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PHOTO- AND THERMAL-INDUCED ANTIFERROMAGNETIC
INTERLAYER COUPLING IN Fe/(Fe-Si) SUPERLATTICES*

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We have grown via sputtering, Fe - iron silicide superlattices that exhibit novel thermal and photomagnetic effects. For $\sim 15\text{\AA}$ nominal thickness of the silicide layer the superlattices are antiferromagnetically (AF) coupled at room temperature. Upon cooling below 100K they gradually revert to ferromagnetic alignment, but the AF coupling is restored at low temperature by exposure to visible light of sufficient intensity ($>10\text{ mW/mm}^2$). These effects are due to charge carriers in the silicide spacer layer which, when thermally or photo-generated, are capable of communicating spin information between the Fe layers.

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There is considerable interest in antiferromagnetically coupled trilayers and superlattices that consist of ferromagnetic films coupled across nonmagnetic, metallic spacer layers.¹ The coupling mechanism is thought to result from the polarization of conduction electrons in the spacer layer via an RKKY-like interaction mediated by the band-structure of the spacer.² A spin-dependent quantum-well description of the electronic structure has been proposed.³ The experimental results most readily compared with theory have been the multiple period oscillations of high-quality MBE wedged trilayers,⁴ and related electron-spectroscopy results.^{3,5} Studies of sputtered metallic multilayers have found that oscillatory coupling is a general property of transition metal spacer layers.¹ Recently it has been discovered that AF interlayer coupling is observed in evaporated Fe/Si/Fe trilayers, by Toscano *et al.*⁶ and in sputtered Fe/Si superlattices by Fullerton *et al.*^{7,8} and Mattson *et al.*⁹ found that sputtered Fe/Si superlattices also exhibit AF interlayer coupling. For the superlattices, the antiferromagnetism has been confirmed by polarized neutron reflectivity.^{7,8} The samples of ref. 6 were grown at 40K and the coupling is thought to occur across an amorphous semiconducting Si spacer layer, while for the superlattices in Ref. 7-9, it was shown that the spacer layer consists of crystalline iron silicide. Regardless of the different identifications of the active spacer species, the main point of these studies is that present theories need to be extended to explain coupling across nonmetallic spacers.

In this paper we provide an overview of our recent work on the Fe/Fe-Si superlattice system. We provide new structural analyses of samples that were fabricated to have relatively homogeneous silicide spacers. The homogeneity is accomplished by interleaving 2-4Å thicknesses of Fe and Si to build up the spacer. In our earlier work of Ref. 7 we only deposited Si, and the spacer became a crystalline silicide by interfacial reaction for $t_{\text{Si}} < 20\text{\AA}$. The artificially-formed silicide of the present work (and of Ref. 9) is structurally and compositionally more homogeneous for thicker silicide layers than the naturally-formed interfacial silicides we worked with in Ref. 7. We describe SQUID

magnetometry, magneto-optic and Mössbauer characterizations in addition to the structural analyses. Most importantly we highlight that AF coupling at low temperature can be photo-induced. Since a number of nonmagnetic Fe-Si phases are semi-metals or small-gap semiconductors¹¹⁻¹² we attribute the photo-effects to excitation of carriers into the conduction band. There are many examples of photo-generated phenomena in materials physics *e.g.* photoconductivity¹³ and photodiffusion¹⁴ in amorphous semiconducting Si:H, and photo-enhanced superconducting transition temperatures in oxygen deficient YBaCuO films.¹⁵ The present case adds an example taken from the realm of magnetic superlattices. In our case the photomagnetic effects provide a novel approach to study AF-coupling phenomena, and also could be of technological importance in future optoelectronic devices.

The samples were grown at ambient temperature by dc magnetron sputtering. Sapphire substrates were used throughout with the exception that the transition Mössbauer samples were prepared on Kapton substrates. The samples were made by the alternate sputter deposition of Fe and iron silicide layers with nominal composition FeSi. Each of the silicide layers was made by interdiffusion of alternately deposited 2-Å doses of Fe and 3 or 4 Å doses of Si.

The structural character of the films was determined with low- and high-angle x-ray diffraction (XRD). Low-angle XRD was used to determine the chemical modulation wavelength Λ and to provide an independent determination of the individual layer thicknesses, as shown in Fig. 1. The intercept of the Λ plot in Fig. 1 indicates a 28-Å Fe layer and the slope indicates ~ 3.5 Å of additional thickness for each nominal 3-Å dose of Si added to the spacer. Similar results are obtained with the samples based on 4-Å doses of Si, except that the Fe layer is only ~ 20 -Å thick (indicating some further interdiffusion is taking place in the films) and the additional spacer thickness is ~ 4 Å for each Si dose. In both the 3- and 4-Å Si-dose sets of films the total spacer-layer thickness is approximately given by the total Si thickness.

The structural coherence of the films can be determined from the full-width at half-maximum (FWHM) of the central Bragg peak in the high-angle XRD spectra. A plot of the coherence length ξ vs. Si thickness is shown in Fig. 1. The films retain an ~ 150 -Å coherence length even for spacer layers as thick as 40 Å. The relative constancy of the ξ value is in contrast to what we found in Refs. 7-8 for our Fe/Si multilayers. In that earlier work we found a dramatic decrease in ξ for >20 -Å Si layer thickness. The decrease is due to the formation of an amorphous Si rich composition within the spacer. Thus, by artificially making the silicide spacer layer homogeneous, we can greatly increase the spacer thickness over which the film remains structurally coherent.

SQUID magnetometry was used to probe the magnetic coupling in the films. We show in Fig. 2 the saturation field H_S vs. spacer thickness. AF coupling occurs in the thickness region of 15 - 20 Å for both sets of spacers. This is approximately the same spacer thickness and similar coupling strength as observed in our Fe/Si multilayers.^{7,8} Additionally, as in for the Fe/Si studies, no additional AF regions appear out to spacer thicknesses of 40 Å.

Hysteresis loops for a film that is AF coupled at room temperature are shown in Fig. 3. At low temperature the film becomes more ferromagnetically aligned, as indicated by an increase in the remanence in Fig. 3 at 4.2 K. The increase in remanence could be due to a number of possibilities. For example, if the spacer becomes ferromagnetic upon cooling the adjacent Fe layers could couple directly. Alternatively, if the spacer is semiconducting the freezing out of carriers could effectively uncouple the Fe layers.

Mössbauer spectroscopy was used to determine that the spacer does not become ferromagnetic upon cooling. Transmission Mössbauer spectra at 297 and 4.2 K are shown in Fig. 4 for a $[\text{Fe}(30\text{Å})/\text{FeSi}(17\text{Å})]_{20}$ superlattice. The spectra show both nonmagnetic and magnetically split contributions at all temperatures. The magnetic layers consist of two components: α -Fe (42%), with lesser amounts of $\approx \text{Fe}_{0.85}\text{Si}_{0.15}$ (29%) and other magnetic Fe-Si phases with >15 atomic % Si (5%). 15 at.% is near the solubility limit of Si in α -

Fe.¹⁷ The parameters for the nonmagnetic contribution, which represent the spacer, are given in Table I. The nonmagnetic fractional area shows no significant statistical change upon cooling (24% at 297K and 21% at 4.2K). *Thus, there is no evidence that the spacer becomes ferromagnetic upon cooling.* The isomer shifts and quadrupole splittings provide additional information about the spacer. The room-temperature isomer shift is characteristic of several crystalline Fe-Si phases.^{16,17} Unfortunately, the nonmagnetic spectral component is not sufficiently resolved to accurately determine the quadrupole splitting, but can be fitted with a single broad line, as shown in Fig. 4. The nonmagnetic linewidth increases by nearly 50% on cooling from room temperature to 4.2K, and the isomer shift exhibits the same temperature dependence as that of the magnetic layer. Both results are consistent with previous work on ϵ -FeSi.¹² ϵ -FeSi has an anomalously strong temperature dependence of its quadrupole splitting, with a similar increase of $\sim 50\%$ from room temperature to 4.2K, due to its small semiconducting energy gap of only 0.05eV.¹² This suggests that our observed nonmagnetic component may be related to ϵ -FeSi.

A two-laser pump-probe arrangement was used to explore the effects of incident light on the magnetic coupling. A low-power (25mW) He-Ne laser ($\lambda = 6328\text{\AA}$) was used as the probe to obtain magneto-optic Kerr loops, and a variable power (10-1000mW) Kr (6471 or 5309 $\text{\AA}$) or Ar (5145 $\text{\AA}$) laser was used to illuminate the sample. The unfocused ($\approx 2\text{mm}$ diameter) laser beams were superimposed on the sample. The experiment consists of varying the power of the Kr(Ar) laser and monitoring the magnetization via the Kerr effect using the He-Ne laser. The measurements were performed as a function of temperature using an optical cryostat.

Figure 5 shows Kerr loops for a superlattice with a nominal 17- $\text{\AA}$ FeSi spacer. At room temperature the film is AF coupled and does not noticeably change when irradiated with 100mW of pump power. Upon cooling to 53K the film becomes ferromagnetic; but, most strikingly, *the AF coupling is restored by irradiation with 100mW of pump power.* Every one of the Fe/(Fe-Si) superlattices investigated which exhibited AF coupling at room

temperature, becomes increasingly ferromagnetically aligned (as indicated by an increase in the remanence of the sample) upon cooling, and then recovers its AF coupling upon exposure to visible laser light of sufficient intensity at low temperature ($>10 \text{ mW/mm}^2$). An obvious possibility is to attribute the observed effects to laser heating, but heating is not significant and cannot explain our observations, as we discussed in Ref. 9. Sample heating of only a few Kelvin typically is reported in the literature for Raman experiments with similar laser power densities.¹⁸

An additional feature of importance is that there is a time constant of ~ 10 -60 seconds associated with the photo-induced change in the interlayer magnetic coupling. An example of this effect is shown in Fig. 6 where the degree of AF alignment is monitored as a function of time after the pump laser is turned on. Similar time constants are seen when turning off the pump laser. The existence of long time scale photo-excitation effects have been encountered by researchers in diverse fields, and are attributed, in general, to trapping of electrons or hole in metastable states.¹³⁻¹⁵

We also find that it can take less pump power at 53K than at 75K to restore the AF alignment in some of the films, as shown in Fig. 7. Figure 7(a) shows the evolution of the Kerr loops at 75K as the pump power is increased from 0 to 200mW and returned back to 0mW. It takes $>100\text{mW}$ to completely restore the AF coupling at 75K. Figure 7(b) shows the same measurements performed at 53K and demonstrates that the AF coupling is restored at only $\sim 50\text{mW}$. The last curves of Figs. 7(a) and (b) show that the data superimpose within reasonable limits before and after irradiation.

We believe that the photo-induced AF coupling described above is caused by the excitation of carriers by the radiation. If the mechanism involving conduction electrons proposed for metallic layers is applicable to the present samples, then the strength of the AF coupling should depend on the number of carriers, as is consistent with our observation. If the number of carriers changes significantly, as expected in a semiconductor, the Fermi wavevector also should change, which might alter the period of the coupling. Then if the

semiconducting energy gap is greater than kT , it should be possible either induce or inhibit coupling even at room temperature, an effect we did not observe. The long time constants also seem to be somewhat inconsistent with lifetimes of intrinsic conduction electrons, and are more reminiscent of impurity or defect induced states. Furthermore, the observation in Fig. 7 that lower power is necessary to restore AF coupling at lower temperatures is consistent with there being less thermal energy available to de-excite the trapped electrons or holes. However, until a theoretical model is developed to explain the phenomenon of photo-induced AF coupling, no definite conclusion can be drawn regarding the nature of the states involved.

In conclusion, we highlighted our recent studies that show evidence in Fe/(Fe-Si) superlattices of the restoration of AF interlayer coupling at low temperature by exposure to intense visible light. We present some new structural studies that support our previous work. We suggest that the thermal and photo-induced coupling results from thermally and photo-generated carriers, respectively, which conduct spin information between adjacent ferromagnetic layers.

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REFERENCES:

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1. P. Grünberg, R. Schreiber, Y. Pang, M.B. Brodsky and C.H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986); M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988); S.S.P. Parkin, Phys. Rev. Lett. **67**, 3598 (1991); J.E. Mattson, C.H. Sowers, A. Berger, and S.D. Bader, Phys. Rev. Lett. **68**, 3252 (1992).
 2. Y. Wang, P.M. Levy, and J.L. Fry, Phys. Rev. Lett. **65**, 2732 (1990); D.M. Edwards, J. Mathon, R.B. Muniz, and M.S. Phan, Phys. Rev. Lett. **67**, 493 (1991); P. Bruno and C. Chappert, Phys. Rev. B **46**, 261 (1992); R. Coehoorn, Phys. Rev. B **44**, 9331 (1991).
 3. J.E. Ortega and F.J. Himpsel, Phys. Rev. Lett. **69**, 844 (1992).
 4. J. Ungaris, R.J. Celotta, and D.T. Pierce, Phys. Rev. Lett. **67**, 140 (1991); S.T. Purcell, W. Folkerts, M.T. Johnson, N.W.E. McGee, K. Jager, J. ann de Stegge, W.B. Zeper, W. Hoving, and P. Grünberg, Phys. Rev. Lett. **67**, 903 (1991); Z.Q. Qiu, J. Pearson, A. Berger, and S.D. Bader, Phys. Rev. Lett. **68**, 1398 (1992).
 5. N.B. Brookes, Y. Chang and P.D. Johnson, Phys. Rev. Lett. **67**, 354 (1991).
 6. S.Toscano, B. Briner, H. Hopster, and M. Landolt, J. Magn. and Magn. Mater. **114**, L6 (1992).
 7. E.E. Fullerton, J.E. Mattson, S.R. Lee, C.H. Sowers, Y.Y. Huang, G.Felcher, S.D. Bader, and F.T. Parker, J. Magn. and Magn. Mater. **117**, L301 (1992).
 8. E.E. Fullerton, J.E. Mattson, S.R. Lee, C.H. Sowers, Y.Y. Huang, G.Felcher, S.D. Bader, and F.T. Parker, J. Appl. Phys. **73**, 6335 (1993).
 9. J.E. Mattson, Sudha Kumar, Eric E. Fullerton, S.R. Lee, C.H. Sowers, M. Grimsditch, S.D. Bader, and F. T. Parker, Phys. Rev. Lett. **71**, 185 (1993).

10. Yi-hua Liu, Xiao-ding Ma and Liang-mo Mei, *J. Phys.: Condens. Matter* **3**, 3571 (1991).
11. I. Nishida, *Phys. Rev B* **7**, 2710 (1973).
12. G.K. Wertheim, V. Jaccarino, J.H. Wernick, J.A. Seitchik, H.J. Williams, and R.C. Sherwood, *Phys. Lett.* **18**, 89 (1965).
13. J.-H. Zhou and S.R. Elliot, *Phys. Rev. B* **46**, 12402 (1992); D.L. Staebler and C.R. Wronski, *J. Appl. Phys.* **51**, 3262 (1980).
14. P.V. Santos, N.M. Johnson, and R.A. Street, *Phys. Rev. Lett.* **67**, 2686 (1991).
15. G. Nieva, E. Osuiguil, J. Guimpel, M. Maenhoudt, B. Wuyts, Y. Bruynseraede, M.B. Maple and I.K. Schuller, *Appl. Phys. Lett.* **60**, 2159 (1992); *Phys. Rev. B* **46**, 14249 (1992).
16. G. Marchal, P. Mangin, M. Piecuch, and C. Janot, *J. de Phys. (Paris)* **37**, C6-763 (1976).
17. M. B. Stearns, *Phys. Rev.* **129**, 1136 (1963).
18. M. Boekholt, M. Hoffmann, and G. Güntherodt, *Physica C* **175**, 127(1991).

FIGURE CAPTIONS:

- Fig. 1 Structural parameters of Fe/Fe-Si superlattices. The measured modulation wavelength Λ (a), and crystalline coherence length ξ (b), are plotted vs Fe-Si layer thickness. ξ is obtained from the width of the high-angle diffraction spectra. The solid lines are a guide to the eye.
- Fig. 2 The saturation field H_S vs FeSi layer thickness for films with spacers constructed from dosings of Fe(2Å)Si(3Å) (solid circles) and Fe(2Å)Si(4Å) (open squares), as described in the text.
- Fig. 3 Hysteresis loops for the film [Fe(30Å)/Fe-Si(17Å)]₂₀ at room temperature (upper panel) and 10K lower panel. Note the higher remanence at 10K
- Fig. 4. Transmission Mössbauer spectra for an [Fe(30Å)/Fe-Si(15Å)]₈₀ film. The upper panel shows the spectrum taken at room temperature and the lower panel shows the spectrum taken at 4.2 K. Open circles are experimental data, thin solid lines represent the separate magnetic and nonmagnetic components of the fit to the data, and the thick solid line is the combined fit.
- Fig. 5 Kerr loops for an [Fe(30Å)/Fe-Si(17Å)]₂₀ superlattice: (a) shows the loop at room temperature with no pump-laser power; (b) shows the loop at 53K also with no pump power; (c) shows the loop at 53K with 100mW of pump power and illustrates the photo-induced restoration of the AF coupling.
- Fig. 6 Kerr loops taken at 53K for the [Fe(30Å)/Fe-Si(17Å)]₂₀ sample at the indicated number of seconds after the (200mW) pump laser was turned on.
- Fig. 7 Kerr loops for an [Fe(30Å)/Fe-Si(17Å)]₂₀ superlattice: (a) shows the evolution of the loops at 75K for the indicated pump powers; (b) shows the evolution at 53K.

Table I. Mössbauer parameters for the nonmagnetic spacer phase measured for an $[\text{Fe}(30\text{\AA})/\text{Fe-Si}(17\text{\AA})]_{80}$ film. Numbers in parentheses denote statistical standard deviation.

T (K)	HWHM ^a (mm/s)	Isomer Shift ^b (mm/s)	Nonmagnetic Fraction (%)
4.2	0.57(2)	0.390(8)	21(1)
79	0.49(3)	0.375(4)	22(1)
297	0.38(1)	0.257(5)	24(1)

a) Half-width at half-maximum of the nonmagnetic component in the spectrum.

b) Isomer shift relative to Fe metal at room temperature.

Fig. 1a Mattson et al.

Fig 1b Mattson et al

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Fig. 7 a Mattsson et al

Fig 7b Mattson et al

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