Photoinduced Antiferromagnetic Interlayer Coupling in Fe/(Fe-Si) Superlattices

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We report photoinduced antiferromagnetic (AF) interlayer coupling in sputtered Fe/(Fe-Si) superlattices. The superlattices are intrinsically AF coupled at room temperature and become increasingly ferromagnetically coupled when cooled below 100 K, but the AF coupling is restored at low temperature by exposure to visible light of sufficient intensity (> 10 mW/mm^2). These effects are due to charge carriers in the Fe-Si spacer layer which, when thermally or optically 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 be induced by the polarization of conduction electrons in the spacer layer via an RKKY-like interaction [2]. A spin-dependent quantum-well description of the electronic structure also has been proposed [3]. The most revealing experimental results for comparison with theory have been the multiple period oscillations of high-quality molecular beam epitaxy wedged trilayers [4] and related electron-spectroscopy results [3,5]. Though present theories describe coupling in systems with metallic spacers, it is not clear how these theories can be extended to explain coupling across nonmetallic spacers. Recently, it was discovered by Toscano *et al.* [6], that evaporated Fe/Si/Fe trilayers, and by Fullerton *et al.* [7,8], that sputtered Fe/Si superlattices exhibit antiferromagnetic (AF) interlayer coupling. Also, Liu, Ma, and Mei [9] reported hysteresis loops that are characteristic of AF coupling for $Fe_{0.8}Si_{0.2}$/Si multilayers, but attributed them to perpendicular anisotropy. For the superlattices, the antiferromagnetism has been confirmed by polarized neutron reflectivity [8]. However, AF coupling in the superlattices was observed only for *crystalline* spacer layers [7], while in the trilayers [6] the spacer was claimed to be amorphous semiconducting Si. The crystallinity of the spacer layers in the superlattices was attributed to iron-silicide formation. Mössbauer spectroscopy of the superlattices provides support for this supposition [7,8]. It was also found for these superlattices that the degree of AF coupling decreases dramatically as temperature decreases, an effect which could be due either to the spacer becoming ferromagnetic or to the reduction of thermally activated carriers in a semiconducting spacer.

In this Letter we provide evidence from low-temperature Mössbauer spectroscopy that the spacer does not become ferromagnetic, and we show that AF coupling at low temperature can be photoinduced. Since a number of nonmagnetic Fe-Si phases are semimetals or small-gap semiconductors [10,11], we attribute the photoeffects to excitation of carriers into the conduction band. Although there are many photogenerated phenomena (e.g., photoconductivity [12] and photodiffusion [13] in amorphous semiconducting Si:H and photoenhanced superconducting transition temperatures in oxygen deficient YBaCuO films [14]), the present report is the first example, to our knowledge, of photoinduced changes in the interlayer coupling of magnetic superlattices. Such photomagnetic effects provide not only a novel approach to the study of AF-coupling phenomena, but also could be of technological importance in the optoelectronic industry.

Two sets of samples were grown at ambient temperature on Si and sapphire substrates. The first set was made by the alternate sputter deposition of Fe and Si layers, as discussed in Refs. [7] and [8]. To summarize those results, the samples are AF coupled for nominal Si layer thicknesses of 13–17 Å (with a coupling energy of 0.5 erg/cm^2) and ferromagnetically coupled or uncoupled outside this range. The Si (silicide) layer becomes amorphous for thicknesses ≥ 20 Å. In addition, Mössbauer spectra indicate that the spacer consists of a nonmagnetic $FeSi_{1+x}$ ($x \geq 0$) alloy. The second set of superlattices also was made by sputter deposition and consists of 30 Å Fe layers and Fe-Si layers with nominal composition FeSi; each of these spacer layers was made by alternately depositing ~ 1 atomic layer of Fe and Si. These samples also showed room-temperature AF coupling with the same nominal Si composition as the first set of samples. Although the spacer in these samples remains crystalline for thicknesses ≥ 20 Å, no AF coupling is observed for spacer thicknesses ≥ 20 Å [7,8]. Both high- and low-angle x-ray diffraction superlattice peaks are observed for all AF-coupled samples (from both sample sets) indicating well layered and crystalline films. Resistivity measurements (made using a standard four-terminal, dc tech-

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nique with the current in the plane of the film) exhibit no change between room temperature and 4 K. This is because the Fe layers have much higher conductivity than the Fe-Si and mask any change in the conductivity of the latter. Samples used for Mössbauer experiments were prepared similarly to the ones discussed above, but were thicker and Kapton substrates were used.

Mössbauer spectroscopy was used to determine if the spacer becomes ferromagnetic upon cooling. A ferromagnetic spacer would give direct ferromagnetic exchange coupling of the Fe layers and explain the increased remanence observed at low temperature [7]. Typical transmission Mössbauer spectra at 297 and 4.2 K (for the second set of samples) are shown in Fig. 1. The spectra show both nonmagnetic and magnetically split contributions at all temperatures. The magnetic layers in the first set of samples consist of two components: α-Fe and an $Fe_{85}Si_{15}$ phase [8] which is near the solubility limit of Si in α-Fe. For the second set of samples, in which the Fe-Si spacer was codeposited, the major magnetic component is α-Fe (42%), with lesser amounts of $Fe_{0.85}Si_{0.15}$ (29%) and other magnetic Fe-Si phases with greater than 15 at.% Si (5%). 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 297 K and 21% at 4.2 K). *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 [15]. 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. 3. The nonmagnetic linewidth increases by nearly 50% on cooling from room temperature to 4.2 K, 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 [11]. ε-FeSi has an anomalously strong temperature dependence of its quadrupole splitting, with a similar increase of $\sim$50% from room temperature to 4.2 K, due to its small semiconducting energy gap of only 0.05 eV [11]. This suggests that our observed nonmagnetic component may be related to ε-FeSi; however, this identification needs to be confirmed by direct structural studies which are presently underway.

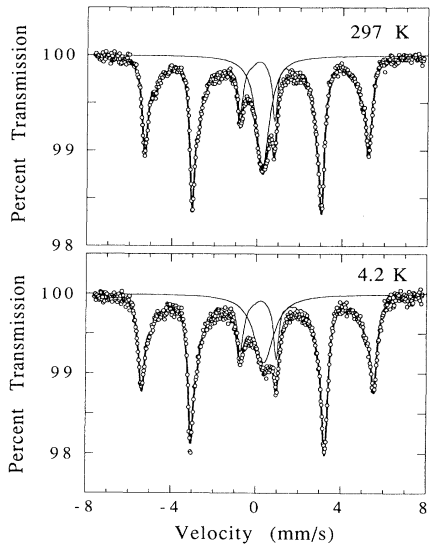

FIG. 1. Transmission Mössbauer spectra for an [Fe(30 Å)/Fe-Si(15 Å)]$_{80}$. The upper panel shows the spectrum taken at room temperature and the lower panel shows the spectrum taken at 4.2 K. Open circles are experimental data, thin solid lines represent the separate magnetic and nonmagnetic components of the fit to the data, and the thick solid line is the combined fit.

TABLE I. Mössbauer parameters for the nonmagnetic spacer phase measured for an [Fe(30 Å)/Fe-Si(17 Å)]$_{80}$ 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.

We used a two-laser pump-probe arrangement to explore the effects of incident light on the magnetic coupling. A low power (25 mW) He-Ne laser (λ=6328 Å) was used as the probe to obtain magneto-optic Kerr loops, and a variable power (10–1000 mW) Kr (6471 or 5309 Å) or Ar (5145 Å) laser was used to illuminate the sample. The unfocused ($\approx$2 mm diam) 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 2 shows Kerr loops for a superlattice with a nominal 17 Å FeSi spacer. At room temperature the film is AF coupled and does not noticeably change when irradiated with 100 mW of pump power. Upon cooling to 53 K the film becomes ferromagnetic; but, most strikingly, *the AF coupling is restored by irradiation with 100 mW of pump power.* Every one of the ten Fe/(Fe-Si) superlattices investigated, which exhibited AF coupling at room temperature, becomes ferromagnetically coupled or uncoupled upon cooling, and recovers its AF coupling upon exposure to visible laser light of sufficient intensity at low temperature (>10 mW/mm^2). An additional feature of importance is that there is a time constant of

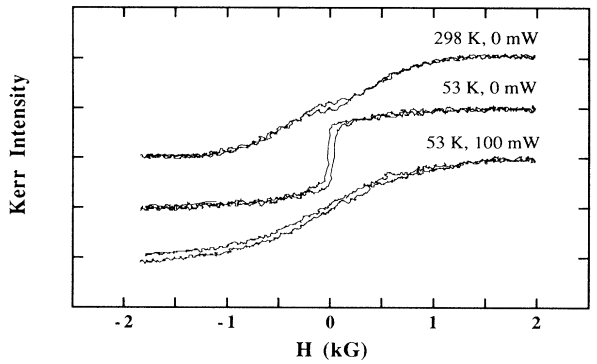

FIG. 2. Kerr loops for an [Fe(30 Å)/Fe-Si(17 Å)]$_{20}$ superlattice. Top, the loop at room temperature with no pump-laser power; middle, the loop at 53 K also with no pump power; bottom, the loop at 53 K with 100 mW of pump power.

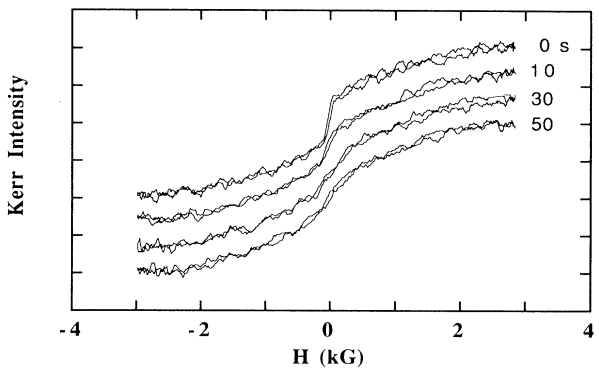

FIG. 3. Kerr loops taken at 53 K for the [Fe(30 Å)/Fe-Si(17 Å)]$_{20}$ sample at the indicated number of seconds after the (200 mW) pump laser was turned on.

$\sim$10–60 s associated with the photoinduced change in the interlayer magnetic coupling. An example of this effect is shown in Fig. 3 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 photoexcitation effects have been encountered by researchers in diverse fields, and are attributed, in general, to trapping of electrons or holes in metastable states [12–14].

Since an obvious possibility is to attribute the observed effects to laser heating, we now discuss why heating is not significant and cannot explain our observations. We find that it takes *less* pump power at 53 than at 75 K to restore the AF alignment in some of the films, as shown in Fig. 4. If laser heating were the cause of the AF coupling, one would expect that the lower-temperature experiments would require more, not less, power. Figure 4(a) shows the evolution of the Kerr loops at 75 K as the pump power is increased from 0 to 200 mW and returned back to 0 mW. It takes $>$ 100 mW to completely restore the AF coupling at 75 K. Figure 4(b) shows the same measurements performed at 53 K and demonstrates that the AF coupling is restored at only $\sim$50 mW. [The last curves of Figs. 4(a) and 4(b) show that the data superimpose within reasonable limits before and after irradiation.] Additionally, we measured a rise of only 10 K when we placed a 2-mil chromel/alumel thermocouple in the laser beam, in agreement with a calculated estimate which yields a maximum temperature rise of $<$ 10 K for 1 W of incident power (based on conservative values of both film and substrate thermal conductivities). Also, sample heating of only a few kelvin is typically reported in the literature for Raman experiments with similar laser power densities [16].

We believe that the photoinduced 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 wave vector also should change, which might alter the period of the coupling; then it should be possible either to 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 intrin-

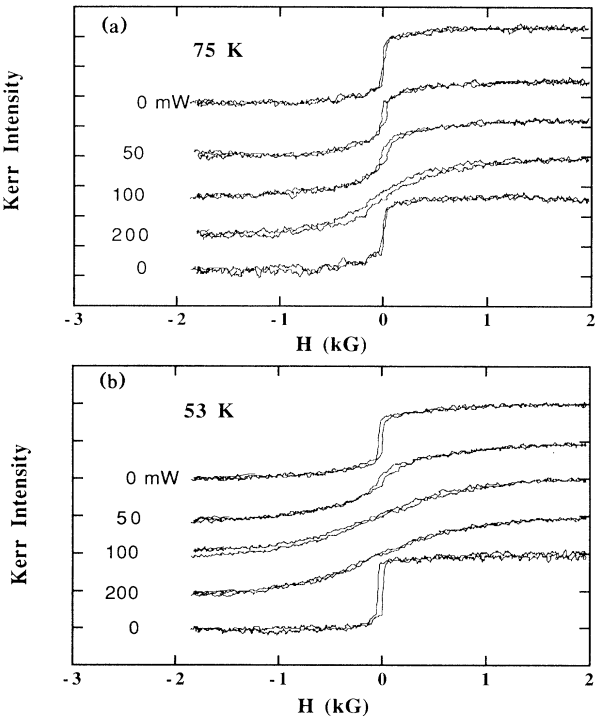

FIG. 4. Kerr loops for an [Fe(30 Å)/Fe-Si(17 Å)]$_{20}$ superlattice: (a) the evolution of the loops at 75 K for the indicated pump powers; (b) the evolution at 53 K.

sic conduction electrons, and are more reminiscent of impurity or defect induced states, further, that lower power is necessary to restore AF coupling at lower temperatures is consistent with there being less thermal energy available to deexcite the trapped electrons (holes). However, until a theoretical model is developed to explain the phenomenon of photoinduced AF coupling, no definite conclusion can be drawn regarding the nature of the states involved.

In conclusion, we present evidence in Fe/(Fe-Si) superlattices of the restoration of AF interlayer coupling at low temperature by exposure to intense visible light. We suggest that this results from photogenerated carriers which conduct spin information between adjacent ferromagnetic layers.

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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. W. 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. 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. 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] Yi-hua Liu, Xiao-ding Ma, and Liang-mo Mei, J. Phys. Condens. Matter **3**, 3571 (1991).

[10] I. Nishida, Phys. Rev. B **7**, 2710 (1973).

[11] G. K. Wertheim, V. Jaccarino, J. H. Wernick, J. A. Seitchik, H. J. Williams, and R. C. Sherwood, Phys. Lett. **18**, 89 (1965).

[12] 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).

[13] P. V. Santos, N. M. Johnson, and R. A. Street, Phys. Rev. Lett. **67**, 2686 (1991).

[14] 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); G. Nieva, E. Osuiguil, J. Guimpel, M. Maenhoudt, B. Wuyts, Y. Bruynseraede, M. B. Maple, and I. K. Schuller, Phys. Rev. B **46**, 14249 (1992).

[15] G. Marchal, P. Mangin, M. Piecuch, and C. Janot, J. Phys. (Paris), Colloq. **37**, C6-763 (1976).

[16] M. Boekholt, M. Hoffmann, and G. Güntherodt, Physica (Amsterdam) **175C**, 127 (1991).