Properties of magnetic superlattices with antiferromagnetic interfacial coupling: Magnetization, susceptibility, and compensation points

R. E. Camley*
Max Planck Institut für Festkörperforschung, Heisenbergstrasse 1, 7000 Stuttgart 80, Federal Republic of Germany
(Received 3 October 1988; revised manuscript received 9 January 1989)

Magnetic superlattices composed of ferromagnetic films (Fe and Gd) which couple antiferromagnetically at the interfaces are studied. In a magnetic field a variety of different spin configurations are possible. We show how these phases are reflected in magnetization and susceptibility measurements. In particular, a twisted spin configuration is characterized by a much larger susceptibility than an aligned spin configuration. We show that small changes in the layering pattern can lead to large changes in $M(T)$ and static susceptibility. In contrast, changes in the value of the interface exchange parameter cause large changes only in the weak-coupling limit.

In a previous paper,[1] we considered a superlattice made of alternating ferromagnetic films, but where the exchange coupling at the interfaces was antiferromagnetic. Within mean field theory, we investigated a model system where the two ferromagnets were Gd and Fe. A number of different phases were found, depending on the applied magnetic field and temperature. The most interesting of these phases were (1) aligned-Fe phase where all Fe spins were parallel to the external field and all Gd spins were antiparallel, (2) aligned-Gd phase where all Gd spins were parallel to the external field and all the Fe spins were antiparallel, (3) twisted phase where the the spins in each layer make a different angle with the external field. In this phase the spin configuration can vary from something very similar to the spin flop phase in an antiferromagnet to a configuration similar to that in a domain wall.

The different phases arise from a competition between exchange and Zeeman energies. For small magnetic fields the Zeeman energy is less important and the minimization of the exchange energy generally favors the aligned states. Which aligned state (aligned Gd or aligned Fe) is chosen depends on the net magnetic moment in each film. Clearly the film with the larger moment will align parallel to the field. This introduces a strong temperature dependence since the thermal averaged magnetic moment in Gd changes rapidly compared to that of Fe. For moderate fields both Zeeman and exchange energies will be important. This favors the twisted state. In addition the case where the net magnetic moments in each film are about the same also favors a twisted state, but where the spin configuration looks similar to a spin-flop state.

It is the purpose of this paper to show how the different microscopic ground states can be observed in macroscopic measurements such as magnetization and susceptibility. In addition, since the nature and strength of the interface coupling is of fundamental interest, we will investigate the role that the interface coupling constant plays in superlattice properties.

The situation of two ferromagnets which couple antiferromagnetically is actually quite common.[2] In addition to the examples given in Ref. 1, there are some recent results on $GdCo_2/Co$ (Ref. 3) and Gd/Co (Ref. 4) superlattices which have the structure discussed here. Also, Gd/Y superlattices show effective antiferromagnetic coupling between the Gd films for some thicknesses of Y spacer layers.[5] Thus the results obtained here will have a reasonably wide application. Clearly anisotropy effects, not included here, may play an important role in some systems.

The mean field method for determining the ground state of the superlattice was described in detail previously.[1] Once the ground state is found, it is simple to find the net spin moment of the unit cell S_{total}. A quantity proportional to the static susceptibility dM/dH can then be found numerically by taking the difference of S_{total} evaluated at two close values of magnetic field.

The spin values for this calculation are $S_{\text{Fe}}=1$, $S_{\text{Gd}}=\frac{7}{2}$. The ratio of exchange constants $J_{\text{Gd}}/J_{\text{Fe}}=0.0355$ is obtained by comparing the transition temperatures of Gd and Fe. We consider two limiting values for the interface exchange constant J_I. The weak-coupling limit has $J_I=J_{\text{Gd}}$, and the strong-coupling limit has $J_I=J_{\text{Fe}}$. The external field is measured in dimensionless units; $h=g\mu_B H_0/J_{\text{Fe}}S_{\text{Fe}}$. Here H_0 is the external field applied in the z direction. A value of $h=0.01$ corresponds to an external field of about 7.3 kG. In Ref. 1 a value of $S_{\text{Fe}}=\frac{5}{2}$, appropriate to the Fe^{3+} ion, was incorrectly used for Fe metal. This also gave a different ratio for the exchange constants of Gd and Fe.

We can obtain a crude estimate for the length of the twist as a function of magnetic field as follows. Consider a single ferromagnetic film with a total of N layers. The outer spin layer on one side of the film is artificially fixed antiparallel to an applied field to simulate our antiferromagnetic coupling. The number of layers n_0 over which a 180° twist will occur is found by minimizing the sum of the exchange and Zeeman energies. A simple calculation[6] gives $n_0=(z\pi^2/2h)^{1/2}$ where h is $g\mu_B H_0/JS$ as above and z is the number of nearest neighbors. For $h=0.01$ and $t=0$, n_0 is about 62 layers in Fe and 22 layers in Gd. At higher temperatures, the temperature dependence of S_{Gd} would result in a narrower twist region in Gd. Clearly the twists in the superlattice system do not take place only in Fe or only in Gd, but in both. Also, the twisted states found here do not always correspond to full 180° turns. Nonetheless, this simple estimate sets an order of magnitude length scale for our problem.

In Figs. 1–3 we give examples of how the microscopic phases are related to the macroscopic properties of magnetization and susceptibility. Figure 1(a) shows the phase diagram for a structure with a unit cell of 13 layers of Fe and 5 layers of Gd (a 13Fe/5Gd). The temperature is measured in dimensionless units with $t=T/T_{C\mathrm{Fe}}$. Figure 1(b) shows S_{total} as a function of t for an external field of $h=0.03$. At $t=0$, the net spin moment in 5 Gd layers is larger than the net spin of the 13 Fe layers, i.e., $S_{\mathrm{total\,Gd}}=\frac{35}{2}=17.5$ and $S_{\mathrm{total\,Fe}}=13$. The total spin of the unit cell is then the difference of the two values above, 4.5. As t increases the Gd spins reduce in magnitude quickly compared to the Fe spins and the magnetization is reduced. Near $t=0.2$ there is a phase transition to the twisted state. Here the magnetization changes only very slowly as a function of temperature. Near $t=0.33$ there is a second phase transition to the aligned-Fe state. In this case the magnetization initially increases (due to the still decreasing Gd moments) and then decreases as the average Fe moments are reduced by thermal fluctuations.

Figure 1(c) shows the susceptibility as a function of temperature at $h=0.03$. Here χ is low for the aligned states and high in the region of the twisted state. The reason for this is clear. Susceptibility measures the ability of the spin configuration to change due to an external field. In the aligned states only the magnitudes can change. In the twisted state the magnitudes and the orientation of the spin can both change, leading to a larger susceptibility.

Comparing Fig. 2 (for a 14Fe/4Gd structure) to Fig. 1, we see that small changes in the structure of the unit cell lead to large changes in both the phase diagram and the macroscopic parameters of magnetization and susceptibility. Tailoring magnetic properties is thus quite practical in these structures.

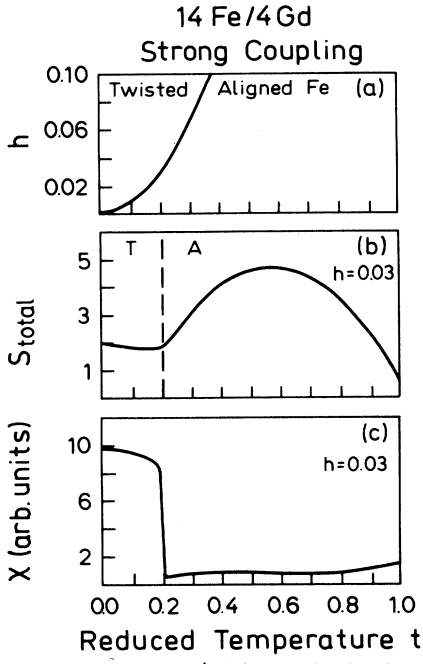

FIG. 2. Results for a 14Fe/4Gd superlattice in the strong-coupling limit. (a)-(c) are as in Fig. 1.

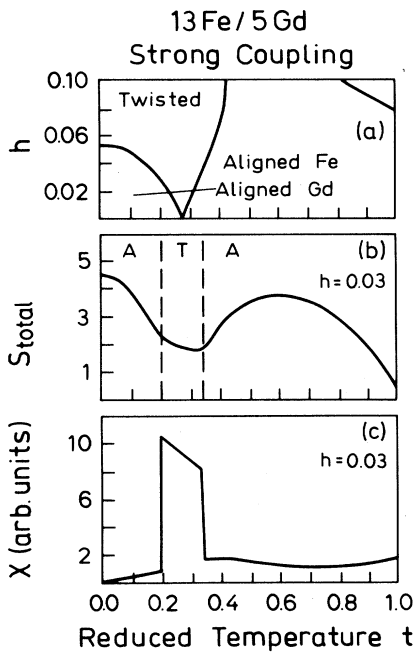

FIG. 1. Results for a 13Fe/5Gd superlattice in the strong-coupling limit; (a) the h-t phase diagram, (b) S_{total} vs t at a field of $h=0.03$, (c) the susceptibility vs t, again for $h=0.03$. The labels A and T in (b) indicate the regions of the aligned and twisted states found from the phase diagram.

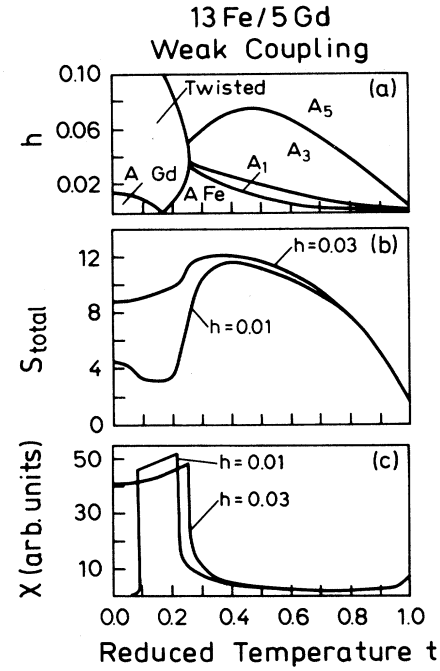

FIG. 3. Results for a 13Fe/5Gd superlattice in the weak-coupling limit. (a)–(c) are as in Fig. 1. $A_1(A_3, A_5)$ are aligned Fe states, but with the middle (3,5) Gd spins aligned with the field. AGd and AFe indicate the aligned-Gd and aligned-Fe states, respectively. Note in (b) that changing the magnetic field has a significant effect on magnetization as a function of temperature.

In a comparison of Fig. 1(a) (strong coupling) and Fig. 3(a) (weak coupling) we see that the general features remain the same, but the numerical values are quite different. In particular the aligned-Gd state occurs in only a very small portion of the phase diagram of Fig. 3(a) when compared to Fig. 1(a). Here the decreased interface exchange has allowed the transition from the aligned-Gd state to the twisted state to occur at much lower values of external fields, i.e., in the competition between Zeeman and exchange energies, the minimization of the exchange energy has become less important and the twisted state is favored.

A second major difference is visible in the comparison of Figs. 1(a) and 3(a). In the weak-coupling case the twisted state exists only for temperatures below $t=0.26$ while in the strong coupling case the twisted state is seen to exist beyond $t=0.4$. To understand this difference, we first have to understand the origin of the transition from the twisted state to the aligned-Fe state. As the temperature increases, the thermal averaged values of the Gd moments become quite small, and the Zeeman contribution of the Gd thus is also small. As a result, the compromise of the twisted state is no longer energetically favorable and the system changes to the aligned-Fe state. The difference between the weak and strong interface coupling is then simply related to the magnitudes of the Gd moments as a function of interface coupling.

We explore this idea further in Fig. 4 with a plot of the thermal averaged spin magnitude as a function of Gd layer number for different temperatures and for strong and weak coupling cases. For strong coupling, the value for $\langle S\rangle$ is held up by the coupling to the Fe layers. In contrast, for weak coupling the exchange field from the Fe is weaker than that from the Gd layers and as a result $\langle S\rangle$ is lower, especially at the boundaries of the Gd film.[7] In comparing the strong coupling and weak coupling for the same temperature ($t=0.25$) we see that $\langle S\rangle$ is much larger for all positions in the strong coupling case. Thus the twisted mode, which needs the Zeeman energy from the Gd spins for its existence, survives at higher temperatures for the strong coupling case.

Figure 5 explores the influence of the interfacial exchange parameter on several different variables. We consider a 13Fe/5Gd structure, and $t=0$. Figure 5(a) shows how the angle between the Fe and Gd spins at the interface changes as a function of J_I. The applied field is given by $h=0.06$ so the superlattice is in the twisted state. We see here that for all but the smallest values of J_I the Gd and Fe spins at the interface are nearly 180° apart. This is to be expected since, for J_I moderate to large, it would cost too much in exchange energy to have a large angular deviation at the interface. In Figs. 5(b) and 5(c) we plot the experimentally measurable parameters of χ and h_c as a function of the interface exchange. h_c is the field at which the transition from the aligned to twisted state takes place at $T=0$. The susceptibility is nearly constant except for small J_I where the spin orientation can be more flexible with a corresponding increase in the susceptibility. Figure 5(c) shows that as the interface coupling is reduced h_c is also reduced. Thus for very weak interface coupling there is no aligned-Gd state.

As noted previously, the multilayer geometry allows tailoring of the material parameters. For systems with an aligned-Gd phase, the temperature t_c at which the spin configuration changes from the aligned-Gd state to the aligned-Fe state (at $h=0$) corresponds to the case of total magnetization equal to zero. This has previously been called the compensation point, in analogy with ferrimagnets.[3] Figure 6 presents t_c as a function of N for an

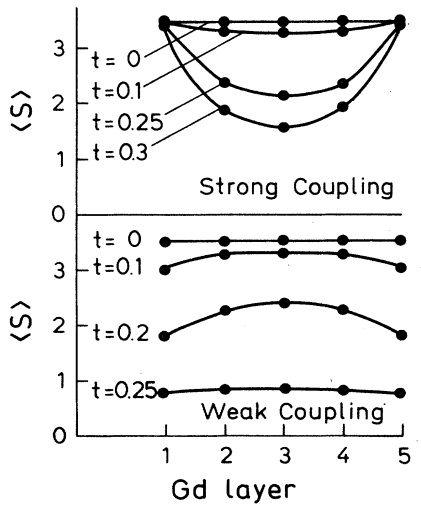

FIG. 4. Average spin moment as a function of position and temperature for the Gd spins in a 13Fe/5Gd superlattice. In the strong-coupling limit the Gd spins at the interface are increased in magnitude compared to those in the bulk while in the weak-coupling limit the reverse is true.

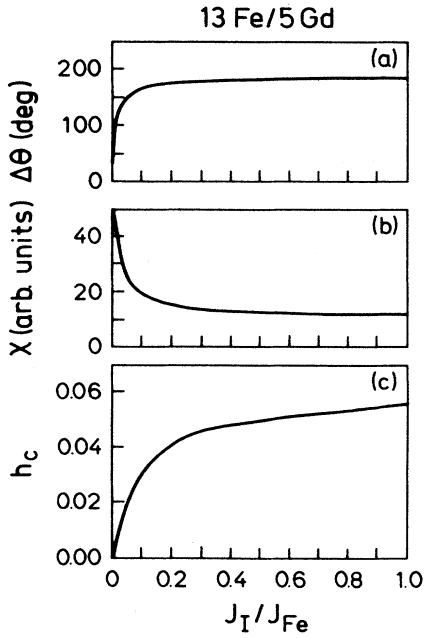

FIG. 5. The effect of interfacial exchange on (a) the angle between the Fe and Gd spins at the interface, (b) susceptibility, and (c) the critical field h_c.

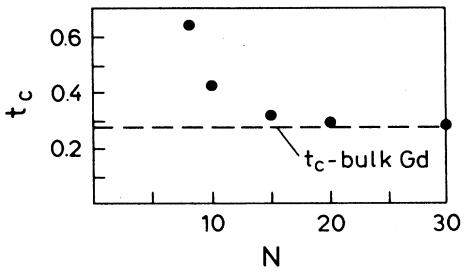

FIG. 6. The compensation temperature as a function of number of layers N for a NFe/NGd superlattice. $J_I/J_{Fe}=0.706$. As N decreases, the effect of the interfaces becomes more important.

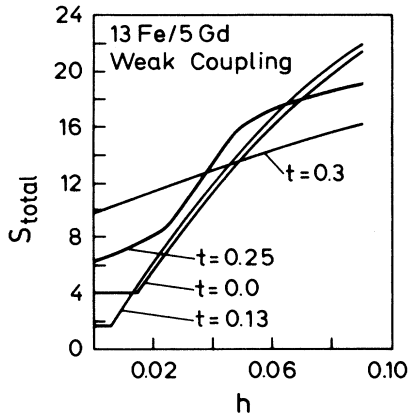

FIG. 7. S_{total} as a function of h for the 13Fe/5Gd superlattice in the weak-coupling limit.

NFe/NGd superlattice structure. The interface coupling constant here is rather strong, $J_I/J_{Fe}=0.706$, a value obtained from alloy mean field calculations.[2] For large N the superlattice phase transition essentially occurs near the magnetic phase transion of bulk Gd. As N decreases, the influence of interfaces becomes more important. Here with the strong interface coupling the Gd spins are "held up" at the edges of the Gd film as seen in Fig. 4(a), and the phase transition occurs at a higher temperature. This result is qualitatively similar to one found for a Gd/Co multilayer deposition, but the experimental result apparently occurs due to film morphology.[3] From the figure we see that $N=15$ is the smallest value for which t_c is not too far from the bulk Gd value. Thus the influence of the interface on the thermal properties of the bulk spins extends out only about 7 layers.

A common experimental characterization of the magnetic materials is an M vs H_0 curve. In Fig. 7 we present a theoretical calculation for S_{total} vs h for the 13Fe/5Gd structure in the weak-coupling limit. For low temperatures and fields the aligned states are characterized by nearly horizontal lines where the magnetization looks as if it is saturated. Twisted states are indicated by lines with nearly constant slope, i.e., the susceptibility is nearly independent of the applied field (and to a lesser extent temperature). The results are in excellent qualitative agreement with some recent experimental results for Gd/Co superlattices.[4] It is worth noting, however, that for $h=0$ the experiment shows a monotonic increase in S_{total} with temperature, while the theoretical work shows (see Fig. 3) first a decrease in S_{total} and then an increase. The reason for this difference is that the experiments were carried out only for temperatures above 80 K, presumably above, or at least near, the compensation temperature for this structure. It would be interesting to have experimental data for temperatures below 80 K in order to compare experiment and theory more completely.

In summary:

(1) The microscopic aligned and twisted states can easily be observed in macroscopic measurements such as magnetization and susceptibility as a function of temperature. In particular the twisted state has a much higher susceptibility than the aligned state due to its orientational polarizability.

(2) Strong interface exchange allows thin Gd films (less than 15 layers) to remain ferromagnetic well above the Curie temperature for bulk Gd. This, in turn allows the twisted state to exist at temperatures above the bulk transition temperature of Gd. Changes in the interface exchange cause large changes in the magnetic properties only in the weak coupling region.

This work was supported by the Army Research Office Grant No. DAALO3-88-K-0061. The author would also like to thank Peter Fulde for the hospitality and support of the Max Planck Institut where most of this work was carried out.

*Permanent address: Department of Physics, University of Colorado, Colorado Springs, CO 80933-7150.

[1]R. E. Camley and D. R. Tilley, Phys. Rev. B **37**, 3413 (1988).

[2]See, K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elseiver, New York, 1984), Chap. 6.

[3]D. J. Webb, A. F. Marshall, A. M. Toxen, T. H. Geballe, and R. M. White, IEEE Trans. Magn. **24**, 2013 (1988); D. J. Webb, R. G. Walmsley, K. Parvin, P. H. Dickinson, T. H. Geballe, and R. M. White, Phys. Rev. B **32**, 4667 (1985).

[4]S. Tsunashima, T. Ichikawa, M. Nawate, and S. Uchiyama, in *International Conference on Magnetism, Paris, 1988* [J. Phys. (Paris) (to be published)].

[5]J. Kwo, E. M. Gyorgy, D. B. McWhan, M. Hong, F. J. DiSalvo, C. Vettier, and J. E. Bower, Phys. Rev. Lett. **55**, 1402 (1985); C. F. Majkrzak, J. W. Cable, J. Kwo, M. Hong, D. B. McWhan, Y. Yafet, and J. V. Waszczak, *ibid.* **56**, 2700 (1986).

[6]See the simple calculation for domain wall thicknesses in C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1986).

[7]A similar result was found using a Landau-Ginzburg approach by R. M. White and D. J. Friedman, J. Magn. Magn. Mater. **49**, 117 (1985).