf} is $\sim$ 14 % larger with respect to that of bulk Fe. If the effective B_{hf} of Fe at the interface is mostly from the polarization of the core electrons, the calculated Fe moment of 2.8 μ_B is consistent with the theoretical prediction [6].

REFERENCES

[1] J. Crangle, Phil. Mag. **5**, 335 (1960).

[2] R. M. Bozorth, P. A. Wolff, D. D. Davis, V. B. Compton, and J. H. Wernick, Phys. Rev. **122**, 1157 (1961).

[3] Z. Celinski, B. Heinrich, J. F. Cochran, W. B. Muir, A. S. Arrott, and J. Kirschner, Phys. Rev. Lett. **65**, 1156 (1990).

[4] Z. Celinski, B. Heinrich, and J. F. Cochran, J. Appl. Phys. **70**, 5870 (1991).

[5] J. R. Childress, R. Kergoat, O. Durand, J.-M. George, P. Galtier, J. Miltat, and A. Schuhl, J. Magn. Magn. Matt. **130**, 13 (1994)

[6] D. Stoeffler, K. Ounadjela, J. Sticht, and F. Gautier, Phys. Rev. B **49**, 299 (1994).

[7] W. Weber, D. A. Wesner, G. Güntherodt, and U. Linke, Phys. Rev. Lett. **66**, 942 (1991).

[8] J. Vogel, A. Fontaine, V. Cros, F. Petroff, J.-P. Kappler, G. Krill, A. Rogalev, and J. Goulon, Phys. Rev. B **55**, 3663 (1997).

[9] S. Blugel, B. Drittler, R. Zeller, and P. H. Dederichs, Appl. Phys. A **49**, 457 (1989).

[10] N. Hosoito, T. Shinjo, and T. Takada, J. Phys. Soc. Jpn. **50**, 1903 (1981).

[11] A. Boufelfel, R. M. Emrick, and C. M. Falco, Phys. Rev. B **43**, 13152 (1991).

[12] M. Li, X. D. Ma, C. B. Peng, J. G. Zhao, L. M. Mei, Y. H. Liu, B. G. Shen, and D. S. Dai, J. Phys.: Condens. Matter **6**, L785-L790 (1994).

[13] X. Le Cann, C. Boeglin, B. Carrière, and K. Hricovini, Phys. Rev. B **54**, 373 (1996).

[14] C. Liu and S. D. Bader, J. Appl. Phys. **67**, 5758 (1990).

[15] C. Liu and S. D. Bader, Phys. Rev. B **44**, 2205 (1991).

[16] C. Boeglin, H. Bulou, J. Hommet, X. Le Cann, H. Magnan, P. Le Fèvre, and D. Chandesris, Phys. Rev. B **60**, 4220 (1999).

[17] C. Binns, C. Norris, Gwyn P. Williams, M. G. Barthes, H. A. Padmore, Phys. Rev. B **34**, 8221 (1986).

[18] J. Quinn, Y. S. Li, H. Li, D. Tian, F. Jona, and P. M. Marcus, Phys. Rev. B **43**, 3959 (1991).

[19] O. Rader, C. Carbone, W. Clemens, E. Vescove, S. Blügel, W. Eberhardt, W. Gudat, Phys. Rev. B **45**, 13823 (1992).

[20] R. Ingalls, F. Van der Woude, and G. A. Sawatzky in *Mössbauer Isomer Shifts* edited by G. K. Shenoy and F. E. Wagner, (North-Holland, 1978) Chapter 7, p365.

[21] H. J. Elmers, G. Liu, and U. Gradmann, Phys. Rev. Lett. **63**, 566 (1989).

[22] S. Ohnishi, A. J. Freeman, and M. Weinert, Phys. Rev. B **28**, 6741 (1983).

[23] S. Ohnishi, M. Weinert, and A. J. Freeman, Phys. Rev. B **30**, 36 (1984).

[24] Soon. C. Hong, A. J. Freeman, and C. L. Fu, Phys. Rev. B **38**, 12156 (1988).

Figure Captions:

Figure 1: The RT-CEMS spectra of [Pd 53 Å/Fe t Å]$_{25}$, t = 5, 10, 15, 20, 30, and 40 Å as labeled in the figures. The vertical lines are experimental data and the solid lines are calculations. For the spectra fitted using two sextets, the dashed lines are calculations for interface-like Fe.

Figure 2: The temperature dependence of the hyperfine fields. The solid lines are for fits using $T^{3/2}$, while the dotted lines are for the fit using linear T dependence. The open squares and solid squares are for the interface-like and bulk-like Fe respectively of S_{15}; the triangles are for the hyperfine fields of S_{10}.

FIGURES

Figure 1:

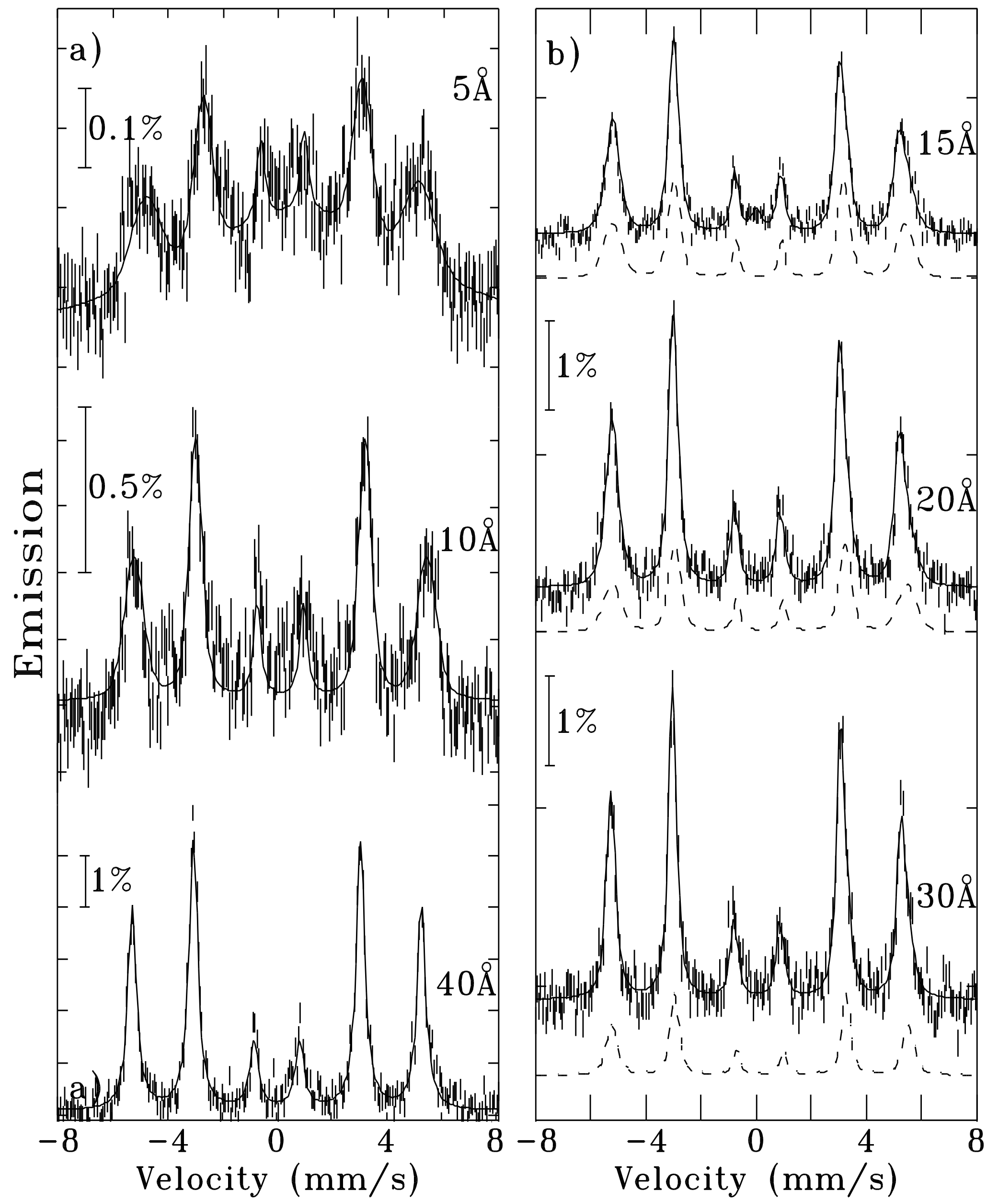

Figure 2: