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Temperature-dependent biquadratic coupling in antiferromagnetically coupled Fe/FeSi multilayers

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Fe/FeSi multilayers are known to exhibit a strong antiferromagnetic interlayer coupling peak centered at a nominal FeSi spacer thickness of ~15±2 Å at room temperature, and to develop remanence in the magnetic hysteresis loop upon cooling to ~100 K. An analysis of the hysteresis loops is found to require the inclusion of a temperature-dependent biquadratic (90° coupling) in addition to the bilinear coupling term in the energetics. The temperature dependence of the Fe/FeSi multilayer coupling can then be understood in terms that are applicable to conventional metallic multilayer systems such as Fe/Cr.

The exploration of metallic multilayers with oscillatory interlayer magnetic coupling[1] and giant magnetoresistance,[2] such as Fe/Cr and Co/Cu superlattices, has been a very active and rewarding field. The physics of the coupling is described in general starting from the Ruderman-Kittel interaction between ferromagnetic layers separated by the metallic spacer,[3] or equivalently by spin-dependent quantum well states in the spacer.[4] An exciting materials challenge to gain deeper insights into this technologically promising class of materials is to alter the electronic or structural nature of the spacer and observe the resultant change in the magnetic coupling and/or the magnetotransport properties. Thus, reports of antiferromagnetic (AF) interlayer coupling across amorphous (a)Si in Fe/a-Si/Fe trilayer structures,[5] and across iron silicide spacers in sputtered Fe/FeSi multilayers[6] stimulated much interest, especially due to announcements of photoinduced coupling effects attributed to the photogeneration of carriers across the semiconducting gap of the spacer.[7,8] These studies stimulated additional experimental work on the growth, structure,[9–11] and magnetotransport,[12] and theoretical models that are distinctly different from those for metallic spacers.[13,14]

For Fe/FeSi multilayers with an FeSi thickness of 15 Å, the coupling is AF at room temperature and at low temperature (<100 K) the magnetization loops show increased remanent magnetization.[6] The results were suggestive of a transition from AF to ferromagnetic or uncoupling upon cooling. The original photoeffect reports supported the hypothesis that carriers in the spacer could be thermally generated *or* photogenerated to restore AF coupling.[7,8] The photoeffect was the main support that the FeSi spacer was semiconducting and, therefore, "exotic," as opposed to metallic and relatively traditional.[7] However, further progress in these systems demonstrated that the restoration of AF coupling due to the light was a local laser heating effect, not associated with photogeneration of carriers.[15] This is now known to be the case for the sputtered multilayers of Ref. 7 as well. Therefore, the temperature dependence of the coupling in these multilayers is still an open question.

In the present work, we reinterpret the earlier temperature-dependent FeSi results within a more traditional context. We propose that the bilinear (J_1) AF coupling interaction persists upon cooling and even increases in strength, but with the simultaneous and more rapid increase of a biquadratic (J_2) (90° coupling) term that can overcome the bilinear term and add remanence to the low-temperature hysteresis loops. This is analogous to known behavior of biquadratically coupled Fe/Cr superlattices, for example.[16] In the Fe/Cr case J_2 is ascribed to thickness fluctuations according to the theory of Slonczewski.[17] In the present case presumably it is compositional fluctuations in the silicide layer that drives J_2. Strong temperature-dependent increases in J_2 are common in metallic multilayer systems, although the theoretical underpinnings have not been explored, except for the "loose" spins model.[18]

Before presenting the new biquadratic coupling interpretation of the FeSi data, it is useful to summarize background information and recent developments in the field. In Refs. 6 and 7 multilayers were prepared via magnetron sputtering at ambient temperature typically onto Si or sapphire substrates from Fe and Si sources. Interfacial mixing, diffusion and/or reaction produced the silicide from nominal $[\mathrm{Fe}\ (30\ \text{Å})/\mathrm{Si}\ (t_s)]_N$ structures, where N is typically 12–80 bilayers, and the nominal spacer thickness is 10–40 Å. Magnetometry, Mössbauer spectroscopy, polarized neutron reflectivity, and x-ray diffraction were used to deduce that the AF coupling is strong at room temperature (with switching fields ~6 kOe) and centered at $t_s \sim 15$ Å, and also that the spacer is a crystalline and nonmagnetic silicide. For $t_s > 20$ Å the spacer is amorphous and the system is uncoupled magnetically. Even artificial mixing of the spacer to form the suspected FeSi composition by interleaving atomic layers in the spacer deposition process failed to give rise to anticipated higher-order oscillations for $t_s > 20$ Å even though the crystallinity could be retained beyond this limit.[7] This suspected silicide candidate materials were either the ε phase or the CsCl-structural form of FeSi.[19] The ε phase was particularly appealing because it is a narrow-gap bulk semiconductor (~0.05 eV gap) and its nonmagnetic character is ascribed to a Kondo-insulator model.[20] Indeed, the theoretical coupling model of Ref. 14 to describe the usual magnetic properties reported for Fe/FeSi multilayers is based on this description, in which a many-body "hybridization gap" is formed between localized Fe $3d$ states and Si $3s$-$3p$ bands of the FeSi spacer. The gap is temperature dependent; it increases on

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cooling and disappears on warming. Although the model is intriguing, new experimental evidence argues against the presence of ε-FeSi and for the role of the CsCl-structure FeSi as originally suggested in Ref. 19. CsCl-structure FeSi is not an equilibrium bulk phase, but a metallic, metastable, interfacial compound.[21] Transmission electron diffraction measurements in Ref. 9 observed only diffraction peaks characteristic of bcc Fe and showed no evidence of known Fe silicides in samples which were AF coupled. Similar results were observed in Ref. 10 with the exception of an additional weak half-order (001) peak consistent with the metastable CsCl structure. Mössbauer spectroscopy work also provides evidence for the CsCl structure.[11] Also, the magnetoresistance of both the Argonne samples[6] and more recent ion-beam deposited samples of Ref. 10 are small (~0.1%) positive or negative, respectively.

The thrust of the present work is to provide an analysis of the temperature-dependent coupling properties of Fe/FeSi in terms of biquadratic coupling in addition to the AF coupling, as in traditional metallic multilayers.[22,23] This would obviate the need to require that the spacer be semiconducting or the presence of pin holes,[24] and would help rationalize the large magnitude of the AF coupling at $t_s \sim 15$ Å as arising from strong metallic coupling. Changes in the nature of the spacer material for $t_s > 20$ Å are still necessary to understand the lack of strong higher-order AF-coupling oscillations (e.g., the amorphization of the spacer,[6] or the alteration in its texturing, as reported in Ref. 10). This interpretation may not apply to the work in Ref. 15 in which the samples were grown and measured at 40 K.

The interlayer exchange energy can be written as

$$J_1 \mathbf{m}_1 \cdot \mathbf{m}_2 + J_2 (\mathbf{m}_1 \cdot \mathbf{m}_2)^2, \qquad (1)$$

where J_1 and J_2 are the bilinear and biquadratic coupling constants, respectively, and $\mathbf{m}_1$ and $\mathbf{m}_2$ are the magnetization of adjacent ferromagnetic layers. For $J_1 > 0$, the interlayer coupling favors AF alignment, whereas $J_2 > 0$ favors 90° alignment. This phenomenological expression for the interlayer coupling has been successful in interpreting the experimental results in a number of systems.[16,22] In many cases, the temperature dependence of J_2 has been found to be stronger than that of J_1, decreasing exponentially with increasing temperature. Given the importance of J_2 to the interlayer coupling, particularly at low temperatures, we have reexamined previous work on Fe/Si superlattices fitting the magnetization data to the bilinear plus biquadratic coupling scheme. We find that the anomalous temperature dependence can be understood as a result of a strongly temperature-dependent quadratic coupling. This is found to be analogous to the temperature dependence in Fe/Cr superlattices.

The multilayers are textured in the growth direction but polycrystalline in the plane, and the in-plane anisotropies are negligible. Therefore, the magnetization data can be fitted assuming only a contribution from the interlayer coupling and the Zeeman energy. Assuming that adjacent Fe layers are symmetric about the applied field H at angles $+\theta$ and $-\theta$, energy minimization gives the following expression:

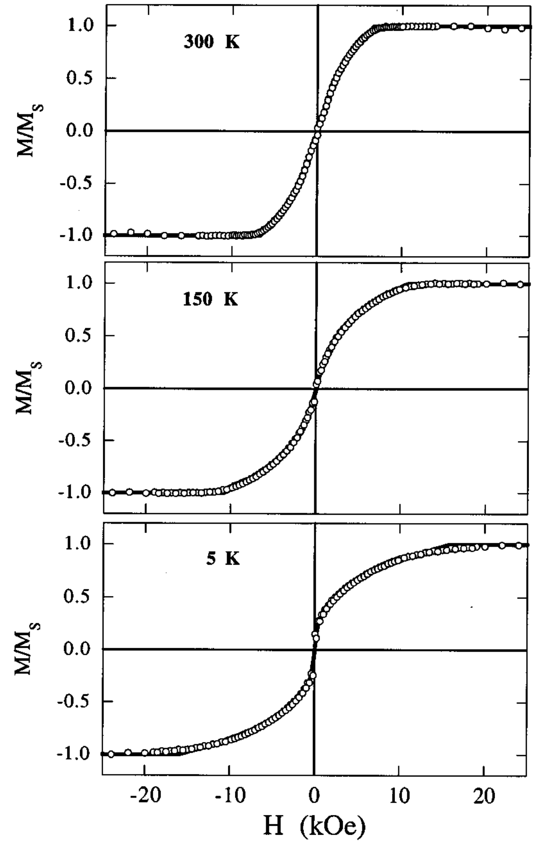

FIG. 1. Magnetic hysteresis loops for an [Fe (45 Å)/Si (15 Å)]$_{50}$ superlattice. Open circles are the measured points and the thick solid line is a fit to Eq. (2). Fitting results are shown in Fig. 3.

$$\frac{H M_s t_{\mathrm{Fe}}}{4} = (J_1 - 2J_2)\cos\theta + 4J_2\cos^3\theta. \qquad (2)$$

The saturation field H_s is given by $4(J_1 + 2J_2)/M_s t_{\mathrm{Fe}}$ and the remanent magnetization M_r/M_s by

$$\frac{M_r}{M_s} = \begin{cases} 0 & \text{for } J_1 > 2J_2 \\ \sqrt{(2J_2 - J_1)/4J_2} & \text{for } J_1 < 2J_2. \end{cases} \qquad (3)$$

Shown in Figs. 1 and 2 are the magnetization results for [Fe (45 Å)/Si (15 Å)]$_{50}$ and [Fe (45 Å)/Si (17 Å)]$_{50}$ multilayers grown at ambient temperature on sapphire substrates. The 15-Å sample was chosen to give the maximum interlayer coupling, and the 17-Å sample is displaced from the maximum and is more weakly coupled. Both samples are completely AF coupled at room temperature with zero remanent moment. As the temperature is decreased, the loop shape changes and the saturation field increases. For the 17-Å sample, below 100 K the remanent magnetization increases as previously observed. The thick solid lines in Figs. 1 and 2 are fitted to Eq. (2), where J_1 and J_2 are the only fitting parameters. The fits are able to reproduce both the shape of the magnetization as it approaches saturation and the remanent magnetizations. The results of the fitting procedure are summarized in Fig. 3. For both samples, the bilinear coupling is relatively weakly temperature dependent, increasing

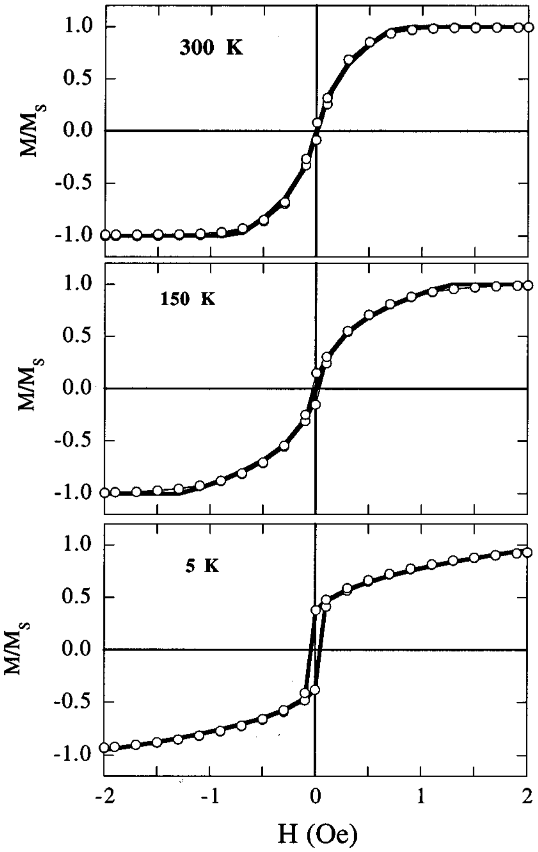

FIG. 2. Magnetic hysteresis loops for an [Fe (45 Å)/Si (17 Å)]$_{50}$ superlattice. Open circles are the measured points and the thick solid line is a fit to Eq. (2). Fitting results are shown in Fig. 3.

by a factor of 2 from room temperature to 5 K. In contrast, the biquadratic coupling increases six-fold over the same temperature range.

These results can be compared to well-studied Fe/Cr superlattices.[25] Shown in Fig. 4 are the results for a (100)-oriented [Fe (14 Å)/Cr (26 Å)]$_{28}$ superlattice. The growth and structural characterization of the superlattice are described in Ref. 26. The magnetization curve in Fig. 4 shows the same qualitative features observed in Fig. 1. Again, the solid lines are fits to Eq. (2), including a cubic anisotropy term, showing a stronger increase in J_2 relative to J_1 with decreasing temperature. The characteristic shape of the hysteresis loops are also similar to those observed for Ag/FeNi multilayers.[23] It was the striking similarity between the Fe/FeSi and Fe/Cr data that suggested to us the biquadratic interpretation presented herein.

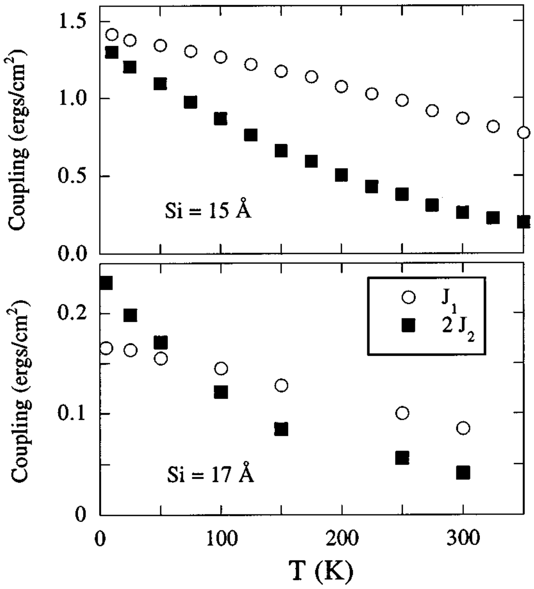

FIG. 3. Interlayer coupling values for J_1 and $2J_2$ determined for the superlattices shown in Figs. 1 and 2.

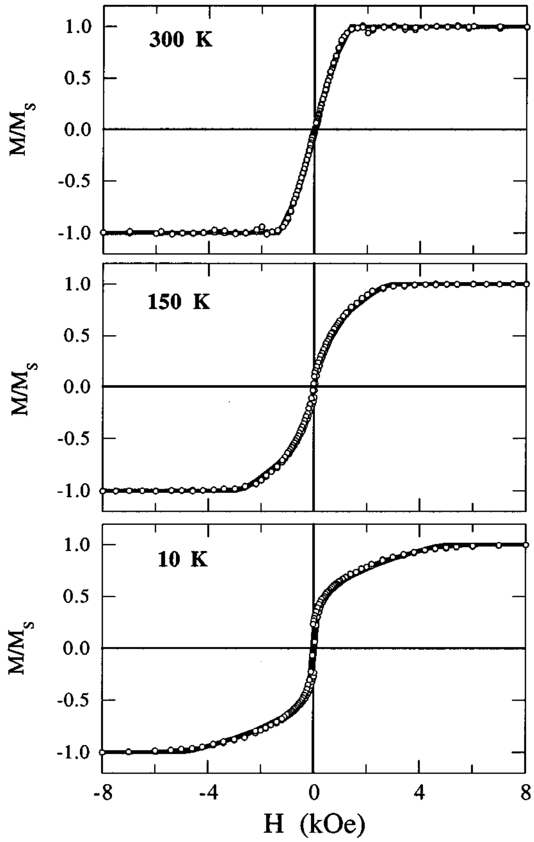

FIG. 4. Magnetic hysteresis loops for a (001) oriented [Fe (14 Å)/Cr (26 Å)]$_{28}$ superlattice measured with the applied field along the Fe[100] easy axis. Open circles are the measured points and the thick solid line is a fit to Eq. (2) including cubic anisotropy. Fitting results are $J_1=0.058$ and $J_2=0.009$ ergs/cm^2 (300 K), $J_1=0.092$ and $J_2=0.031$ ergs/cm^2 (150 K), and $J_1=0.110$ and $J_2=0.068$ ergs/cm^2 (10 K).

The implication is that these results provide a framework to begin to understand the properties of Fe/FeSi multilayers in conventional terms, as have been applied to metallic multilayers. There remain, of course, many interesting questions and challenges associated with the exploration of ''exotic'' spacer materials.

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