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1991 J. Phys.: Condens. Matter 3 4971

(http://iopscience.iop.org/0953-8984/3/26/017)

J. Phys.: Condens. Matter 3 (1991) 4971–4982. Printed in the UK

Monte Carlo simulations of a simple Ising spin model representing 111 and 100 nickel–iron multilayers

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Received 27 February 1991

Abstract. The magnetic behaviour of FCC superlattices of equal-thickness Ni and Fe layers, with repeat lengths between 6 and 30 atomic layers, and with layers parallel to both 111 and 100 crystal planes, is modelled with a short range interaction Ising model, using parameters determined from a previous study of homogeneous Ni_cFe_{1-c} alloys. The ground states of these models and the dependence of their magnetizations on temperature and magnetic field are studied using Monte Carlo simulation. We pay particular attention to macroscopic manifestations of the antiferromagnetic coupling between the Fe sites, and the surprisingly strong ferromagnetic Ni–Fe coupling deduced from the simulations of the solid solutions. We suggest experiments which would critically test the model.

1. Introduction and details of the model

The properties of metallic multilayers are of considerable current interest because they are new materials with new and possibly useful properties, perhaps hosting new phenomena; for a review of work in this area see [1]. In this paper we will be concerned with the magnetic properties of Ni/Fe multilayers of 50% composition, i.e. in which both Ni and Fe layers are composed of L monolayers, with values of L between 3 and 15. Previous work in this field notwithstanding, our work is prompted by the recent discoveries of Jennet and Dingley [2]. They have found that under appropriate conditions, for $L < 10$ monolayers per Fe layer, the Fe layers formed an FCC coherent lattice with the Ni layers rather than the BCC lattice characteristic of bulk Fe at low temperatures. This makes the system particularly interesting magnetically, due to the fact that while Ni is ferromagnetic, there is mounting evidence [3–10] that Fe on an appropriate FCC lattice is antiferromagnetic. Superlattices composed of alternating ferromagnetic and and antiferromagnetic layers have received considerable attention in the past [11–13], one of the many interesting features they exhibit being the much studied 'exchange anisotropy effect' [14,15]

Our aim here is to investigate a simple but reasonably reliable model for the magnetic properties of the Ni/Fe multilayers in order to stimulate experimental interest in measuring them.

The model is a perfect rigid FCC lattice on which sites behave magnetically according to the Ising Hamiltonian:

$$\mathcal{H} = -\sum_{i>j} J_{ij}\sigma_i\sigma_j - H\sum_i \sigma_i \qquad (1)$$

0953-8984/91/264971+12$03.50

where σ_i is an Ising spin variable $\sigma_i = \pm 1$, H is an external magnetic field, and J_{ij} is defined in terms of the occupation variable ξ_i, which is 1 if the atom at site i is Ni and 0 if it is Fe, and the composition-specific exchange integrals, as follows:

$$J_{ij} = \xi_i\xi_j J_{ij}^{\mathrm{NiNi}} + \xi_i(1-\xi_j)J_{ij}^{\mathrm{NiFe}} + (1-\xi_i)\xi_j J_{ij}^{\mathrm{NiFe}} + (1-\xi_i)(1-\xi_j)J_{ij}^{\mathrm{FeFe}}. \tag{2}$$

In the case of a multilayer with a repeat length of $2L$ monolayers, every site in the layers $2nL+1, \ldots, 2nL+L$ has $\xi_i = 1$ (i.e. is occupied by a Ni atom) and every site in the layers $2nL+L+1, \ldots, 2nL+2L$ has $\xi = 0$ (i.e. is occupied by a Fe atom), for integer n.

The simplifying assumptions used in this model are considerable. Instead of a full quantum mechanical description of the itinerant magnetism the magnetic part of the Hamiltonian is spin-only and uses Ising, rather than vector spins. Moreover the interaction strengths are assumed to be constant not only with temperature, field and state of order, but also with position within the inhomogeneous sample. The significance of our results in the light of these idealizations is discussed in the Conclusion.

In [3] we have used the Hamiltonian (1) to study compositional and ferromagnetic ordering in the Ni-rich FCC homogeneous alloys. We found that using the following values of the exchange integrals, as well as a large nearest neighbour compositional interaction $V \approx -130$ meV which is not relevant for the fixed-site multilayer case, we were able to reproduce most of the experimental observations:

$$\begin{aligned} J_{\mathrm{NN}}^{\mathrm{NiNi}} &= 5.3\ \mathrm{meV} & J_{\mathrm{NNN}}^{\mathrm{NiNi}} &= 0.000\ \mathrm{meV} \\ J_{\mathrm{NN}}^{\mathrm{NiFe}} &= 11.5\ \mathrm{meV} & J_{\mathrm{NNN}}^{\mathrm{NiFe}} &= -0.115\ \mathrm{meV} \\ J_{\mathrm{NN}}^{\mathrm{FeFe}} &= -3.3\ \mathrm{meV} & J_{\mathrm{NNN}}^{\mathrm{FeFe}} &= 0.033\ \mathrm{meV} \end{aligned} \tag{3}$$

where the subscripts NN and NNN refer to nearest neighbour and next nearest neighbour respectively. The next nearest neighbour interactions are small and little effect is expected from them; they were introduced to break ground state degeneracy and are retained principally for uniformity with [3]. On a FCC lattice the ordering temperatures defined by these values in a Monte Carlo simulation are as follows: the Curie temperature of Ni $T_{\mathrm{C}}^{\mathrm{Ni}} \approx 634$ K and the Néel temperature of Fe $T_{\mathrm{N}}^{\mathrm{Fe}} \approx 120$ K.

Central to the motivation of this paper are the two noteworthy features of the interaction parameters listed in equation (3). These are the antiferromagnetic coupling (−3.3 meV) between Fe sites and the strong ferromagnetic coupling (11.5 meV) between Ni and Fe sites. Our principle aim is to identify experimentally accessible consequences of these microscopic interactions.

In the simulations periodic boundary conditions were employed outside a system of one or two times the repeat length. This sometimes led to a system with one dimension quite small (down to 6 atomic spacings), and normally one would be worried about finite size effects compromising the accuracy of the results, particularly near phase transitions. However, this will only occur if the correlation length ξ exceeds the size of the system in such a way as to wrap around the periodic boundary conditions. Since ξ diverges only at $T_{\mathrm{C}}^{\mathrm{Ni}}$ in the Ni region and only at $T_{\mathrm{N}}^{\mathrm{Fe}}$ in the Fe region, and these two are widely separated in temperature, a spin will never 'feel the effect' of its own image through both layers.

In order to interpret our simulations we have also worked out the theory within the mean field approximation. Except for the well known failure of the mean field

theory to deal with frustration in an ordering process on the FCC lattice we have found a broad agreement between the mean field theory and simulations.

In the simulation runs of between 800 and 5000 Monte Carlo steps per site for each result were used as appropriate, care being taken to ensure that all data represents equilibrium states of the system. Lateral system size was 32×32 sites. The simulations were carried out on an AMT Distributed Array Processor DAP 510.

2. Zero-field magnetic ground states of the systems

The important features of the ground states of the above systems can be anticipated directly from examination of the exchange interactions (3). Since the Ni–Ni coupling is ferromagnetic and the Fe–Fe coupling is antiferromagnetic, the multilayer will consist, to a first approximation, of magnetic zones each L monolayers thick alternately ferromagnetic and antiferromagnetic. Since in addition the Ni–Fe interaction is ferromagnetic, the ferromagnetic layers will tend to be $L+2$ monolayers thick, extending one monolayer into the Fe layers on either side of each Ni layer, and the antiferromagnetic layers $L-2$ monolayers.

For the systems with layers parallel to the 111 crystal planes this leads to a simple alternation of ferromagnetic Ni layers and antiferromagnetic Fe layers with no long range coupling between layers. Thus on average, for a large number of repeats of the superlattice unit cell, there will be zero ground state magnetization of the multilayer, since of the ferromagnetic Ni layers as many will be magnetized with spin up as with spin down.

The magnetic structure of Fe on a FCC lattice is type I antiferromagnetic (the $\hat{z}$ axis of the Ising model is taken to be along an assumed 100 easy axis), that is the planes of parallel spin are parallel to the 100 planes of the crystal. Thus we see that in this case the Fe layers will consist of monolayers each ferromagnetically aligned within themselves, successive monolayers having magnetizations alternately parallel and antiparallel to that of the ferromagnetic Ni layer. This has the consequence that if L is odd then adjacent Ni layers are ferromagnetic parallel to each other, while if L is even then adjacent Ni layers are ferromagnetic but antiparallel to each other, so that the magnetic repeat length is twice the compositional repeat length. We have here a 'supercoupling' effect in which coupling between ferromagnetic regions is alternately ferromagnetic and antiferromagnetic with change in L. The magnetic ground states are summarized in figure 1.

A similar effect has been produced experimentally by separating ferromagnetic regions with certain non-magnetic metals [16–19], giving rise to oscillatory exchange coupling and the so-called giant magnetoresistance. The origin of this is, however, quite different; in contradistinction to the coupling in the present case it is electronic. It turns out that the exchange coupling between ferromagnetic layers communicated by the conduction electrons of the non-magnetic spacer layer oscillates with a period of typically 8–10 monolayers (though see [18] for an example of an oscillation length near a single monolayer) and dies out over similar scales. On the other hand the period of the effect noted above must be exactly equal to two monolayers and the effective superlayer interaction strength for a perfect sample ought not to decrease with L.

This state is somewhat reminiscent of the one-dimensional Ising model; at low temperatures each Ni layer will have only a single degree of freedom, all spins being

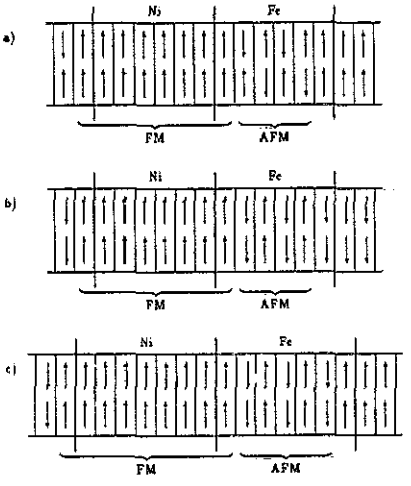

Figure 1. Magnetic ground states. (a) 111 multilayer: each plane in the antiferromagnetic region AFM is antiferromagnetically aligned within itself, and there is no coupling between the ferromagnetic regions FM. (b) 100 multilayer with even L: each plane in the antiferromagnetic region is ferromagnetically aligned within itself, leading to 'super-antiferromagnetic' coupling between the ferromagnetic regions. (c) 100 multilayer with odd L: again, each plane in the antiferromagnetic region is ferromagnetically aligned, here leading to 'superferromagnetic' coupling between the ferromagnetic regions.

either up or down, and adjacent Ni layers will interact via an effective exchange interaction proportional to the number of sites at each interface. It is well known that the 1D Ising model has no phase transition at finite temperature, and it might be thought that this system resembles it in this respect. In fact the two systems are significantly different from each other: the 1D Ising model lacks a phase transition because introducing a domain boundary leads to a finite increase $2J$ in the internal energy E but also to an increase in the entropy S which tends logarithmically to infinity with the length of the chain of spins. Thus for an infinite chain the free energy $F = E - TS$ will not be minimized for a fully ordered state for any temperature $T > 0$. This is only true in virtue of the finite strength of the interaction and the infinite length of the chain of spins. While it is possible to conceive of a multilayer sample of infinite length and finite interface size, this is hardly a natural geometry, and certainly for a cubic sample ordering will not be prevented by these considerations†.

It should be noted that although this 'supercoupling' is a definite feature of our model it might be very difficult to reproduce experimentally since the difference

† The Curie temperature of this system can be shown to be

$$T_C^{1D} = 306 \text{ kelvin} \times \frac{A}{\ln R(L-1)}$$

where A is the number of sites in each crystal plane parallel to the Ni–Fe interfaces, L is the number of monolayers per Fe layer and R is the number of repeats of the superlattice unit cell perpendicular to the interfaces. Of course this gives only an upper bound to the actual Curie temperature of the system, since the reasoning only holds while each monolayer has a single degree of freedom, i.e. at zero temperature, but the fact that for any reasonable sample geometry $T_C^{1D} \gg T_C^{\text{multilayer}}$ indicates that this case is very far from the 1D Ising case.

between aligned and antialigned ferromagnetic regions is so sensitive to Fe layer thickness. Experimentally there is likely to be some roughness in layer boundaries and unevenness of deposition, and these slight deviations from the sharp boundaries and exact layer thicknesses of our model could easily destroy the effect. Moreover, since $T_N^{Fe} \ll T_C^{Ni}$ it would be extremely difficult to reach the ground state by cooling even on a perfect multilayer sample; since the ergodicity of the ferromagnetic regions is broken at a temperature at which the Fe regions are highly disordered it is more likely for the system to become trapped in a local minimum and at low temperatures achieve a state like that of the 111 multilayers, of random alignment of each ferromagnetic region. Extremely slow cooling would be needed to counteract this tendency.

Simulations are in broad agreement with the above reasoning concerning the ground states of the system. However, we find that the stability of the 111 multilayer ground states is extremely delicate: the $L = 3$ and $L = 4$ cases are so degenerate as to appear to possess finite entropy at $T = 0$, and the energetically equivalent ground states of these systems have a range of different magnetizations so that the zero-temperature spontaneous magnetization is undefined. The $L > 4$ cases are more robust because of the small next nearest neighbour interactions which anyway become unimportant at temperatures above even a few kelvins. Since this state of affairs is rather strongly dependent on the details of the model, in particular the assumption that interactions beyond nearest neighbour are unimportant, it seems best not to take too seriously the predictions of this model for 111 multilayers at very low temperatures (a few tens of kelvins).

3. Results of the simulations and discussion

Given the origin of the exchange interactions (3) it is natural to compare our results of simulating the multilayers with the corresponding results for homogeneous $Ni_{0.5}Fe_{0.5}$ alloys. Of this latter, two sets are considered, one for an alloy quenched from near its melting point of about 1500 K [21], and hence being fixed in a state lacking compositional long range order and with only magnetic freedom, and one for an alloy which is annealed and so has both magnetic and compositional freedom. Experimental results for $Ni_{0.5}Fe_{0.5}$, which due to the long ordering time of the system are likely to be from partially ordered states, may be expected to lie between results for the ordering and disordered alloys. The difference between these two homogeneous alloys was discussed in [3] and it was attributed to the state of compositional order. From this point of view the multilayer system is simply another compositional configuration.

3.1. Temperature dependence of the zero-field magnetization

Graphs of M versus T for various values of L are given for 111 and 100 multilayers in figures 2 and 3 respectively.

The most dramatic feature of these results is the difference in Curie temperature T_C between the homogeneous alloys and the multilayers. This is the consequence of the strong Ni–Fe coupling ($J^{NiFe} \approx 2J^{NiNi}$) and may be understood as follows: the bulk magnetization of the multilayers is non-zero in virtue of the spontaneous magnetization of the Ni layers, which are ferromagnetic islands in a non-ferromagnetic sea of Fe. The Fe monolayers adjacent to the Ni layers contribute to the magnetization below T_C also, but can only do so while they have the ferromagnetic Ni layer with which to interact, so can produce no bulk magnetization above T_C^{Ni}. So we see that

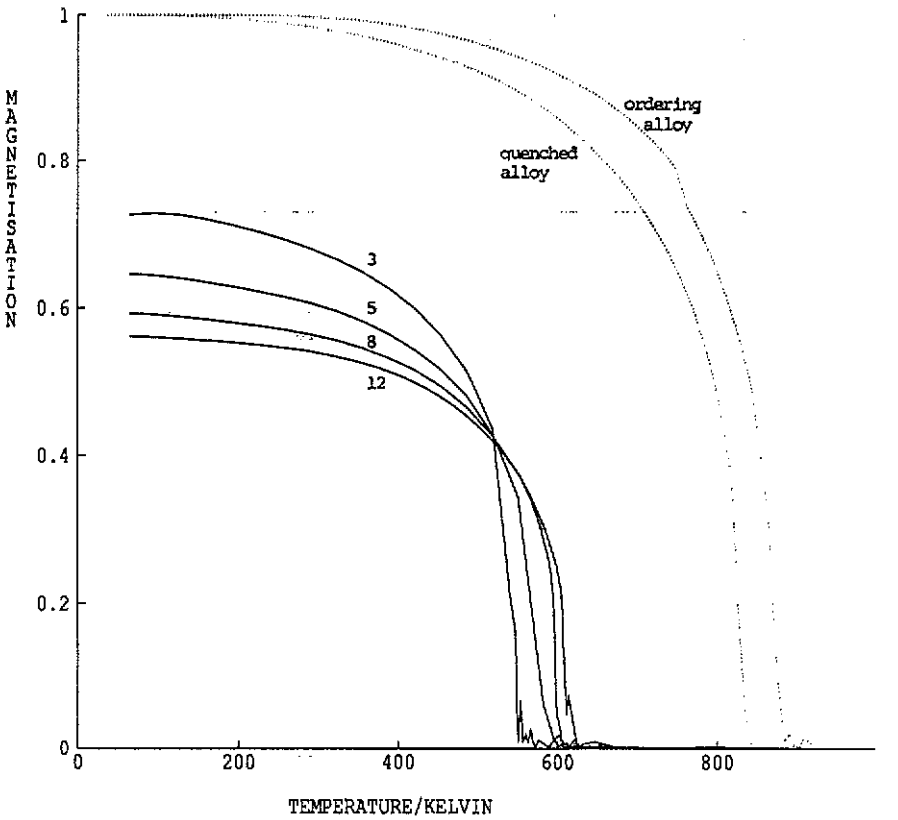

Figure 2. Temperature dependence of magnetization for 111 multilayers. Data are given for multilayers of varying layer thickness (full curves), and homogeneous alloys (dotted curves). Note the discrepancy in Curie temperatures: for the multilayers T_C is near T_C^{Ni} and for the alloys it is much higher.

the Curie temperature of the multilayer is determined by the Ni–Ni interaction J^{NiNi} only. For the homogeneous alloy the magnetization is of the whole bulk and, since in bulk there will be interacting pairs of all the types Ni–Ni, Ni–Fe and Fe–Fe, the Curie temperature will be dependent in some non-trivial way on all the interaction values J^{NiNi}, J^{NiFe} and J^{FeFe}. In short, going from the homogeneous alloy to the multilayer the number of Ni–Fe nearest neighbour pairs decreases dramatically and hence T_C drops. Evidently, useful information about the Ni–Fe interactions could be gained by an experiment in which the Curie temperature of a multilayer is measured and then remeasured after annealing out of the compositional modulation. A major advantage of such an experiment is the fact that, in so far as Ni and Fe have different moments, and these are transferable from the solid solution to the multilayer, they are automatically compensated for, since the number of atoms of each element in the sample does not change, merely their lattice positions. It may be that the lattice parameter, or lattice strains, change from one arrangement to the other however, and this may complicate matters. Moreover, the compositional modulation may anneal out below $T_C^{multilayer}$, making it unobservable.

The above argument suggests that all Ni–Fe multilayers (for $L > 2$) should exhibit

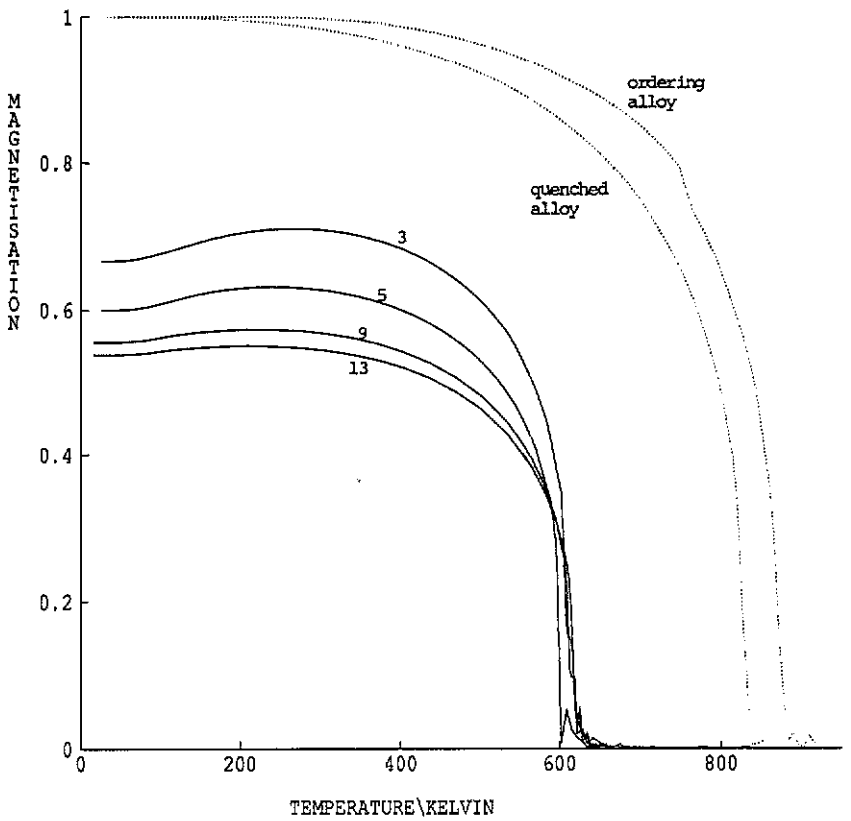

Figure 3. Temperature dependence of magnetization for 100 multilayers. Data are given for odd-L multilayers and homogeneous alloys. All even-L 100 multilayers have zero magnetization at all temperatures. Note the discrepancy in Curie temperatures between the alloys and the multilayers. Note also that the maximum of multilayer magnetization is not at $T = 0$, especially for low values of L.

precisely the same Curie temperature, namely $T_C^{Ni} = 634$ K (from [21]). This is to a first approximation true, but close inspection of the mean field theory and simulation results reveals a weak dependence of T_C on L, the multilayers with larger repeat lengths tending asymptotically up to the bulk value. This is due to the fact that the magnetization is not that of an infinite three-dimensional bulk but of an (infinite) array of quasi-two-dimensional systems, i.e. three-dimensional systems of which one dimension is finite in extent. In other words the modulation period L is a thermodynamic variable like the concentration or the chemical potential.

The other main feature of the multilayer–homogeneous alloy comparison is the difference in magnetization at low temperatures. This has been essentially explained in section 2, but is clarified below.

Firstly, the spontaneous magnetization of even-L 100 multilayers is zero for all temperatures. This is because, although part of the multilayer is ferromagnetic, half the ferromagnetic regions have spins antiparallel to those of the other half, so there is no bulk magnetization from the ferromagnetic regions. This behaviour is due to the antiferromagnetic coupling between the internally ferromagnetic Ni layers

by the antiferromagnetic Fe layers. Naturally, there is no contribution to the bulk magnetization from the antiferromagnetic regions either.

As noted in section 2 the above will on average be the case for all 111 multilayers as well. In order to see some bulk magnetization however, we have constrained all ferromagnetic regions to be magnetized parallel to each other. This is not energetically unfavourable, merely unlikely, but is easily experimentally realised by application of a small magnetic field while cooling through T_C^{Ni}, or of a larger magnetic field while below T_C^{Ni}.

For the odd-L 100 multilayers, the magnetization at $T = 0$ is $(L + 1)/2L$, following the argument in section 2. This is also true for 111 multilayers, subject to the comments made at the end of section 2 above.

A less pronounced effect is the maximum in the magnetization of the odd-L 100 multilayers at non-zero temperature. This occurs because the first parts of the system to lose magnetic order are the antiferromagnetic Fe layers, since $T_N^{Fe} \ll T_C^{Ni}$. There is an odd number of these monolayers and always one more aligned antiparallel to the ferromagnetic Ni than parallel to it (see figure 1(c)), so that the uniform loss of magnetic order in this region will tend to increase the magnetization of the sample. Clearly, the effect is more pronounced for low values of L. This effect may be difficult to observe in experiment for the same reasons as the superantiferromagnetism described above.

3.2. Field dependence of the magnetization

Magnetic M versus H hysteresis loops are given for 111 and 100 multilayers in figures 4 and 5 respectively. These were taken from simulations at 190 K. At higher temperatures below T_C^{Ni} the curves remained much the same, except that the loops were smaller and the remanences lower. These results presuppose a single magnetic domain.

The principle feature of the above M–H loops is the low remanence of the multilayer curves compared to those of the alloys. This is a simple consequence of the combination of ferromagnetic and non-ferromagnetic properties of the systems; the Ni layers which order ferromagnetically (like the homogeneous alloys) have a very square hysteresis loop, while the Fe which is in its paramagnetic phase above $T_N^{Fe} \approx 120$ K displays no hysteresis at all, but a magnetization which goes through the origin. Asymptotically as $L \rightarrow \infty$ this would lead to a form just half way between the ferromagnetic and non-magnetic curves, but as can be seen from the graphs the increasing importance of the interfacial effects at small L leads to some deviation from this form. This argument is illustrated by the magnetization profile of figure 6.

The second, and less obvious, feature of the field response is the small step in the sudden jump between large negative and large positive magnetization, observed only for the even-L 100 multilayers. At large negative field the system is saturated, with all spins pointing down. As the field increases some Fe atoms reverse spin but nearly all Ni atoms retain their original orientation; in particular the overall magnetization of each Ni layer remains negative—the system is in a 'superferromagnetic' state. As H reaches the coercivity, *half* of the ferromagnetic Ni layers reverse their magnetization, leaving the system in a superantiferromagnetic state similar to the ground state of figure 1(b), this arrangement being slightly more stable than the previous one in the presence of a field. The difference in stability is small however, and when the applied field is increased a little, the overall magnetization in all the layers flips to positive.

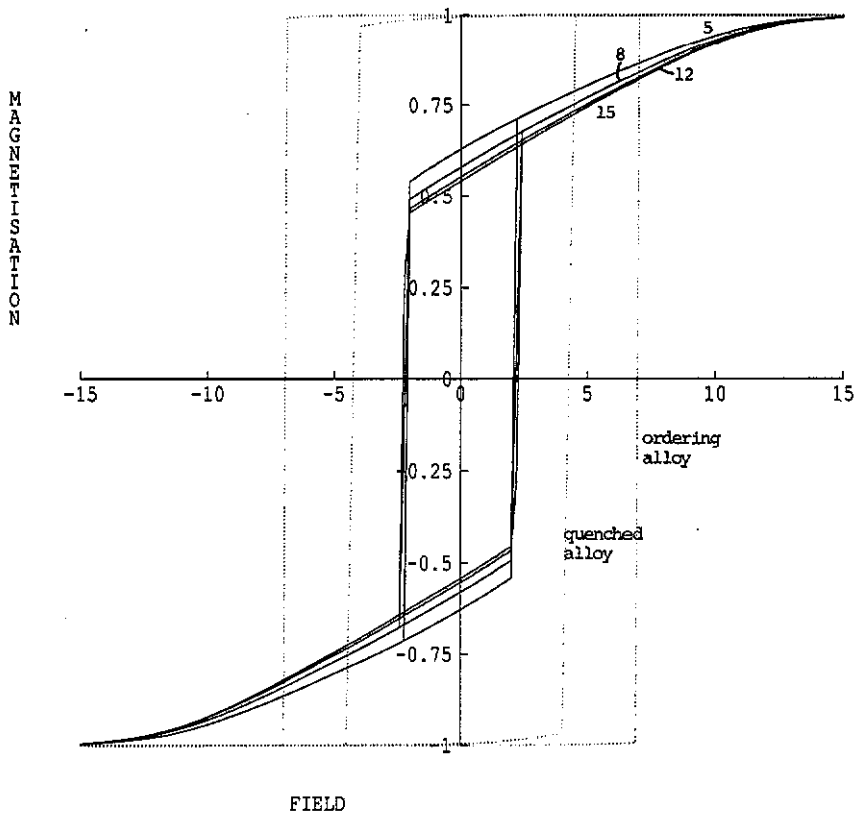

Figure 4. H-field dependence of the magnetization for 111 multilayers at 190 K. Field is in units of J_{NN}^{NiNi}. Note that the hysteresis loops of the partially ferromagnetic multilayers are much less square than those of the entirely ferromagnetic alloys.

This is not only a useful signature of the nature of these structures, but may provide a more practicable method than extremely slow cooling of putting the even-L 100 multilayers into their true ground states rather than one of the local minima discussed in section 2, subject to the sample perfection constraints mentioned there. However, the region in which the superantiferromagnetic state is stable and the superferromagnetic one is not is quite small and may even vanish with different model parameters, so that it might prove difficult, or impossible, to locate experimentally.

4. Conclusion

We have performed simulations on a spin-only Ising model of a perfect Ni/Fe multilayer system with equal Ni and Fe layer thicknesses of between 3 and 15 monolayers, and we have identified certain characteristic features of the temperature and field dependence of the bulk magnetization of these materials. In particular we found that the Curie temperature T_C and the zero-temperature magnetization M_0 are significantly smaller for the multilayers than for the corresponding $Ni_{0.5}Fe_{0.5}$ homogeneous alloys. Moreover, we stressed that these prominent features are the consequences of the

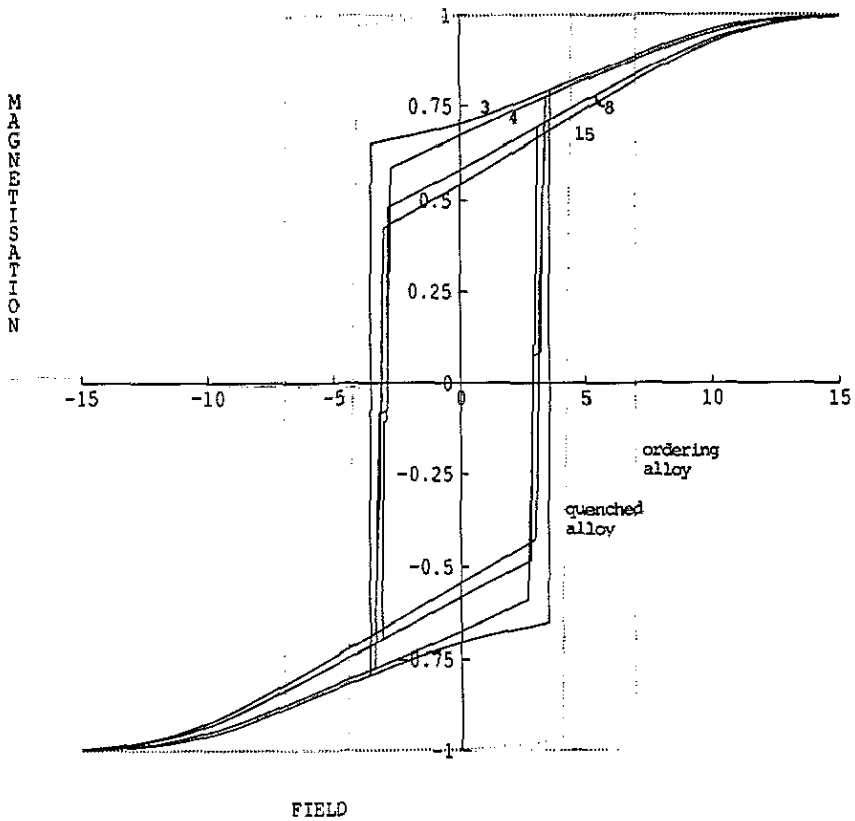

Figure 5. H-field dependence of the magnetization for 100 multilayers at 190 K. Field is in units of J_{NN}^{NiNi}. Note the relative slantedness of the multilayer hysteresis loops. Note also the small intermediate 'superantiferromagnetic' plateaus in the magnetization between the two 'superferromagnetic' states for the even-L multilayers.

largeness of J_{NN}^{NiFe} and the negative sign of J_{NN}^{FeFe}. The interesting question is whether or not experiment can be used to substantiate or reject these critical characteristics of our theoretical model.

The main obvious deficiency of our model is that the spins are Ising spins, rather than quantum mechanical vector spins with anisotropy arising from various sources. By examining the reasoning in this paper it should be clear that most of the properties described arise simply from the ferromagnetism of the Ni, the antiferromagnetism of the Fe, the strong ferromagnetic coupling across the Ni–Fe interfaces, and the rough values of the ordering temperatures associated with these interactions, so that as long as these are not in question, and as long as the geometry of the sample is a fairly close approximation to that assumed here (and we believe this to be the case) the behaviour of this model will be broadly similar to that of a real sample, subject to the qualifications made in the text. For a theoretical treatment of a similar system employing a vector spin model, which comes to similar conclusions at least concerning the ground state, see the work of Hinchey and Mills [13].

One simplification of our work which may turn out to affect our conclusions is the operation of magnetic interactions beyond nearest neighbour. Although we have

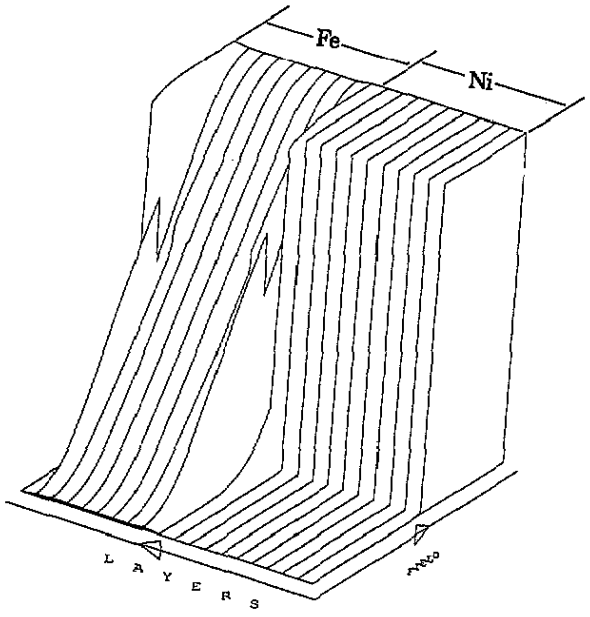

Figure 6. Layer-by-layer profile of the 111 $L = 10$ multilayer magnetization-field behaviour at 190 K. Only one half of the hysteresis loop is shown. The ferromagnetic Ni on the right has an almost entirely square hysteresis loop, while the paramagnetic Fe on the left displays no hysteresis behaviour, the curves going through the point $(M = 0, H = 0)$. Interfacial effects are evident in the outermost Fe layers.

included a next nearest neighbour term it is small, and this was an assumption rather than fitted from experimental data. If contributions beyond nearest neighbour are important this may significantly affect our results, particularly those for small layer thickness L.

We plan in the future to extend our research using a vector spin model.

Acknowledgments

MBT was financially supported by an SERC studentship and the calculations were carried out on the departmental DAP provided by the Computational Science Initiative.

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