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Electronic and magnetic structure of the ternary fcc Mn-Fe-Ni system. II. Disordered alloys and ferromagnetic-antiferromagnetic interfaces

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We have developed a novel statistical theory of fcc ternary Mn-Fe-Ni alloys based on a decomposition of the fcc lattice into space-filling atomic tetrahedra whose magnetic properties are estimated from the magnetic properties of analogous tetrahedra in ordered fcc ternary compounds. Using this theory, we have calculated the average total moment and root-mean-square moment per atom as a function of alloy composition and lattice constant. As an application of this statistical theory, we discuss the magnetic properties of the permalloy–MnFe-alloy interface, which is an example of a two-dimensional disordered fcc ternary Mn-Fe-Ni alloy. Because of the hybrid nature of the atomic environment at this interface, there is a ferrimagnetic transition layer between ferromagnetic permalloy and antiferromagnetic MnFe regions. This feature has been largely ignored in discussions of this structure but should be taken into account in treating the magnetic properties of this interface.

INTRODUCTION

We have recently calculated the magnetic structure of 15 ordered fcc Mn-Fe-Ni compounds and determined the spin arrangements as a function of lattice constant and composition.[1] These compounds may be regarded as crystals having the translational periodicity of a simple-cubic lattice with four atoms per unit cell. These compounds are defined by the chemical formula $\mathrm{Mn}_m\mathrm{Fe}_n\mathrm{Ni}_{4-m-n}$, $0 \le m, n \le 4$, where m and n are integers. In the present paper we carry our study of the Mn-Fe-Ni system further, developing a statistical theory of the magnetic structure of disordered fcc Mn-Fe-Ni alloys (solid substitutional solutions). The novel feature of our theory is the decomposition of the fcc alloy into space-filling atomic tetrahedra whose magnetic properties can be inferred from the magnetic properties of closely related ordered fcc compounds. As an application, we will consider ferromagnetic-antiferromagnetic interfaces represented by the permalloy $(\mathrm{Fe}_{25}\mathrm{Ni}_{75})/\mathrm{Mn}_{50}\mathrm{Fe}_{50}$ system. These interfaces are two-dimensional examples of disordered fcc Mn-Fe-Ni alloys and as such can be described by the present statistical theory.

In an earlier study,[2] we investigated permalloy-MnFe interfaces by carrying out spin-polarized superlattice calculations using ultrathin slabs of ordered FeNi_3 and MnFe as model systems. In the present paper we deal with this interface more directly as a ternary fcc Mn-Fe-Ni alloy. As already indicated in our preceding paper,[1] ferromagnetic-antiferromagnetic interfaces are of interest for scientific as well as technological reasons. Rapid advances in vacuum technology and surface characterization techniques have increased the likelihood that nearly atomically abrupt ferromagnetic-antiferromagnetic interfaces will soon be grown by molecular-beam epitaxy.[3] Accordingly, the magnetic properties of ultrathin ferromagnetic-antiferromagnetic multilayers and superlattices will become increasingly important subjects for theoretical and experimental studies. The present theory provides a means for studying such interfaces in a relatively straightforward manner.

MODELS FOR SOLID SUBSTITUTIONAL ALLOYS

In this section we will develop a statistical theory of alloys with a view to estimating the magnetic properties of solid substitutional fcc ternary Mn-Fe-Ni *alloys*. We will start with the results given in Ref. 1 for the atomic moments of 15 fcc ternary Mn-Fe-Ni *compounds* as a function of lattice constant. In order to avoid repetition, we will assume that the reader is familiar with Ref. 1. We will denote the magnetic moment of an atom of type t (t=Mn, Fe, Ni) at site s in compound q by $\mu_{s,t}(q)$, so that the average total moment per atom and the root-mean-square (rms) moment per atom for compound q are given by

$$\mu_{\mathrm{comp}}^{\mathrm{avg}}(q)=\frac{1}{4}\sum_{s=1}^{4}\mu_{s,t}(q),\quad \mu_{\mathrm{comp}}^{\mathrm{rms}}(q)=\left[\frac{1}{4}\sum_{s=1}^{4}\mu_{s,t}(q)^2\right]^{1/2}. \tag{1}$$

If atoms of the same type have different moments at different positions in the 4-atom unit cell, they can be distinguished from one another by the site index s. It will be understood that all atomic moments are lattice-constant dependent, although our notation does not indicate this explicitly. In Ref. 1, the individual atomic and average total moments are listed for three different lattice constants; values at intermediate lattice constants are readily determined by interpolation. The 15 ordered compounds belonging to our so-called basic model and some of their properties are listed in Table I.

Our treatment of the Mn-Fe-Ni alloys will be based on a number of simplifying assumptions. We will assume that the atoms are arranged at random on a fcc lattice having an average lattice constant that depends only on composition. We will neglect slight variations in interatomic distances that may occur because of fluctuating local atomic environments. Since no long-range or short-range *chemical* order is assumed, the occupancy of each atomic site is determined solely by the atomic composition, which is denoted by c, where $c=\{c_{\text{Mn}},c_{\text{Fe}},c_{\text{Ni}}\}$, and $c_{\text{Mn}}+c_{\text{Fe}}+c_{\text{Ni}}=1$.

In traditional statistical theories of magnetic alloys,[4-8] the magnetic moment of an atom is usually assumed to depend on its local environment, which is normally described by the number of nearest-neighbor atoms of each type, n, where $n=\{n_{\text{Mn}},n_{\text{Fe}},n_{\text{Ni}}\}$. The effects of more distant neighbors are typically neglected, as are the effects of different spatial arrangements of nearest- (and further-) neighbor atoms. This approach works reasonably well for ferromagnetic systems, where all the atoms have the same spin, but may become unwieldy for antiferromagnetic systems, where the magnetic environment (spatial arrangement of up and down spins) must also be taken into account.

If atomic moments in disordered fcc ternary Mn-Fe-Ni alloys depended only on the number of nearest neighbors of each type, we could easily calculate these moments: For a given alloy stoichiometry, we could calculate the average numbers of Mn, Fe, and Ni atoms surrounding each type of atom in the alloy. For those sets of numbers that occur in the 15 ordered compounds of our basic model (cf. Table I), the magnetic moment of the central atom could be taken as the moment of that particular atom in one of the compounds that has exactly the same set of nearest-neighbor numbers. For other sets of numbers, the central-atom moment could be obtained by interpolation. We could then carry out a statistical calculation taking fluctuations in the nearest-neighbor numbers into account, and averaging over different types of atoms, ultimately obtaining the average total moment, as in traditional statistical theories.[4-8]

However, in antiferromagnetic systems the atomic moments also depend on the local magnetic environment, that is to say, on the signs as well as the magnitudes of the nearest-neighbor moments. Thus, the information contained in Table I is insufficient for investigating the Mn-Fe-Ni system, which is antiferromagnetic over a substantial portion of the phase diagram (cf. Fig. 1). Therefore, we will have to proceed differently. Instead of focusing on individual atoms and their chemical and magnetic environments, we will take advantage of the fact that a fcc lattice can be regarded as a simple-cubic lattice having four atoms per unit cell. Thus, a disordered fcc ternary alloy can be decomposed into a periodic array of space-filling atomic tetrahedra.

In the spirit of a mean-field theory, we will begin by assuming that the average atomic moment of each type of tetrahedron depends only on its chemical composition, i.e., on the number of Mn, Fe, and Ni atoms belonging to this tetrahedron. We would expect some variation in the atomic moments of a given type of tetrahedron embedded in different alloy environments, but we will proceed on the assumption that the average over all possible local environments is roughly equal to the average atomic moment of the corresponding atomic tetrahedron in one of the 15 ordered fcc Mn-Fe-Ni compounds. At a later stage we will improve upon this simplified model by taking magnetic correlations between adjacent tetrahedra into account.

The present treatment involving tetrahedral spatial

TABLE I. Number of nearest Mn, Fe, and Ni neighbors surrounding central Mn, Fe, and Ni atoms in the 4-atom compounds (ignoring spin). The compound serial number is denoted by q. The spin patterns of the compounds are denoted by plus and minus signs. Missing signs denote zero atomic moments. In the right-hand column, FM denotes a ferromagnetic compound, while AFM1 and AFM2 denote antiferromagnetic compounds of the first and second kind, respectively. Weakly ferrimagnetic spin arrangements are represented by the closest antiferromagnetic spin arragnements.

					Central Mn			Central Fe			Central Ni			
q	Compound				Mn	Fe	Ni	Mn	Fe	Ni	Mn	Fe	Ni	FM-AFM
1	Mn+	Mn−	Mn+	Mn−	12									AFM1
2	Mn+	Mn+	Mn−	Fe+	8	4		12						AFM2
3	Mn+	Mn−	Fe+	Fe−	4	8		8	4					AFM1
4	Mn−	Fe+	Fe+	Fe+		12		4	8					AFM2
5	Fe+	Fe−	Fe+	Fe−					12					AFM1
6	Mn+	Mn+	Mn−	Ni	8		4				12			AFM2
7	Mn+	Mn−	Fe−	Ni+	4	4	4	8		4	8	4		AFM1
8	Mn−	Fe+	Fe+	Ni+		8	4	4	4	4	4	8		AFM2
9	Fe+	Fe+	Fe+	Ni+					8	4		12		FM
10	Mn+	Mn−	Ni	Ni	4		8				8		4	AFM1
11	Mn+	Fe−	Ni	Ni		4	8	4		8	4	4	4	AFM1
12	Fe+	Fe+	Ni+	Ni+					4	8		8	4	FM
13	Mn+	Ni+	Ni+	Ni+			12				4		8	FM
14	Fe+	Ni+	Ni+	Ni+						12		4	8	FM
15	Ni+	Ni+	Ni+	Ni+									12	FM

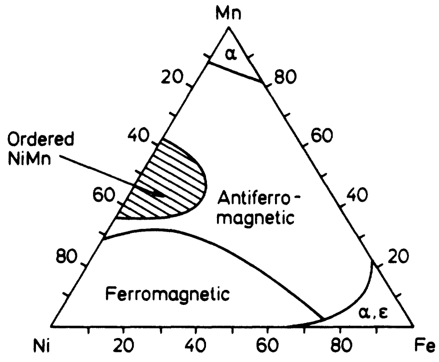

FIG. 1. Schematic phase diagram of ternary Mn-Fe-Ni system.

decomposition is obviously geared to the fact that we already know the magnetic properties of 15 compounds, each constructed entirely from such tetrahedra.[1] In view of the one-to-one correspondence between any atomic tetrahedron chosen at random in the fcc Mn-Fe-Ni alloy system, and the 4-atom unit cell of one of the 15 compounds q, we can label each alloy tetrahedra by the corresponding compound serial number q.

We will now introduce a phase factor $f_m(q,r)=\pm 1$, partly to allow for the arbitrariness of spin-up and spin-down directions,[9] and partly to simplify the treatment of magnetic correlations between adjacent tetrahedra, which we will come to in due course. The phase factor $f_m(q,r)$ is common to all the atoms in a given tetrahedron, but may be different for different types of tetrahedra q, and for different spatial orientations r of the same tetrahedral type. The orientation index r can range from 1 to 12 depending on the symmetry of the tetrahedron.[10] The phase factor will also depend on the magnetic order, as specified by the subscript m. Accordingly, we can now write the average total moment of alloy tetrahedron q having orientation r as

$$\bar{\mu}_m^{\mathrm{avg}}(q,r)=\mu^{\mathrm{avg}}(q)f_m(q,r)=\pm\mu^{\mathrm{avg}}(q)\ , \qquad (2)$$

where $\mu^{\mathrm{avg}}(q)$ is defined by Eq. (1), and the bar over μ indicates that the phase factor is included in its definition. Phase factors do not appear in expressions for rms moments since $|f_m(q,r)|=1$ for all m, q, and r.

By introducing $f_m(q,r)$, we will be able to describe antiferromagnetic arrangements on fcc lattices that require more than four sublattices.[11,12] In Fig. 2 we display antiferromagnetic arrangements of the first, second, and third kind, which we will refer to as AFM1, AFM2, and AFM3, respectively. Here the chemical identities of the various sites are ignored, and only the spin directions are indicated. Four sublattices are sufficient only for antiferromagnetic arrangements of the first kind shown in Fig. 2(a). Here sublattices (or sites) 5–8 are equivalent to sublattices 1–4, respectively. Note that all atoms on a given (001) sheet have the same spin, but alternate (001) sheets have opposite spins. This would be a possible antiferromagnetic arrangement for fcc alloys composed predominantly of Mn_4, Mn_2Fe_2, and Fe_4, which have exactly this spin pattern.[1] The same may be true for Mn_2FeNi, and possibly also for Mn_2Ni_2 and $MnFeNi_2$, where the two Ni atoms have zero moments and the remaining two atoms opposite moments. All of these compounds are identified in Table I by the symbol AFM1.

In Fig. 2(b) we illustrate antiferromagnetic arrangements of the second kind, which may be characterized by (111) sheets, all atoms on each sheet having the same spin, with alternating sheets having opposite spins. Such arrangements require eight distinct sublattices, sublattices 1–4 having spins opposite to those at sublattices 5–8, respectively. The atomic tetrahedra 1-2-3-4 and 5-6-7-8 each have three spins of one sign and one spin of the other sign. This would be a possible antiferromagnetic spin arrangement for 8-atom compounds formed predominantly from one or two 4-atom compounds having such spin arrangements, for example, Mn_3Fe, $MnFe_3$, Mn_3Ni, and $MnFe_2Ni$, although some orientations of their respective tetrahedra may be less favorable than others. These compounds are identified in Table I

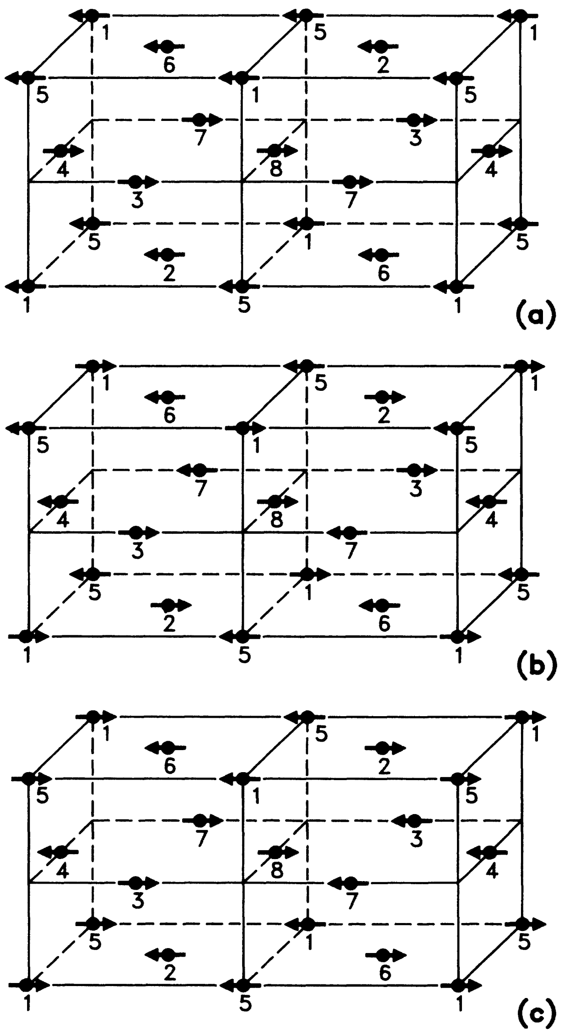

FIG. 2. Antiferromagnetic arrangements of the first, second, and third kind for fcc lattice regarded as eight interpenetrating simple-cubic lattices.

by the symbol AFM2.

In Fig. 2(c) we illustrate antiferromagnetic arrangements of the third kind, which again require eight sublattices. Here sublattices 1–4 have spins opposite to those for sublattices 5–8, respectively, but now the atomic tetrahedra 1-2-3-4 and 5-6-7-8 each have two spins up and two spins down. This would be a possible antiferromagnetic arrangement for the compounds already mentioned in connection with antiferromagnetic arrangements of the first kind, except that now we are in effect dealing with 8-atom compounds composed of two 4-atom compounds each of which has two spin-up and two spin-down atoms per unit cell, but oppositely directed with respect to one another.

Clearly, we can encompass all of these antiferromagnetic arrangements by representing the fcc lattice by eight simple-cubic sublattices.[11,12] The first four of these form a periodic array of tetrahedra lying in the red cubes of a three-dimensional checkerboard; similarly, the second four sublattices form a periodic array of tetrahedra lying in the black cubes. It is convenient to describe the geometry in these pictorial terms, and we will continue to do so.

In principle, we could evaluate the average moment (per atom) of the alloy by summing over all pairs of red and black tetrahedra, allowing for different permutations of the atoms in each tetrahedron, and weighing each pair of tetrahedra according to their likelihood of occurrence for the specified alloy composition c. This would be a very ambitious approach indeed, requiring a knowledge of the magnetic structure of all possible 8-atom compounds.[1] Instead, we will ignore environmental and orientational effects, and assume—provisionally—that the average total moment of a tetrahedron depends only on its chemical composition. This moment is thus given by Eq. (2), or Eq. (1), depending on whether or not we include magnetic correlations.

By neglecting environmental (chemical) and orientational (spatial) correlations between adjacent tetrahedra, but allowing for magnetic correlations, we can greatly simplify the expression for the average total moment per atom. This becomes

$$\bar{\mu}^{\mathrm{avg}}_{\mathrm{alloy},m}(c)=\sum_{p=1}^{15}\sum_{q=1}^{15}P_4\{n(p),c\}P_4\{n(q),c\} \times\tfrac{1}{2}[\mu^{\mathrm{avg}}(p)f_m(p)+\mu^{\mathrm{avg}}(q)f_m(q)]\ , \tag{3}$$

where the two types of tetrahedra (in the red and black cubes) are identified by p and q, respectively, and where the probability factors have the form

$$P_4(n,c)=\frac{(n_{\mathrm{Mn}}+n_{\mathrm{Fe}}+n_{\mathrm{Ni}})!}{n_{\mathrm{Mn}}!n_{\mathrm{Fe}}!n_{\mathrm{Ni}}!}c_{\mathrm{Mn}}^{n_{\mathrm{Mn}}}c_{\mathrm{Fe}}^{n_{\mathrm{Fe}}}c_{\mathrm{Ni}}^{n_{\mathrm{Ni}}}\ . \tag{4}$$

Here $n=n(p)$ or $n(q)$, and $n=n_{\mathrm{Mn}}+n_{\mathrm{Fe}}+n_{\mathrm{Ni}}=4$. The first factor on the right-hand side of Eq. (4) accounts for the different spatial orientations of the atomic tetrahedron. The number of such orientations depends on its composition n, where $n=(n_{\mathrm{Mn}},n_{\mathrm{Fe}},n_{\mathrm{Ni}})$. The remaining factors account for the likelihood of occurrence of any tetrahedron (p or q) consistent with the alloy composition c. Because of the assumption of orientation independence, the index r does not appear in the phase factors in Eq. (3).

The analogous expression for the rms alloy moment does not depend on phase factors and is given by

$$\mu^{\mathrm{rms}}_{\mathrm{slloy}}(c)=\sum_{p=1}^{15}\sum_{q=1}^{15}P_4\{n(p),c\}P_4\{n(q),c\} \times\{\tfrac{1}{2}[\mu^{\mathrm{rms}}(p)^2+\mu^{\mathrm{rms}}(q)^2]\}^{1/2}\ . \tag{5}$$

This provides a common measure of the magnetization strength for ferromagnetic and ferrimagnetic as well as antiferromagnetic arrangements.

MAGNETIC STRUCTURE OF 8-ATOM EXTENDED MODEL COMPOUNDS

We will now proceed to estimate the magnetic structure of 8-atom compounds formed from all possible three-dimensional checkerboard arrangements of red and black 4-atom compounds. Since we already know the magnetic structure of all the 4-atom compounds, we must choose phase factors that will lead to the most plausible long-range magnetic order for all pairs of 4-atom compounds. For example, we would expect $\mathrm{Fe}_8=\mathrm{Fe}_4+\mathrm{Fe}_4$ to have an antiferromagnetic arrangements of the first kind. In the absence of any evidence to the contrary, we will assume that Fe_4 remains antiferromagnetic even when it combines with another 4-atom compound to form an 8-atom compound.[13] In order to obtain the most favorable 8-atom spin pattern, it might be necessary to change the sign of one of the 4-atom phase factors. Since the average total moment of Fe_4 is zero, the sign of its phase factor is irrelevant so far as Eq. (3) is concerned. (In fact, this is true for all antiferromagnetic compounds.)

Ignoring magnetic correlations between red and black tetrahedra, and assuming that all 8-atom compounds have positive average total moments, we can set all phase factors equal to $+1$, thereby defining our provisional model. According to this model, the magnetic properties of the alloys are very similar to those of compounds having the same compositions. This can be seen in Table II, where the average total moments per atom for the compounds and alloys are listed in the columns headed $\mu^{\mathrm{avg}}_{\mathrm{comp}}$ and $\mu^{\mathrm{avg}}_{\mathrm{alloy}}$. The rms moments per atom for the compounds and corresponding alloys are also listed here in the columns headed $\mu^{\mathrm{rms}}_{\mathrm{comp}}$ and $\mu^{\mathrm{rms}}_{\mathrm{alloy}}$. All of these moments were calculated at the interpolated Wigner-Seitz radii appropriate to each composition, which are also shown.

It is clear that the alloy moments differ from the corresponding compound moments only to the extent that compositional averaging is included in the definition of the former [cf. Eq. (3)]. This averaging tends to smooth out the composition dependence, lowering high moments and raising low moments slightly. However, this smoothing is a relatively minor effect which fails to account for the large changes in average total moments that are known to occur for Mn-Ni (but not Fe-Ni) order-disorder transitions.[5]

TABLE II. Average total and rms moments for Mn-Fe-Ni compounds and disordered alloys. The atomic compositions are denoted by c_{Mn}, c_{Fe}, and c_{Ni}. All moments are listed at the interpolated Wigner-Seitz radii, R_{WS}, which are given in bohr units. The average total and rms moments for the compounds and alloys appear in the remaining columns. The inclusion of short-range magnetic order leads to the moments denoted by $\hat{\mu}^{\mathrm{avg}}_{\mathrm{alloy}}$. The bottom row refers to the permalloy-MnFe interface.

q	Compound	c_{Mn}	c_{Fe}	c_{Ni}	R_{WS}	$\mu^{\mathrm{avg}}_{\mathrm{comp}}$	$\mu^{\mathrm{avg}}_{\mathrm{alloy}}$	$\hat{\mu}^{\mathrm{avg}}_{\mathrm{alloy}}$	$\mu^{\mathrm{rms}}_{\mathrm{comp}}$	$\mu^{\mathrm{rms}}_{\mathrm{alloy}}$
1	Mn_4	1	0	0	2.70	0.00	0.00	0.00	2.09	2.09
2	$\mathrm{Mn}_3\mathrm{Fe}_1$	$\frac{3}{4}$	$\frac{1}{4}$	0	2.69	0.32	0.16	0.02	1.68	1.69
3	$\mathrm{Mn}_2\mathrm{Fe}_2$	$\frac{1}{2}$	$\frac{1}{2}$	0	2.68	0.00	0.20	0.05	1.64	1.64
4	$\mathrm{Mn}_1\mathrm{Fe}_3$	$\frac{1}{4}$	$\frac{3}{4}$	0	2.67	0.50	0.23	0.02	1.63	1.64
5	Fe_4	0	1	0	2.66	0.00	0.00	0.00	1.35	1.35
6	$\mathrm{Mn}_3\mathrm{Ni}_1$	$\frac{3}{4}$	0	$\frac{1}{4}$	2.68	0.25	0.16	0.02	1.84	1.85
7	$\mathrm{Mn}_2\mathrm{Fe}_1\mathrm{Ni}_1$	$\frac{1}{2}$	$\frac{1}{4}$	$\frac{1}{4}$	2.67	0.40	0.33	0.20	1.76	1.77
8	$\mathrm{Mn}_1\mathrm{Fe}_2\mathrm{Ni}_1$	$\frac{1}{4}$	$\frac{1}{2}$	$\frac{1}{4}$	2.66	0.23	0.61	0.47	1.79	1.80
9	$\mathrm{Fe}_3\mathrm{Ni}_1$	0	$\frac{3}{4}$	$\frac{1}{4}$	2.65	1.83	1.16	0.79	1.72	1.74
10	$\mathrm{Mn}_2\mathrm{Ni}_2$	$\frac{1}{2}$	0	$\frac{1}{2}$	2.65	0.00	0.37	0.09	1.72	1.75
11	$\mathrm{Mn}_1\mathrm{Fe}_1\mathrm{Ni}_2$	$\frac{1}{4}$	$\frac{1}{4}$	$\frac{1}{2}$	2.64	0.27	0.65	0.52	1.69	1.71
12	$\mathrm{Fe}_2\mathrm{Ni}_2$	0	$\frac{1}{2}$	$\frac{1}{2}$	2.63	1.57	1.37	1.28	1.69	1.71
13	$\mathrm{Mn}_1\mathrm{Ni}_3$	$\frac{1}{4}$	0	$\frac{3}{4}$	2.63	1.06	0.65	0.32	1.37	1.42
14	$\mathrm{Fe}_1\mathrm{Ni}_3$	0	$\frac{1}{4}$	$\frac{3}{4}$	2.62	1.14	1.09	1.09	1.32	1.37
15	Ni_4	0	0	1	2.60	0.62	0.62	0.62	0.62	0.62
	$\mathrm{Mn}_2\mathrm{Fe}_3\mathrm{Ni}_3$	2/8	3/8	3/8	2.65	0.25	0.65	0.54	1.77	1.78

In order to develop a more realistic statistical theory of Mn-Fe alloys, we must take short-range magnetic correlations into account. The first of these is the tendency of nearest-neighbor Mn atoms to have antiparallel spins.[5] Antiferromagnetic coupling also occurs for nearest-neighbor Mn-Fe pairs. To illustrate the experimental evidence for antiferromagnetic coupling between nearest-neighbor Mn-Mn pairs, consider Ni-rich–disordered Mn-Ni alloys. Starting with pure Ni and adding Mn, we find that the average total moment per atom first increases and then decreases, falling to 0 at about 30 at. % Mn. The standard interpretation of these results is as follows. For dilute concentrations, the Mn atoms are far apart, so they will couple ferromagnetically to the host Ni atoms. Since a Mn moment is larger than the Ni moment it replaces, the average total moment will increase at first with increasing Mn content. However, as the Mn content continues to increase, the Mn atoms get closer together, and increasing numbers become nearest neighbors. Since nearest-neighbor Mn atoms have antiparallel spins, their net contribution to the average total moment is zero. As more and more nearest-neighbor Mn pairs form, the average total moment decreases. By the time we reach disordered MnNi_3, the average total moment is zero. On the other hand, for ordered MnNi_3, there are no nearest-neighbor Mn pairs, and the average total moment is substantial. Similar arguments suggest antiferromagnetic coupling between Mn and Fe atoms.

The results of our earlier studies of 4-atom compounds are consistent with this picture of antiparallel nearest-neighbor Mn spins. In all 4-atom compounds containing an even number of Mn atoms, all Mn atoms in the unit cell are antiferromagnetically coupled to one another. In $\mathrm{Mn}_3\mathrm{Fe}$ and $\mathrm{Mn}_3\mathrm{Ni}$, the moment of one of the three Mn atoms is larger than the moments of the other two. These two Mn atoms have equal moments but signs opposite to that of the other one. This arrangement is the best accommodation that the three frustrated Mn atoms can make, since this arrangement tends to minimize the average total Mn moment. Nevertheless, we see that two of the three Mn atoms are antiferromagnetically coupled. The spin patterns for compounds containing both Mn and Fe also suggest that Mn and Fe atoms prefer antiparallel spin alignments (cf. Table I).

We can take these antiferromagnetic tendencies of Mn and Fe into account by exploiting the phase factors that we have already built into our formalism. We will continue to assume that the atomic moments of each tetrahedron as given by Eq. (1) are fixed, but we will change the signs of phase factors to satisfy the antiferromagnetic tendencies of Mn and Fe atoms in the red and black tetrahedra, when they are brought together to form 8-atom compounds. In this way we can express magnetic correlations between red and black atomic tetrahedra entirely in terms of their phase factors.

As an example, consider red and black atomic tetrahedra having only one Mn atom each, and consider all possible 8-atom compounds formed by bringing the red and black tetrahedra together with all possible relative spatial orientations. It is easily seen that there are 3 times as many 8-atom compounds having red and black Mn atoms as nearest neighbors as next-nearest neighbors. For some

8-atom compounds, the red and black Mn atoms already have opposite spin, so the red and black phase factors can both be set equal to +1. However, for other 8-atom compounds, where the red and black Mn atoms start out with parallel spin, it would be necessary to flip one of the Mn spins $\frac{3}{4}$ of the time (nearest-neighbor situation), and retain the original parallel spins $\frac{1}{4}$ of the time (next-nearest-neighbor situation). Flipping a Mn spin implies flipping the spins of the remaining three atoms in its tetrahedron as well.

If we proceed in this fashion, and treat tetrahedra containing three Mn atoms similarly, thereby avoiding the complexities of frustrated Mn spin arrangements, we find that the average total moments of the alloys are reduced only slightly in the Mn-rich regions, and remain the same in the Fe- and Ni-rich regions. In the case of concentrated disordered Mn-Fe-Ni alloys (as opposed to ordered compounds), it becomes difficult to estimate the relative number of nearest- and next-nearest neighbor Mn atoms. Accordingly, we will ignore this distinction and treat all red and black Mn atoms as nearest neighbors, dropping the $\frac{3}{4}:\frac{1}{4}$ ratio mentioned earlier. This procedure reduces the average total moments of many Mn-rich alloys by an additional 25%, but does not, for example, suppress the magnetism of disordered $MnNi_3$ as experiment dictates. In order to reduce the average total moment of $MnNi_3$ and other Mn-containing alloys still further, it is necessary to investigate additional magnetic correlations.

Where are such correlations most likely to arise? If we return to Eq. (3) and take into consideration the fact that the magnetic properties of the atomic tetrahedra could be influenced by their local environments, we can imagine that some environments would favor positive and others negative phase factors. Such effects would be least likely for ferromagnetic tetrahedra embedded in ferromagnetic environments, and most likely for weakly magnetic tetrahedra embedded in weakly magnetic environments. Clearly, environmental averaging can reduce or eliminate certain terms in Eq. (3) because of partial or total cancellation between positive and negative phase factors.

With these considerations in mind, we will refine our previous model by adding the following feature. In all 8-atom compounds containing a 4-atom antiferromagnetic compound, this represents such a weak magnetic environment that its partner compound could have moments that are just as likely to be positive or negative. By summing over all occurrences of such pairs, we obtain a zero-net moment, since the first member has zero moment by virtue of being antiferromagnetic, and the second has zero moment due to cancellation of positive and negative phase factors.

Using this refined model, we have repeated the earlier calculations obtaining average total moments for the alloys which are listed in Table II under the heading $\hat{\mu}_{\text{alloy}}^{\text{avg}}$. In the Mn-rich region, these alloy moments are generally smaller than the alloy moments $\mu_{\text{alloy}}^{\text{avg}}$ obtained by ignoring magnetic correlations. Both sets of moments incorporate compositional averaging, thus distinguishing them from the moments for the ordered compounds, $\mu_{\text{comp}}^{\text{avg}}$. Similar comparisons of average alloy moments could be made for arbitrary compositions, and in fact the last line of Table II shows such a comparison for the composition corresponding to the permalloy-MnFe interface (see the following).

Our statistical theory provides a simple framework for investigating other plausible descriptions of magnetic correlations. Clearly, it would be more satisfactory to base the theory on the outcome of spin-polarized self-consistent calculations for 8-atom compounds, but such results are not available. It will be interesting to see whether future 8-atom calculations will bear out the approximations we have made above.

Overall, our refined model provides a satisfactory picture of the magnetic phase diagram as shown in Fig. 1. The Mn-rich region of the phase diagram is weakly ferrimagnetic or antiferromagnetic, while the Fe- and Ni-rich regions are ferromagnetic. The fact that the antiferromagnetic region extends closer to pure Fe than to pure Ni reflects the greater weakening of the Fe moments (relative to Ni) as the Mn content is increased. The average magnetic moment of disordered $MnNi_3$ has been reduced by a factor of 3 relative to ordered $MnNi_3$, and further reduction is possible by the inclusion of additional magnetic correlations. The changeover from ferromagnetic to antiferromagnetic behavior and the region of spin-glass behavior[14] occur in that range of the phase diagram where there are roughly three Mn atoms per 8-atom unit cell. This can be understood qualitatively by examining the delicate spin balances and resultant frustrations that describe this region.

MAGNETIC STRUCTURE OF PERMALLOY–MnFe-ALLOY INTERFACES

In an earlier paper,[2] we investigated the atomic spin arrangements at (001) interfaces between ordered Ni_3Fe and Mn_2Fe_2 compounds in a prototypic study of a ferromagnetic-antiferromagnetic interface. In that paper, we carried out spin-polarized self-consistent linearized-muffin-tin-orbital band-structure calculations for superlattices consisting of two layers of Ni_3Fe alternating with two layers of Mn_2Fe_2. The repeat period consisted of four successive layers with two atoms per layer. Hence there are eight atoms per unit supercell. Consistent with the specified atomic compositions of the alternating slabs, we can construct three geometrically inequivalent superlattices:

(i) FeNi/NiNi//FeFe/MnMn or (e)/(f)//(b)/(a) ,

(ii) FeNi/NiNi//MnMn/FeFe or (e)/(f)//(a)/(b) ,

(iii) FeNi/NiNi//MnFe/MnFe or (e)/(f)//(c)/(c) ,

where symbols such as (a) refer to the various panels in Fig. 3 and double strokes represent the interfaces. The atomic moments obtained in Ref. 2 for these three structures are listed in Table III.

Since each atom lies at the interface, the net or average moment at the interface is given by the average moment for each case ($0.70\mu_B, 0.55\mu_B, 0.55\mu_B$), the overall average being $0.60\mu_B$. Another way of looking at this problem is to regard the three cases above as members of an 8-atom per unit cell extended model having the chemical

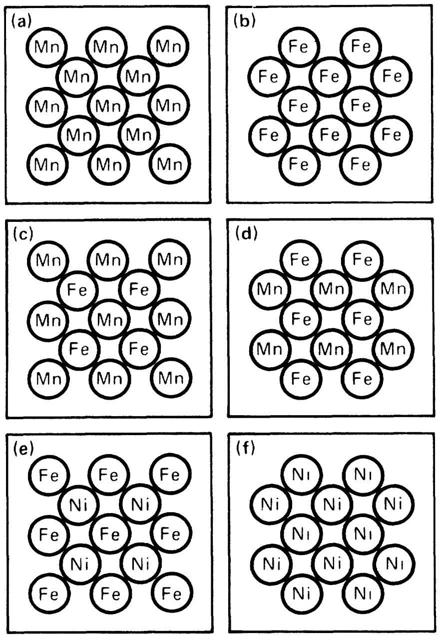

FIG. 3. Atomic layers in $Ni_3Fe/MnFe$ superlattices.

formula $Mn_2Fe_3Ni_3$. In contrast to the 8-atom extended model discussed in the preceding section, where the 8-atom cell is divided into red and black 4-atom cells which form a three-dimensional checkerboard, in the present instance the red and black cells form alternating layers in the [001] direction, each layer being all red or all black. In the preceding section, each cell (or atomic tetrahedron) has six nearest-neighbor cells of the opposite color and none of the same color, while in the present instance there are four nearest neighbors of the same color, and two of the opposite color. The assumptions underlying Eqs. (3) and (5) are more fully justified in the present instance because any given tetrahedron is identical in composition and orientation to four of its six nearest neighbors. Because of the multilayer structure, there are fewer geometrical possibilities for orientational fluctuations, and so only three different cases arise.

For both the checkerboard and multilayer geometries, the compound $Mn_2Fe_3Ni_3$ lies midway between $MnFe_2Ni$ and $MnFeNi_2$ in the phase diagram.[1] As can be seen from Ref. 1, the Ni moment is nearly zero at composition 2:3:3, and the Mn and Fe moments are opposite in sign, the Mn moment being slightly larger than the Fe moment. The average total moment given by Eq. (1) is $0.25\mu_B$, while the more realistic value given by Eq. (3) is $0.63\mu_B$ or $0.54\mu_B$, depending on the treatment of magnetic correlation effects. Note that these values bracket the overall average moment given by our earlier calculation, $0.60\mu_B$. On the other hand, there is a modest difference in the rms moments per atom, these being $1.77\mu_B$ or $1.78\mu_B$ according to the statistical theory, and $1.36\mu_B$ according to the earlier superlattice calculations. These comparisons are sufficiently favorable, however, to suggest that our statistical approach could be used more generally to predict the average magnetic structure of interfaces having different local stoichiometries or different crystallographic orientations.

We can conclude from both sets of calculations that the hybrid nature of the atomic environment at the interface leads to a ferrimagnetic transition layer between the ferromagnetic permalloy and antiferromagnetic MnFe regions. This feature has been largely ignored in discussions of this structure but should be taken into account in treating the magnetic properties of this interface. It can also be seen from the superlattice calculations that there is a significant fluctuation in the interfacial magnetic properties, since these depend on the precise local geometrical arrangement.

As indicated in our earlier paper,[2] the number of different crystal structures that must be considered goes up rapidly as we consider ordered $Ni_3Fe/MnFe$ superlattices containing larger numbers of atoms per layer and/or larger numbers of layers. Studies of such extended superlattices could provide more detailed information about fluctuations in the interfacial magnetic properties,

TABLE III. Individual atomic moments, average total moments, and rms moments (in μ_B/atom) for ordered $Ni_3Fe/MnFe$ superlattices I, II, and III (adapted from Ref. 2).

					Superlattice I					
Atom i	Fe	Ni	Ni	Ni	Fe	Fe	Mn	Mn	$\mu_{\text{super}}^{\text{avg}}$	$\mu_{\text{super}}^{\text{rms}}$
$\mu(i)$	2.34	0.34	0.63	0.57	2.09	1.83	−1.17	−1.03	0.70	1.43
					Superlattice II					
Atom i	Fe	Ni	Ni	Ni	Mn	Mn	Fe	Fe	$\mu_{\text{super}}^{\text{avg}}$	$\mu_{\text{super}}^{\text{rms}}$
$\mu(i)$	2.41	0.46	0.28	0.24	−1.00	−1.16	1.54	1.63	0.55	1.30
					Superlattice III					
Atom i	Fe	Ni	Ni	Ni	Mn	Fe	Mn	Fe	$\mu_{\text{super}}^{\text{avg}}$	$\mu_{\text{super}}^{\text{rms}}$
$\mu(i)$	2.36	0.30	0.53	0.42	1.26	1.92	−1.54	−0.84	0.55	1.34
					Superlattice (average)					
									$\mu_{\text{super}}^{\text{avg}}$	$\mu_{\text{super}}^{\text{rms}}$
									0.60	1.36

but these studies would be exceedingly tedious, considering the large number of distinct geometrical arrangements that must be taken into account with 16-atom supercells, for example. It would be far better to carry out coherent-potential-approximation (CPA) alloy calculations and avoid dealing with individual structures separately. But until extended superlattice calculations or self-consistent spin-polarized CPA calculations are performed for ternary alloys, our simple statistical theory, based on the results for 15 ordered 4-atom compounds, provides a useful interim solution for average interfacial magnetic properties.

DISCUSSION

The present theory is concerned primarily with average magnetic properties and ignores various types of magnetic fluctuation effects that may occur in principle but would be very difficult if not impossible to estimate reliably by present-day methods.

Firstly, it is assumed that all atoms in each ternary Mn-Fe-Ni compound and alloy lie on a common fcc lattice. Thus our models explicitly ignore local magnetovolume fluctuations associated with the slightly different sizes of the various chemical species, as well as the slightly different sizes these species will have when magnetized to different degrees. As can be seen from Table II, R_{WS} is 2.60 and 2.70 bohrs for Ni in fcc Ni and for Mn in fcc Mn, respectively. Since the average value of R_{WS} is 2.65 bohrs for the interface ($Mn_2Fe_3Ni_3$), it is clear that local atomic size fluctuations are of the order of $\pm 2\%$.

Secondly, in representing the interfacial region by a ternary compound or alloy having a suitably averaged lattice constant, we have also ignored the interfacial strain arising from the fact that the bulk lattice constants of Ni_3Fe and MnFe differ by 2%. Thirdly, we have ignored atomic diffusion across the interface, which could, in principle, occur in real systems, and which could change the stoichiometry and the geometrical width of the interface. In spite of these simplifications, we believe that our interface model provides a reasonable estimate of the average magnetic properties of the interface, though we would not be surprised to discover that magnetic fluctuations of the order of 5–10% occur at the interface due to the effects just mentioned.

Finally, there is the intriguing question of multiple magnetic solutions. A number of authors[15] have predicted co-existing magnetic phases for various magnetic elements by using constrained-moment methods. If the various Mn-Fe-Ni compounds and alloys that we have studied could also support multiple-magnetic solutions, it would be logical to average over these solutions in addition to all the other averaging that we have carried out, thereby modifying our final estimates. In the absence of compelling experimental or theoretical evidence indicating the existence of multiple magnetic solutions in the majority of these materials, we decided to deal only with the most stable magnetic solution for each material (see, for example, the discussion of Fe_4 above). A related question that remains unresolved is whether multiple magnetic solutions are favored or suppressed by structural disorder.

In short, the atomic arrangements and magnetic structures of bulk alloys and interfacial regions could be considerably more complex than our model contemplates. We have carried the analysis for average magnetic properties about as far as one can in practical terms using current theoretical and computational methods. There is clearly considerable room for further refinement of our models, but this will require further advances in theoretical and computational techniques.

CONCLUDING REMARKS

In this paper we have demonstrated how disordered fcc Mn-Fe-Ni alloys can be investigated rather simply, starting with the results of magnetic calculations for ordered fcc Mn-Fe-Ni compounds. Because the magnetic character of Mn is considerably different from that of Fe and Ni, the Mn-Fe-Ni system has a far richer magnetic behavior than simpler systems such as Fe-Ni. The two essential ingredients of our model are compositional averaging of the magnetic properties of atomic tetrahedra and inclusion of plausible magnetic correlations between adjacent atomic tetrahedra. To make substantial progress beyond our model would require extensive computational effort, orders of magnitude greater than we have expended. It is gratifying that the present statistical theory corroborates key features of the magnetic structure of permalloy-MnFe interfaces deduced earlier by superlattice methods.

It would also be of considerable interest to study the magnetic properties of the Mn-Fe-Ni system at finite temperatures, as has already been done for ferromagnetic-antiferromagnetic Co/Cr superlattices.[16] Other key problems bearing on the Mn-Fe-Ni system include the Invar problem[17] as well as the region of the phase diagram where spin-glass behavior has been observed.[14] The present statistical theory could also be used to deal with such problems.

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[9] Recall that within the framework of local-spin-density-functional theory, with the spin-orbit interaction absent, the spin-up and spin-down directions are arbitrary. We could just as well have multiplied the four atomic moments for each compound q listed in Ref. 1, Table I by -1 and obtained equally valid sets of moments. (Of course the relative signs for each q are uniquely determined by our electronic structure calculations.) In specifying the common signs in Ref. 1, Table I, we made arbitrary choices for compounds with zero average total moment. For the remaining compounds, we chose the common signs that yielded positive rather than negative average total moments.

[10] In dealing with the 15 compounds q individually, the orientational index r is irrelevant, and the magnetic index may be set equal to $+1$ or -1 depending on whether we want the atomic moments listed in Ref., 1, Table I, or their negatives. On the other hand, if we wish to average some alloy property over all atomic tetrahedra, or if we wish to discuss the influence of neighboring tetrahedra on one another, relative spatial orientations and relative magnetic orders should be taken into account, at least in principle. The numbers of distinct chemical arrangements are 12 for 2:1:1 compounds such as Mn_2FeNi, 6 for 2:2 compounds such as Mn_2Fe_2, 4 for 3:1 compounds such as Mn_3Fe, and 1 for Mn, Fe, and Ni. All of these numbers are doubled if magnetic order ($m=\pm 1$) is also taken into account.

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[12] For more recent discussions, see, for example, M. W. Long and U. Steigenberger, Phys. Scr. **T25**, 220 (1989); also M. S. Seehra and T. M. Giebultowicz, Phys. Rev. B **38**, 11 (1988); **38**, 898 (1988).

[13] Whether Fe_4 is ferromagnetic or antiferromagnetic when forming part of an 8-atom compound, Fe_4 has a weight of 1 in averages taken over the 81 possible 4-atom compounds, allowing for all arrrangements of the four atoms in the unit cell. Therefore, its contribution to the magnetic properties of the Mn-Fe-Ni system is rather minor, except very near pure fcc Fe, where, of course, the equilibrium crystal structure is not fcc in any event. In taking averages over all 4-atom compounds, we will include fcc Fe_4 for the sake of completeness.

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