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Spontaneous local symmetry breaking in frustrated antiferromagnets: non-collinear impurities in MnCu alloys?

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Abstract. Using impurity calculations in the classical Heisenberg model, we show that paramagnetic impurities doped into collinear frustrated antiferromagnets can lead to the spontaneous formation of localised spin components orthogonal to the original quantisation direction. We suggest how transitions from collinear to non-collinear phases might occur as a long-range phase coherence between locally stable non-collinear impurities. There is some recent diffuse neutron scattering evidence for non-collinear impurities in single-domain crystals of MnCu and we calculate the diffuse scattering from our model as a prediction for possible future experimental verification.

1. Introduction

Transition metal alloys based on γ-Mn show very interesting magnetic behaviour. Manganese quenched into a face centre cubic structure exhibits 'type I' antiferromagnetism. This is a sequence of ferromagnetic x–y layers which alternate in spin direction along the z axis. The z axis becomes inequivalent to the other Cartesian directions and the magnetism induces a huge tetragonal distortion of approximately six per cent parallel to the z axis [1]. Neutron scattering shows that the spins align parallel to the z axis where they are held in place by spin–orbit coupling. The fundamental physical picture is of strong short-range antiferromagnetic interactions between spins, leading to long-range antiferromagnetism at about 500 K, corresponding to a spin wave bandwidth of $\sim$120 meV [2], together with a weak coupling to the lattice directions corresponding to a spin wave gap of about $\sim$4–6 meV [3].

When another transition metal is doped into the manganese there are quite dramatic changes in behaviour. Fe [4], Ir [5], Ni [6] and Cu [7] all substantially reduce the tetragonal distortion and for Fe, Ir and Ni there is evidence of a cubic phase which is stabilised at doping concentrations of approximately a quarter. For the Mn–Ni system a sequence of four phases is observed as a function of concentration [6]. This behaviour is being explained in terms of spin reorientation, with spins rotating away from the z axis and into the x–y plane [8].

The face centre cubic lattice is antiferromagnetically frustrated. Most simple lattices are bipartite, which means that there are two natural sublattices with *all* the nearest neighbours of one sublattice on the other sublattice. Antiferromagnetism is unfrustrated on a bipartite lattice, since the spin configuration with spins pointed

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in opposite directions on the two natural sublattices finds *all* nearest neighbours antiparallel. The face centre cubic lattice is *not* bipartite and in any spin configuration only a fraction of nearest neighbours can be made simultaneously antiparallel, the rest are not antiparallel and so are *frustrated*. For the face centre cubic lattice, the maximum possible fraction of antiparallel spins is two thirds, with the other third being parallel (appendix A). This bound is achieved in type I antiferromagnetism. The crucial observation for this topology is that there are other spin configurations which also achieve this bound. Indeed a linear superposition of type I antiferromagnets ordered parallel to different Cartesian directions also achieves the bound. These are the multiple-$\boldsymbol{Q}$ states [9] and they necessarily contain *non-collinear* spins. The magnetic symmetry group of these states is variable and the triple-$\boldsymbol{Q}$ state, an equal amplitude sum of the collinear states in each of the three Cartesian directions, has cubic symmetry. The cubic antiferromagnet found in the γ-Mn alloys is explained as a triple-$\boldsymbol{Q}$ state [8].

In previous work, the questions: 'How might the different multiple-$\boldsymbol{Q}$ states be experimentally separated?' [10] and 'What physical phenomena might stabilise the different phases?' [11] have been studied. In this paper we look at the question of how the system might transform from one phase to another, and in particular whether or not the transition would be sharp. The motivation came from a diffuse scattering experiment [12]. Diffuse scattering from a single-domain crystal of MnCu seems to indicate *non-collinear* spin impurities in a system with pure collinear long-range order. This is a very surprising result because it violates a local symmetry of the system.

Collinear magnets have a quantisation direction to which all the spins in the system are parallel. Spin rotations about this direction constitute local symmetries. Further, substitutional inclusions of paramagnetic impurities maintain these local symmetries, since a paramagnetic atom is symmetric under spin rotations in *all* directions. Non-collinear spins involve a breaking of these symmetries, and the question becomes: 'By what physical mechanism is the symmetry broken?'. There is the possibility of a phase transition to a new phase with long-range order for the broken symmetry, but are there other possibilities associated with the disorder, and how might precursor phenomena be exhibited?

Non-collinear spins have led to confusion in other systems. For the case of disordered ferromagnets the interest revolves around loss of moment. The atoms have a fairly well defined local moment and the whole moment does not seem to be involved in the long-range phase coherence. Where does the moment go? Initially it was thought that the local moment was reduced, but more recently it has been suggested that *non-collinear* fluctuations are the cause. The idea is that short-range atomic order sets up a local lattice symmetry parallel to which spin–orbit coupling tries to align the moments. Longer-range variations in this local direction then cause a non-collinear component which can explain the loss of moment [13]. In this paper we will present an argument for loss of average moment into other directions caused by the local release of antiferromagnetic frustration, a quite distinct physical phenomenon.

There is disorder in the experimental alloys we are considering. Although the atomic positions are regular, the impurities are fairly random with only a certain amount of short-range order. We do not believe that the effects of the disorder are as crucial as in disordered ferromagnets, but neither do we believe that the transitions are as smooth as a pure theory suggests. It has previously been shown that for a pure theory a spin wave mode would soften at a spin reorientation phase transition [14]. One question addressed in this paper is whether spin wave softening would be expected to survive the inclusion of disorder into the description.

The antiferromagnetic interactions are very much stronger than the forces that align the moments with the lattice directions. Any purely magnetic phenomena which cause non-collinear spins can therefore easily dominate. We suggest the formation of localised bound states of multiple-$\boldsymbol{Q}$ distortions as a source for loss of moment and of non-collinear effects.

The mechanism that we propose requires *several* impurities in order to contribute and so we give a brief summary of the types of short-range order found in the γ-Mn transition metal alloys. For the case of MnCu, *clustering* of impurities is observed [15]. Indeed there has been a huge amount of work on the order–disorder transition in this system [16]. Although the tetragonal distortion is reduced as Cu is introduced, the antiferromagnetic moment also decays and indeed vanishes before the advent of the cubic phase. For the case of MnNi, however, the impurities *anti-cluster* [17]. There is a low probability of finding nearest-neighbour impurities and an enhanced probability of finding next-nearest-neighbour impurities. The tetragonal distortion is destroyed in the presence of the antiferromagnetism for this alloy although two other phases are traversed before the cubic antiferromagnet is found. Can we understand the differing stabilities of the cubic phase in these two alloys by considering the effects of short-range order on the magnetism? We consider both clustering and anti-clustering in this paper and compare the suggested pictures for a spin reorientation phase transition.

There is a further clustering consideration and that is: 'What causes the clustering?'. If the magnetic phenomena that we describe are instrumental in the clustering then we ought to consider *low*-energy clusters. In fact the quenching and melting temperatures are quite high in comparison to the magnetic energy scale and the clustering probably occurs for a 'coarser' reason. We do however consider the local energy loss per impurity in our clusters in order to compare them on these grounds. One final subtlety here is that long-range magnetic phase coherence is *not* established at quenching temperatures and so any magnetically induced clustering might be better suited to a different quantisation direction than that for the observed long-range coherence.

It is not clear how these alloys should be modelled. The first row transition series includes itinerant magnetism, but even itinerant systems show evidence of local moments above their transition temperatures. We will model γ-Mn alloys with a *local moment* description with fixed moments of length $2\mu_B$ [18]. The main reason is that such a description is easy to work with and more sophisticated questions can be addressed. We believe, however, that the nearest-neighbour configuration of atoms in the face centre cubic lattice prefers local moment behaviour to itinerancy.

It is also not clear how the impurities should be described in these systems. The charge and spin states of the dopant atoms are unknown but there is fairly direct evidence that the total spin of the nickel dopant is zero [18] and simply counting the electrons in the known copper charge states suggests either spin zero or spin half. We will model the impurities by assuming that the moments are much reduced, but otherwise do not affect the host atoms. If itinerancy were dominant then we might expect the surrounding manganese spins to be altered in length to compensate for the impurity. We accept this possibility but it is not clear how to include it and so we neglect it.

In section 2 we look at various clusters of impurities in the collinear 'type I' antiferromagnet. We also show the possibility of spontaneous local symmetry breaking. In section 3 we deduce a consistent picture for a phase transition based around the non-collinear impurities and in section 4 we make an attempt at describing the likely signatures of the phenomenon.

2. Impurity calculations

We employ the classical limit of the Heisenberg model in our calculations:

$$H = J \sum_{[ii']} \boldsymbol{S}_i \cdot \boldsymbol{S}_{i'} \tag{2.1}$$

where the coupling is antiferromagnetic ($J > 0$), the spins are of fixed length ($\boldsymbol{S}_i \cdot \boldsymbol{S}_i = S^2$), we include only nearest-neighbour bonds (denoted by $[ii']$) and each bond is included once only. We restrict our attention to zero temperature and the search for the ground state. This reduces the problem to determining the preferred *orientations* of the spins.

Impurity spins are included by altering the lengths of some of the spins. We force all impurities to have the same spin length of κS but allow κ to vary. This allows us to compare reduced moments with zero moments. Experiment suggests that the moment on the nickel atoms is small [18], but the copper moment is unclear and we should allow for the possibility of a spin $\frac{1}{2}$ copper atom, corresponding to $\kappa \sim \frac{1}{2}$ for a saturated manganese moment of about $2.2\mu_B$.

In the absence of impurities the ground state of the face centre cubic lattice is multiply degenerate and, although this drives the effects we are studying, it also causes severe interpretational problems. The inclusion of an impurity breaks this degeneracy and stabilises a *non-collinear* phase [19]. In competition with this effect in the real materials are quantum mechanical spin fluctuations and itineracy, namely fluctuations in spin length. Spin fluctuations promote collinearity [19] and itineracy can stabilise any of the spin arrangements [8]. When the concentration of impurities is low, the collinear phase is found in the experimental systems and we include *none* of the effects which stabilise this in our model Hamiltonian! Rather than complicate our description, we stabilise the collinear phase in our choice of *boundary conditions*. We use the collinear phase as a reference state and allow only a few spins around the impurity the freedom to reorientate. The remainder of the spins are held rigidly in the positions dictated by the collinear phase, and the interactions then feed this preferred direction into the cluster. As long as the disturbance decays as a function of distance from the impurity, we feel confident that our calculations make physical sense.

We have considered one impurity previously [19] and we will analyse the single-impurity problem in more detail later. Under the present model assumptions, there is *no* local spin distortion provided that the cluster is small. This is precisely what might be predicted from the local symmetry argument. Each spin sits in the local field of its nearest neighbours and if these spins are collinear then the local field will be collinear. The spins on the surface of the cluster feed a unique quantisation direction into the cluster and unless a local mechanism instigates symmetry breaking the whole cluster will be forced collinear. For non-frustrated bipartite lattices no such mechanism exists and the ground state can be chosen to be collinear *whatever* the number or configuration of impurities. If the cluster in a bipartite lattice is simply connected then the collinear cluster is the unique classical ground state for the given quantisation direction.

For frustrated systems there is a balance between parallel and antiparallel neighbours which can be disturbed by local fluctuations in the configuration of impurities. For the ground state in the absence of impurities, each spin has eight antiparallel and four parallel neighbours. The problems emerge when the impurities happen to

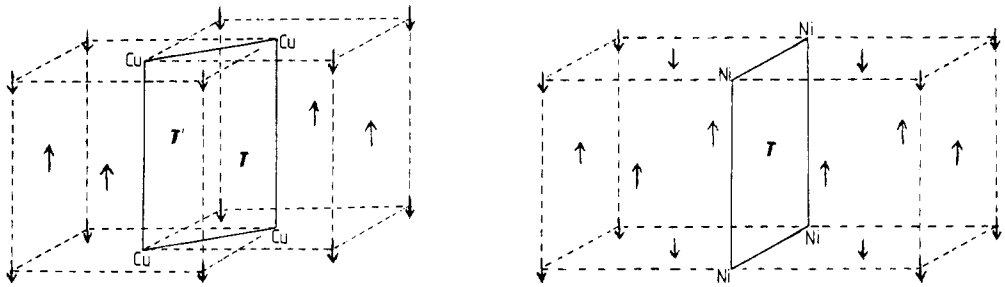

Figure 1. Two simple clusters of impurities which support non-collinear moments. The first is an 'unbalanced' anticluster and the second is a 'balanced' cluster. Both clusters have spins, marked $\boldsymbol{T}$, which are free to rotate with no corresponding change in Heisenberg energy. Primed spins and unprimed spins rotate in opposite directions.

favour one of the two sublattices and locally cancel out the dominance of antiparallel neighbours.

The simplest situations are depicted in figure 1. A careful count of nearest neighbours to the spins marked $\boldsymbol{T}$ shows that there is a balance of four parallel and four antiparallel. The energy of the state remains unchanged for all orientations of spins marked $\boldsymbol{T}$. This energy is the best possible for a collinear arrangement and so any small gain from a non-collinear configuration will stabilise it relative to the collinear arrangement.

There is a simple and often used argument which shows that a non-collinear cluster is stable. If we consider the state where one $\boldsymbol{T}$ is rotated into the plane, then its eight non-impurity nearest neighbours all feel a component of the local field attempting to align them antiparallel to the special spin. If these eight neighbours are infinitesimally rotated, then they achieve a linear gain in energy from the orthogonal component and only lose energy at second order from the collinear component. An infinitesimal rotation is *always* relatively stable and so a non-collinear component is spontaneously created.

There are two types of interpretational difficulty to be overcome in thinking about these non-collinear components. Firstly, in a real dilute alloy, there will be many such impurities with very small interactions between them. At reasonable temperatures we would expect the local orientations of these non-collinear moments to be uncorrelated and hence a macroscopic average would be expected to yield a vanishing non-collinear moment. The possibility of coherence between the non-collinear components will be addressed in the next section. The second difficulty involves the effect of quantum mechanical fluctuations. Quantum mechanics prefers total spin zero ground states over Néel ordered states. For the present example, this would involve averaging the non-collinear component over all possible orientations in the plane. Competing with this are spin–orbit coupling energies which are trying to orient the local order parallel to the crystal axes. We will assume that the time scale for any quantum averaging is slow with respect to the time scale of our experimental probe and hence that the local order is observable.

A study of the second cluster of figure 1 shows that the states where $\boldsymbol{T}$ and $\boldsymbol{T}'$ are constrained to be antiparallel but are orientated as a pair in any direction are also degenerate with the collinear state and make better reference states from which to construct the non-collinear ground state to the cluster.

In case it is thought that all non-collinear clusters have spins for which the numbers

of parallel and antiparallel neighbours balance in the collinear state, in figure 2 we depict two clusters where the maximum number of impurities neighbouring any one spin is three and two respectively and yet the ground states to these clusters are also non-collinear! This may be proven by considering infinitesimal rotations of the spins marked $\boldsymbol{T}$. If $\boldsymbol{T}$ and $\boldsymbol{T}'$ are rotated in opposite directions then the first cluster achieves an energy saving at second order in the angle, whereas the second cluster remains degenerate at this order being destabilised by the coupling to infinitesimal rotations of the other neighbouring spins.

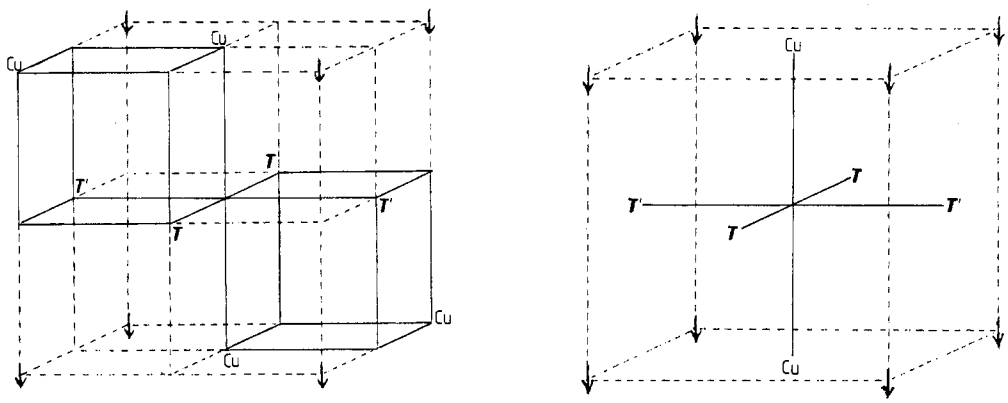

Figure 2. Two clusters which are locally unstable to localised orthogonal Néel fluctuations amongst the spins marked $\boldsymbol{T}$.

In fact even a *single* impurity is unstable to non-collinear fluctuations. The degeneracies of the face centre cubic classical ground state form a one-dimensional family, an order more than the simpler frustrated topology of the triangular lattice for example. The Goldstone modes have corresponding freedom and we find *two-dimensional* Goldstone modes. In the 'single-$\boldsymbol{Q}$' state, infinitesimal orthogonal Néel fluctuations restricted to one ferromagnetic x–y plane are Goldstone modes. Each spin loses energy from its collinear component, from the four net antiparallel neighbours, but gains the same energy from the non-collinear component of the four in-plane neighbours. An impurity in a neighbouring layer acts as a localised attractive potential to the Néel fluctuations, since the spins which neighbour it lose energy from only three net antiparallel neighbours. Any attractive potential in two dimensions *necessitates* a bound state and so the impurity induces two bound states of localised orthogonal Néel fluctuations in the neighbouring planes. About thirty two spins are required to participate in order to stabilise each bound state and the resulting bound states are very long range, with the minor energy gain being unlikely to dominate the effects we have ignored which stabilise the collinear phase. Unless the effect is very short range we will henceforth ignore it.

In this paper we have restricted attention to only one orthogonal component to the quantisation direction. Our reasoning is that firstly, the two-component calculations are simpler, secondly, that the spontaneous appearence of a *third* component would only occur in the case where the first two were locally frustrated—a rare event in the known concentrations doped into collinear alloys—and thirdly, on experimental grounds, the 'double-$\boldsymbol{Q}$' state would seem to be a better state in which to consider fluctuations of the third component.

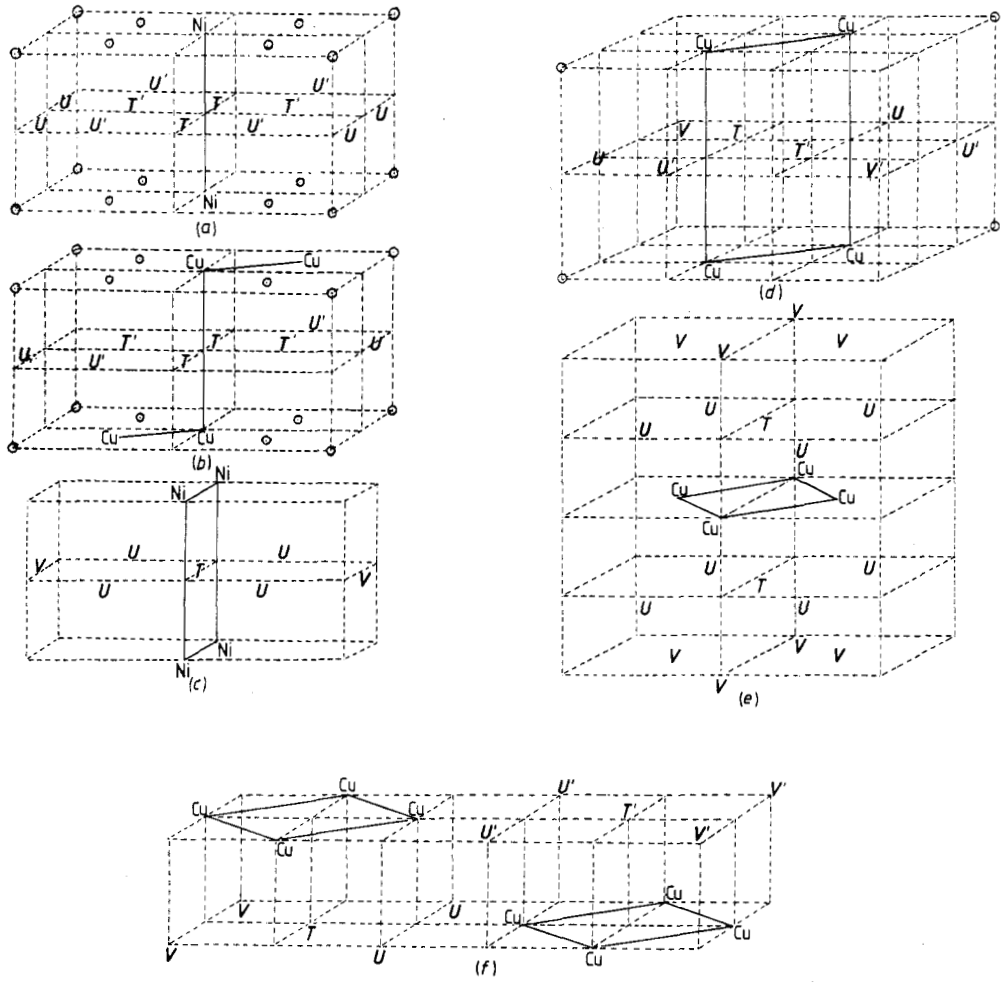

Figure 3. The atomic configurations of the clusters we solve. The spins marked T, U and V are all free to reorientate, but all the other spins are held fixed. The circled spins do not couple to the non-collinear distortion. Primed spins and unprimed spins rotate in opposite directions. The magnitudes of the rotations are presented in figure 4 as a function of the spin scaling factor κ.

We perform cluster calculations on the impurity configurations of figure 3. The calculational details are relegated to appendix B. The angles through which the spins rotate are pictured in figure 4 as a function of the impurity spin length, κ, and the energies of the clusters are quoted in table 1 at $\kappa = 0$, namely for paramagnetic impurities.

As previously noted, spin $\frac{1}{2}$ impurities in a spin 1 host is a possible scenario for copper alloyed into γ-manganese. A cursory inspection of figure 4 immediately indicates that the present model is stable to non-collinear reorientation for $\kappa \sim \frac{1}{2}$. Only the case of paramagnetic copper impurities might be expected to exhibit non-collinear impurities, and so henceforth we will set $\kappa = 0$ and consider spinless impurities.

We chose our clusters according to three criteria: clustering or anticlustering (denoted by Cu and Ni respectively), whether or not the collinear spins were 'balanced' (i.e. for each T there is a T'), and finally according to three instability types, marked

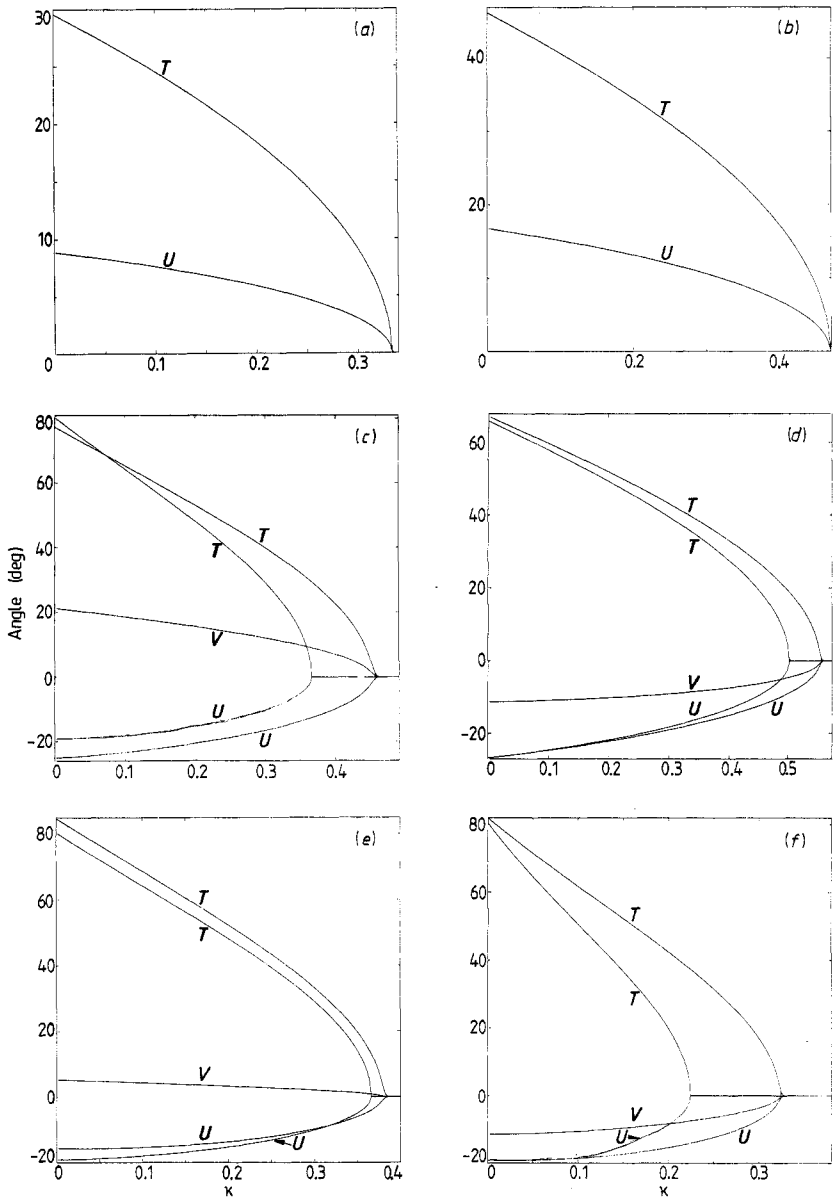

Figure 4. The spin configurations found in our non-collinear cluster calculations. The angles of rotation are plotted as a function of the spin scaling factor, κ, of the doped impurities.

by basic rotations of approximately 0°, 30° and 90°. The final classification comes from solving the '***T***-cluster' and the angle through which the ***T***-spins would rotate with all the other spins held fixed. The final cluster involves two distinct impurities and is a first pass at understanding the interactions between non-collinear impurities.

Cluster (*a*) is the smallest relevant impurity concentration that we have found which leads to short-range non-collinear spins. It is a 'balanced' anticluster which is only just stable with a basic rotation of 0°. The rotations remain modest when the

Table 1. The energies of the non-collinear spin arrangements depicted in figures 3 and 4 for the case where the impurity is paramagnetic (namely $\kappa = 0$). The 'static' contributions, namely those due to the breaking of impurity bonds, for the collinear spin arrangements are given for comparison with the non-collinear energy savings. The cluster marked (*e*)*T*,*U* is that for (*e*) but with the *V* spins held fixed, it is directly comparable with the two-impurity calculation (*f*).

	Energy per impurity (units of JS^2)				
Cluster	Non-collinear state	Collinear state	Non-collinear saving	Collinear state *x*	Collinear state *y*
Impurity	4.000	4.000	0.000	4.000	4.000
(*a*)	3.914	4.000	0.086	4.000	4.000
(*b*)	4.273	4.500	0.227	3.500	3.500
(*c*)	3.769	4.000	0.231	4.000	4.000
(*d*)	4.105	4.500	0.395	3.500	3.500
(*e*)*T*,*U*	4.667	5.000	0.333	3.000	3.000
(*e*)	4.637	5.000	0.363	3.000	3.000
(*f*)	4.742	4.875	0.133	2.875	3.125

surrounding spins are free to reorient and although this is a weak effect it might be argued to dominate when the impurity concentration is low. The energy gain is also modest but should still readily dominate the spin–orbit coupling energy and itinerancy effects restricted to so few spins.

Cluster (*b*) is a 'balanced' cluster with a basic rotation of 30°. This simple picture survives the freeing of surrounding impurities and is surprisingly stable surviving approximately to when the impurity spin is half the length of the host spins. The energies show that this cluster is badly oriented losing a full bond per impurity of 'static' energy. The energy saving from the non-collinear component makes up only a small fraction of this energy and so the quantisation direction would need to be stabilised by similar clusters elsewhere in the crystal oriented in other directions.

Cluster (*c*) is the first example of an 'unbalanced' system. It is also an anticluster with a basic 90° rotation. For a 'balanced' cluster the *total* orthogonal spin component in each plane necessarily vanishes because of the symmetry, but the same is not true for an 'unbalanced' cluster. A careful calculation of this component for this cluster shows that it is almost precisely zero, driven there by the local antiferromagnetic correlations. This fact will prove important in our choice of experimental probe for non-collinear clusters. For comparison, the result for the cluster with *V* fixed is also given and the inclusion of *V* is seen not to change the basic picture. There is no change in the collinear energy if the quantisation direction is changed and the energy saving from the non-collinear component is much larger than for cluster (*a*).

Cluster (*d*) is a 'balanced' cluster with a basic 90° rotation. The decay of the rotation angles as a function of the distance from the impurity is clearly observed. The cluster is very stable being present even when the impurity spins are half the length of the host spins. The collinear energetics of this cluster are the same as cluster (*b*) but the non-collinear impurity is almost twice as stable.

Cluster (*e*) is an 'unbalanced' cluster with a basic 90° rotation. Once again the total orthogonal component in each plane is small, although not negligible for this cluster which is rather small. The cluster is very badly oriented costing two bonds per impurity and although the non-collinear saving is more than in cluster (*b*), it is still only minor.

The coupled impurity calculation of (*f*) shows that firstly, the coupling is strong

with the intermediary spins taking advantage of the second cluster, and secondly, that the two clusters are *coherent* with the symmetry of a single-$\boldsymbol{Q}$ state aligned parallel to the Cartesian direction joining the two clusters. The 'static' contribution also favours the single-$\boldsymbol{Q}$ phase suggested by the non-collinear impurity. The single impurity of cluster (e) where V is held fixed is directly comparable with the full two-impurity calculation. The rotations are extremely similar although the non-collinear component achieves a much smaller energy gain.

If we compare all the clusters (a, b, e, f) in table 1 then we see that the clusters which are most susceptible to non-collinear components have also the worst 'static' contributions. These clusters need to be parasitic on other collinear regions of the crystal. The anticlusters (a, c) have degenerate 'static' contributions and the non-collinear effects break the degeneracy suggesting further stability for clusters oriented in the chosen directions. The magnetic energies favour neighbouring impurities on opposite sublattices and this opposes our mechanism. Is this the source of the differences in the behaviours of MnCu and MnNi?

The spin correlations in the orthogonal component are extremely well described as a 'type I' collinear state orthogonal to the quantisation direction. These states are local multiple-$\boldsymbol{Q}$ distortions.

3. Phase transitions

We now move on to an interpretation of our results in terms of what they suggest for the real systems.

If we consider a doping level of 10%, which is low for the type of alloy usually considered, then, if the impurities are independent, the fraction of spins with two or more nearest-neighbour impurities is about $\frac{1}{3}$ and with three or more is about $\frac{1}{9}$. In order for our mechanism to be operable, however, there is also the restriction that the impurities should be on the opposite sublattice and this cuts down the relevant impurity configurations by an order of magnitude. Nevertheless this is the sort of concentration where our model would suggest that a sizable fraction of spins would start to generate non-collinear components.

As the concentration is increased, initially the non-collinear impurities would be well separated. Although in our classical model the impurities yield long-range distortions that would couple all the impurities together and force long-range coherence, in real materials anisotropy and itinerancy would severely weaken the coupling and the impurities would start out being independent at most reasonable temperatures. These impurities would be the precursor phenomenon to a spin reorientation phase transition.

While the impurities are independent, the *actual* direction for the orthogonal component would be chosen to be parallel to a Cartesian direction by the local spin–orbit coupling. As the concentration of impurities is increased, the impurities will start to interact and to align in order to optimise the Heisenberg contributions. The Heisenberg interactions are optimised by keeping all the orthogonal components parallel.

The next idea which becomes relevant is that of 'phase coherence by percolation'. We may consider the non-collinear impurities as randomly distributed, weakly interacting objects. Impurities within a certain range will align and 'droplets' of coherently connected impurities will form. Long-range phase coherence occurs when one of these 'droplets' percolates through the entire system. This is a more satisfying picture for

how a phase transition might occur, but there is one rather important complication. The non-collinear impurities that we have found are tied predominately to one of the two spin sublattices. The coupling of 'balanced' clusters to neighbouring planes is very weak. Indeed the circled atoms in figure 3 do not couple to the calculated impurity at all. There are two natural choices of coupling between layers which correspond to single-$\boldsymbol{Q}$ states in x and y directions respectively. This 'degeneracy' would be broken by any long-range phase coherence and local configuration fluctuations would decide which of the two directions would be favoured. Cluster (f) is an example of just such a configuration. The two clusters are on different sublattices and are coupled to each other via the $\boldsymbol{U}$ spins. $\boldsymbol{U}$ and $\boldsymbol{U}'$ form two neighbouring ferromagnetic chains which are oriented in opposite directions and pick out the single-$\boldsymbol{Q}$ phase oriented parallel to the line joining the impurities. The picture for the phase transition then becomes a competition between two types of 'droplets' which are predominately x- and y-directed single-$\boldsymbol{Q}$ states. Eventually one of the two types of 'droplets' forms the macroscopic cluster and the symmetry is truly broken at the phase transition.

The phase coherence within a 'droplet' would be expected to be the direction of the orthogonal spin component and not the single-$\boldsymbol{Q}$ character. Spin–orbit coupling acting on the dominant single-$\boldsymbol{Q}$ component would decide the direction of the orthogonal component. The minor single-$\boldsymbol{Q}$ component would lose its spin–orbit coupling contribution if the coupling to the 'droplet' dominates, which is to be expected since the coupling is on the exchange energy scale. The 'droplets' ought to be classifiable by the Cartesian direction of the orthogonal component, which might act like an Ising variable in a description of the phase transition. It is important to realise that some of the components would be forced to lie in an orthogonal direction to that suggested by spin–orbit coupling and this is a crucial experimental consideration.

The state that is transitted into by this line of argument is a double-$\boldsymbol{Q}$ phase with a dominant $\boldsymbol{Q}$ corresponding to the original phase coherence and a minor $\boldsymbol{Q}$ corresponding to the coherently coupled impurities. Such a phase would have orthorhombic symmetry and just such a phase is found in the expected place in the MnNi phase diagram [6].

We are now in a position to try to answer the question of whether spin wave softening would survive the inclusion of disorder into the description. The spin wave in the pure description creates a single-$\boldsymbol{Q}$ spin distortion in an orthogonal direction to the existing quantisation direction. At the pure phase transition this spin wave becomes soft and the fluctuations take the state into the new multiple-$\boldsymbol{Q}$ ground state. In our disordered description we have local multiple-$\boldsymbol{Q}$ distortions; the non-collinear impurities. Each impurity might be expected to 'go soft' at different temperatures according to the local configuration of impurities. Local multiple-$\boldsymbol{Q}$ excitations would be expected to be low energy at the condensation of the impurity, but once the impurity becomes fairly stable, the internal excitations would be expected to drift back to higher energies. The residual low-energy excitations would be associated with changes in the boundaries of the clusters and not a volume effect as suggested by the pure transition. We would therefore expect a large quantity of low-energy scattering in the vicinity of the transition and above in temperature but not a well defined collective excitation corresponding to a softened bulk spin wave.

Unfortunately the transition that we predict is too messy to be picked up by something like a spin wave excitation which requires magnetic coherence over long distances to be easily measurable and is therefore likely to become lost in alloy disorder. Instead the transition must be found with a macroscopic bulk experiment,

such as the magnetic susceptibility, specific heat or magnetic Bragg scattering, which are more sensitive to this less homogeneous but sharp type of transition. The other natural line of experimental attack is to try to observe the precursor phenomena in the untransformed material. With this in mind, we look for the signature of our non-collinear impurities with the more local probe of magnetic diffuse neutron scattering in the next section.

4. Diffuse scattering

We now move on to considerations of how we might experimentally verify the existence of the local non-collinear impurities we predict. The natural probe to consider is magnetic diffuse neutron scattering.

Magnetic diffuse scattering supplies information about the Fourier transform of spin distortions or fluctuations away from the average:

$$\delta \boldsymbol{S}(\boldsymbol{K}) = \sum_{\boldsymbol{R}} \exp(\mathrm{i}\boldsymbol{R} \cdot \boldsymbol{K}) \delta \boldsymbol{S}(\boldsymbol{R}) \tag{4.1}$$

where $\delta \boldsymbol{S}(\boldsymbol{R})$ is the change in spin on the site $\boldsymbol{R}$, away from the average spin as suggested by the Bragg scattering. The observed scattering intensity is proportional to [20] the square of the spin component orthogonal to the momentum transfer of the neutron, namely $\boldsymbol{K}$:

$$\partial\sigma/\partial\Omega \propto I(\boldsymbol{K}) = |\hat{\boldsymbol{K}} \times (\delta \boldsymbol{S}(\boldsymbol{K}) \times \hat{\boldsymbol{K}})|^2 \tag{4.2}$$

There are three main considerations: firstly, the short-range structure of the scattering within a single Brillouin zone; secondly, the decay of the scattering as the magnitude of the momentum transfer increases; and thirdly, the angular dependence of the scattering.

The first consideration yields the basic 'symmetry' of the spin distortion around the impurities. If the scattering is peaked at positions corresponding to a regular spin arrangement, then the local distortion has the same symmetry as the relevant spin structure. The special spin structure may correspond to the original spin structure or to a new spin structure which has been locally stabilised by the impurities.

As with Bragg scattering, the decay of the scattering as the momentum transfer is increased yields the magnetic form factor, from which the spatial extent of the orbital harbouring the spin can be deduced. The form factor will not be directly of interest to us, since the spin almost certainly resides in the d orbitals on the manganese atoms. The form factor of these orbitals is well understood and yields a smooth minor correction to the scattering intensity.

The angular dependence of the scattering is dominated by the fact that the scattering is orthogonal to the spin direction. Usually the direction of the spins can be deduced from an angular analysis of the magnetic Bragg scattering, and then an angular analysis of the magnetic diffuse scattering indicates whether or not the distortions are parallel to the underlying spin structure or not. This is precisely what is required in order to demonstrate the existence of a non-collinear component to the spin distortion.

The experimental situation we have in mind, is of a single-domain single crystal of lightly doped manganese which is a collinear antiferromagnet with a sizable c/a ratio

[1]. The normal situation is of a multi-domain single crystal for which even the ground state is ambiguous, but, with a single collinear domain the ground state is well defined, and whether or not there are static local non-collinear distortions is an interesting and perhaps answerable question.

For the present model there are two relevant spin components: the 'parallel' component, including the original component which is lost into the non-collinear component and the impurity spins, and the non-collinear component itself. Assuming that the original spins are parallel to the z axis and that the non-collinear component is parallel to the x axis, then the magnetic diffuse scattering can be reduced to

$$I(\boldsymbol{K}) = |\delta S_{\parallel}|^2 + |\delta S_{\perp}|^2 - |\delta S_{\parallel}\hat{K}_z + \delta S_{\perp}\hat{K}_x|^2 \tag{4.3a}$$

$$I(\boldsymbol{K}) = |\delta S_{\parallel}\hat{K}_x - \delta S_{\perp}\hat{K}_z|^2 + \hat{K}_y^2(|\delta S_{\parallel}|^2 + |\delta S_{\perp}|^2) \tag{4.3b}$$

in terms of the parallel, $\delta S_{\parallel}$, and perpendicular, $\delta S_{\perp}$, spin components.

The most obvious test for non-collinearity involves looking at momentum transfers parallel to the original spin direction. The scattering reduces to $|\delta S_{\perp}|^2$, which vanishes in the absence of a non-collinear component. Unfortunately, for our model, the perpendicular component in this direction is relatively modest. The cause is fundamental to the mechanism inducing the non-collinear moments. For momenta parallel to the original moment direction, the perpendicular component only depends on the total spin component in each of the x–y planes. Since the non-collinear component is basically a Néel fluctuation in an x–y plane, there is only a small component of total spin in each plane. For the balanced clusters the component vanishes identically, whereas for the unbalanced clusters the component is small and shrinks as the cluster size is increased (see figure 5(*e*)). Only for the two-impurity cluster calculation of figure 5(*f*) is the perpendicular component likely to be non-negligible, but even here it is almost indistinguishable from zero. For this final situation, the Néel fluctuation is balanced by components in two planes and this is effectively a minor component of the original spin density wave but in the non-collinear component. The magnetic diffuse scattering for momentum transfer parallel to the original spin direction is calculated for the relevant clusters in figure 5. The normalisation used is such that one replaced spin would yield unit scattering intensity and so this 'parallel' scattering is clearly minor. It is unlikely that this measurement would be experimentally viable.

The non-collinear components correspond to local multiple-$\boldsymbol{Q}$ fluctuations. This suggests a second possible way to look for non-collinear distortions. Any non-collinear magnetic diffuse scattering from our mechanism is strongly peaked around the magnetic Bragg spots corresponding to the spin density waves perpendicular to the one present. Unfortunately, scattering around these Bragg spots does not prove the existence of a non-collinear component. Although in our model such scattering would constitute proof, if the *lengths* of the spins were allowed to vary, then simply extending the lengths of the nearest-neighbour spins on the *same* magnetic sublattice as the impurity would induce precisely the same scattering [12]. It is difficult to understand why these spins might want to extend, whereas for our model, simple Heisenberg interactions induce the desired effects. Nonetheless, scattering peaked at 'would be' magnetic Bragg spots does not constitute unambiguous proof of a non-collinear component, although we would consider it indicative.

The magnetic diffuse scattering from the sum of both the parallel and perpendicular components is plotted through magnetic Bragg spots and 'would be' magnetic Bragg spots in figure 5, in the combinations that would be experimentally observed. Let us

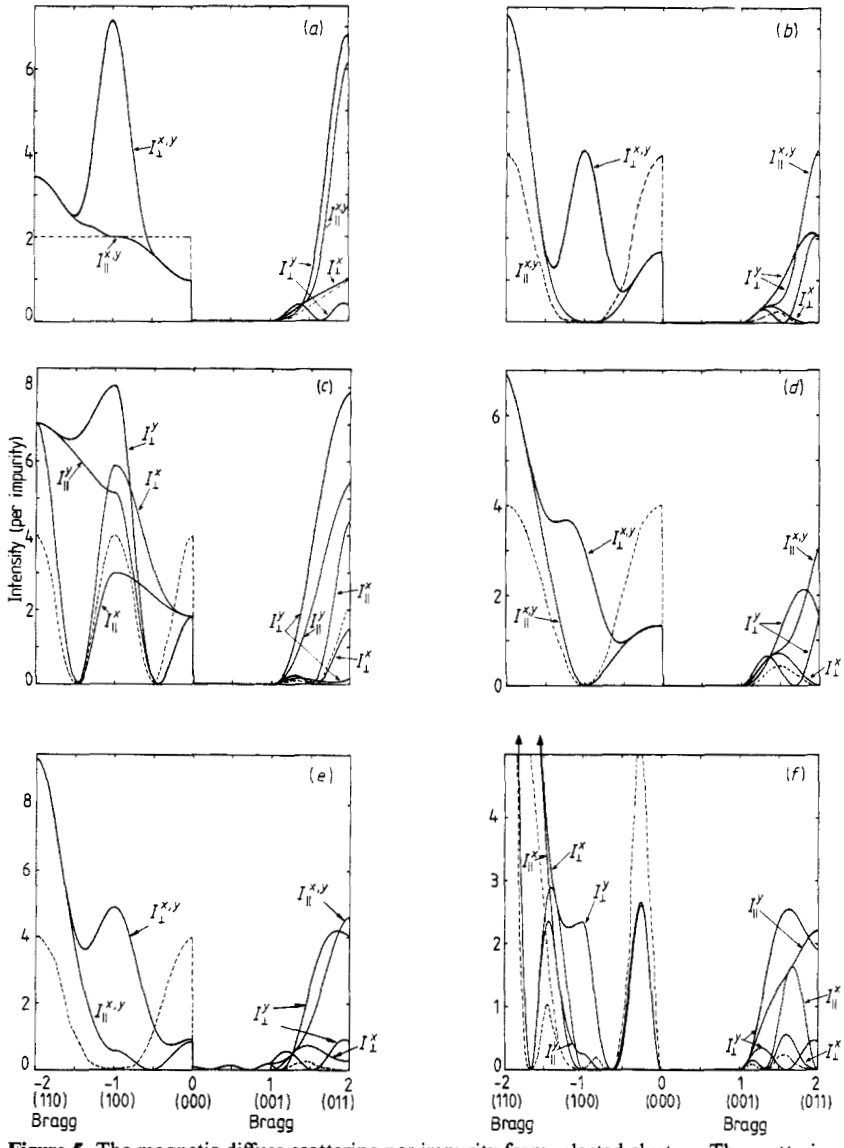

Figure 5. The magnetic diffuse scattering per impurity from selected clusters. The scattering from the bare impurities is depicted by faint broken curves for reference and the clusters are oriented in both directions and with both relative orientations between the spin and lattice. Details are supplied in the text. The bare impurity scattering defines the shape, the collinear component is peaked at the magnetic Bragg spots and the non-collinear component is peaked at the 'would be' magnetic Bragg spots.

focus on figure 5(*a*). The first curve to study is the broken curve. This curve corresponds to the scattering from the collinear cluster with just the impurity spins removed and all the remaining spins in their original positions. This scattering, with the angular factor removed, should be compared to the nuclear diffuse scattering and measures the short-range correlations between the impurity atoms themselves. The important contribution to us is the excess magnetic scattering over and above this contribution. For our more exotic impurity configurations this non-magnetic contribution has a

lot of irrelevant structure which will disappear when many impurities with different configurations are averaged over.

When we allow the surrounding spins to reorient, there is an immediate complication due to the local symmetry breaking. If we assume that the neutron sees a 'frozen' non-collinear distortion, i.e. the quantum fluctuations are on a very long time scale, then the direction of the non-collinear component breaks the spin symmetry in the x–y plane. If we transfer momentum parallel to the x axis, then we assume that the non-collinear component is either parallel or perpendicular to the momentum transfer and denote such calculations $I_{\parallel}$ and $I_{\perp}$ respectively. It is important to realise that $I_{\parallel}$ and $I_{\perp}$ are manifestations of the *same* non-collinear impurity, and reflecting our path in $x = y$ will convert one type of scattering into the other. The experimental intensity might be expected to be an average over the two, corresponding to many uncorrelated clusters.

There is a second complication. As well as the breaking of the local spin symmetry, there is an independent breaking of the spatial symmetry. If we study figure 3(a), then the spins marked $\boldsymbol{T}'$ rotate in the opposite direction to $\boldsymbol{T}$ which breaks x–y symmetry. The non-collinear component can then orient either parallel to the line joining the two $\boldsymbol{T}$s or parallel to the line joining the two $\boldsymbol{T}'$s. These two types of choice correspond to two different types of spin–lattice coupling and are denoted by Cartesian superscripts, x and y. The choice between I^x and I^y intensity is physical and the experimental systems will be expected to have a predominance of one type of scattering. The configuration which has the smallest relative component of (100) when compared with (011), is likely to be the experimental configuration, stabilised by a spin–orbit coupling contribution which has been omitted from the calculations.

We are left with a final complication which is the relative orientation of the non-collinear component to the collinear component. In the presence of the non-collinear component, spins rotate away from the z axis and take up positions at particular fixed inclinations. If the momentum transfer happens to become parallel to a particular spin, then the neutron ceases to scatter from it. This fact then leads to a breaking of the symmetry for scattering in the *plane* of the spins. For a fixed K_z, we find different scattering for positive and negative values of K_x. This final complication then explains why there are sometimes two curves with the same labels, corresponding to orienting the non-collinear component of the spins in the cluster in opposite directions and changing the relative sign of $\delta S_{\perp}$ and $\delta S_{\parallel}$ in (4.3).

For all examples the observed scattering will be a form of average over the presented curves. The path traversed in reciprocal space passes across the most likely regions for experimental investigation. We assume that the collinear spin density wave is parallel to the z axis. On the left-hand side of the figure, we start with the brightest magnetic Bragg spot; the (110) spot. We then move to the 'would be' magnetic Bragg spot corresponding to a spin density wave parallel to the x axis; the (100) spot. We then move to the main beam and then out to the magnetic Bragg spot which vanishes because the momentum transfer is parallel to the spin; the (001) spot. Finally we move to the brightest 'would be' magnetic Bragg spot where we might expect the non-collinear scattering to be the strongest; the (011) spot. All contributions are strongly peaked at both magnetic Bragg spots and 'would be' magnetic Bragg spots and the source of this scattering is easy to understand.

The scattering induced at the existing magnetic Bragg spots comes predominantly from the *loss* of the existing moments, which are turned into the non-collinear components. In the collinear ground state, all moments are optimally positioned and therefore

any change in the collinear component must have the same symmetry as the original spin density wave. This is observed in *all* our calculations and may be deduced by noting that the peak at (110) is huge but that the peak at (001) is invariably absent, due to the orientational factor $(1-\hat{K}_z^2)$ which eliminates only the collinear component.

The scattering at the 'would be' magnetic Bragg spots is dominantly non-collinear. This is the straightforward interpretation of the non-collinear components as 'bound states' of multiple-$\boldsymbol{Q}$ behaviour. The peaks are observed at both (011) and (100) 'would be' magnetic Bragg spots, although in order to save spin–orbit coupling we might have expected the second contribution to vanish. The Néel fluctuations in the x–y layers are a superposition of the two orthogonal spin density waves, but are orientated in a unique direction. One of the two components is orthogonal to where the spin–orbit coupling would align it and so yields scattering at (100). For the coupled pair of impurities depicted in figure 5(*f*), one of the two spin density waves dominates and so the (100) scattering is severely curtailed, although there is still a large peak at (011). This leads to a third possible way to find a non-collinear component and to the first viable experimental possibility.

Magnetic diffuse scattering makes quite a difficult experiment [21] and so the observation of a non-collinear component should be attempted with the dominant scattering contribution if possible. The dominant non-collinear scattering in our model arises at the 'would be' magnetic Bragg spots. Probably the best way to show the existence of a non-collinear component is to compare the magnetic diffuse scattering around the (100) and (011) positions. In terms of the magnetic symmetry group of the original spin density wave, these two positions are equivalent. One of the two positions is further from the origin and so there should be a minor reduction in intensity from the form factor, but the major effect should be the orientational factor.

If we assume that there is no residual 'coupling' between the different components, then the intensity averaged over many impurities would be

$$I(\boldsymbol{K}) = |\delta S_\perp^x|^2(1-\hat{K}_x^2) + |\delta S_\perp^y|^2(1-\hat{K}_y^2) + |\delta S_\parallel|^2(1-\hat{K}_z^2) \tag{4.4}$$

and then the ratio of the intensities at (011) and (100) is

$$R = \frac{I(011)}{I(100)} = \frac{1}{2} + \frac{|\delta S_\perp^1|^2}{|\delta S_\perp^2|^2 + |\delta S_\parallel|^2}(100). \tag{4.5}$$

In the absence of a non-collinear component this ratio would be $\frac{1}{2}$, and the presence of a non-collinear component would increase it. The non-collinear scattering is strongly peaked at these points, whereas the collinear component is not peaked, although the collinear component is expected to be intrinsically larger. The spin–orbit coupling pushes the peak into $|\delta S_\perp^1|^2$ rather than $|\delta S_\perp^2|^2$. This is due just to the fact that Néel fluctuations involve two spin density waves and the major contribution goes into $|\delta S_\perp^1|^2$ whereas the minor contribution goes into $|\delta S_\perp^2|^2$. The parallel component involves the impurity spins themselves, together with the loss of spin into the non-collinear component, whereas the non-collinear component is only relevant for the particular combinations of impurities which *bind* a non-collinear component, suggesting that collinear scattering dominates.

For the cases we have considered, the values of R are 1.063, 1.500, 0.719, 1.500, 1.289 and 1.306 for (*a*) through to (*f*) respectively and we believe that extensions into the real systems should be measurable.

We have allowed only a small fraction of the spins to rotate. We enforced this in our calculations in order to stabilise the collinear phase, but we must point out the consequences of this assumption. The diffuse contribution from the impurities themselves is probably overemphasised in comparison to both the non-collinear component and the collinear spin component. The minor reorientation of the more distant spins has been omitted, and although the more distant spins only move a little, there are many of them which would strongly enhance the non-collinear component. For cluster (*f*) the situation is even worse, because some of the frozen spins should form an (*e*)-like configuration which has been neglected. The shape of the non-collinear scattering is the relevant quantity to be extracted from our calculations, and this is clearly peaked at the 'would be' Bragg spots.

5. Conclusions

We have found a new physical mechanism for the stabilisation of static short-range non-collinear components in antiferromagnets with long-range collinear order. The mechanism only applies to non-bipartite systems and relies heavily upon topological frustration to operate. We have found two ways to interpret the effect. Firstly, in frustrated collinear antiferromagnets, each spin has both parallel and antiparallel neighbours with the antiparallel neighbours dominating. If the local dominance is cancelled out because impurities replace some of the antiparallel neighbours, then the relevant spin can gain nothing from the collinear system and so it forms a local non-collinear 'bound state', gaining energy from antiferromagnetically polarising its neighbours. This argument can be applied to *all* frustrated systems. Secondly, for the face centre cubic lattice, there is a two-dimensional Goldstone mode which corresponds to a non-collinear Néel fluctuation in one x–y plane. Even a single impurity acts as an attractive potential to this Goldstone mode in a neighbouring layer, and because the problem is essentially two dimensional, the impurity potential induces a non-collinear bound state.

These results rely heavily on a Heisenberg description. Quantum mechanical fluctuations and itineracy both interfere, and result in the stabilisation of the collinear phase. We believe that short-range non-collinear components are likely to survive the inclusion of the neglected phenomena. Our first argument is local in content and likely to remain relevant, although the Goldstone mode is likely to be raised to a higher energy thus becoming less relevant.

A consideration of the interactions between non-collinear impurities, suggests a phase transition into an orthorhombic state as the concentration of impurities is increased and just such a transition is observed in the γ-MnNi system [6]. The likely description of the transition involves percolation of a non-collinear cluster and this inhomogeneous description is unlikely to support a softening spin wave excitation as has previously been suggested [14].

The natural probe to look for non-collinear impurities is magnetic diffuse scattering from a single-domain single crystal. Unfortunately, our mechanism suggests that the natural technique, of looking for scattering from momentum transfer parallel to the long-range spin order, will yield only minor scattering and that the best way to find the non-collinear impurities is to study the angular dependence of the magnetic diffuse scattering near 'would be' magnetic Bragg spots.

We believe that the ideas in this paper are more relevant to γ-MnNi than to γ-MnCu, although the second system seems of more experimental interest [12].

We should mention that magnetoelasticity is huge in these systems and involves a certain amount of 'unfrustrating' distortion. The 'size' effects and local spatial rearrangements around impurities is an important effect for future consideration.

The theory awaits experimental verification or refutation.

Acknowledgments

I would like to thank Dr Oscar Moze for introducing me to the problem and for many useful suggestions.

Appendix A

Topological frustration involves the necessity of ferromagnetically aligned spins even in the classical antiferromagnetic ground state. One measure of the degree of frustration is the fraction of ferromagnetic bonds in the ground state. If P_f and P_a denote the probabilities of the bonds being ferromagnetic and antiferromagnetic respectively, then for any configuration $\{\boldsymbol{S}_i\}$:

$$P_f + P_a = 1 \tag{A1a}$$

$$P_f - P_a = \frac{1}{ZNS^2} \sum_{\langle ii' \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_{i'} \tag{A1b}$$

where Z is the coordination number and $\langle ii' \rangle$ denote nearest neighbours and run over all bonds. In the antiferromagnetic ground state, the spins must be chosen to minimise the ferromagnetic bonds. Even if we cannot solve this problem, for a periodic array of spins, it is easy to obtain a bound for these bond probabilities. The problem is best solved in reciprocal space, where

$$P_f - P_a = \frac{\sum_k |\boldsymbol{S}|_k^2 \gamma_k}{\sum_k |\boldsymbol{S}|_k^2} > \gamma_{\min} \tag{A2}$$

where the inequality comes directly from the theory of eigenvalues (namely γ_k) and eigenvectors (namely $\boldsymbol{S}_k$), in terms of the normalised structure factor:

$$\gamma_k = \frac{1}{ZN} \sum_{\langle ii' \rangle} \exp[i\boldsymbol{k} \cdot (\boldsymbol{R}_i - \boldsymbol{R}_{i'})]. \tag{A3}$$

It is clear that $-1 < \gamma_k < 1$ and that $P_f > (1 + \gamma_{\min})/2$ for the antiferromagnetic ground state. For each lattice, the frustration can be measured by studying γ_k. For the face centre cubic lattice, $\gamma_{\min} = -\frac{1}{3}$ and so $P_f = \frac{1}{3}$.

Appendix B

The classical limit of the Heisenberg model involves pointing each spin parallel to the local field defined by the sum of its nearest neighbours.

For 'unbalanced' systems with three distinct types of non-collinear spins, this criterion becomes:

$$\begin{bmatrix} \lambda_T & A & B \\ A & \lambda_U & C \\ B & C & \lambda_V \end{bmatrix} \begin{bmatrix} \boldsymbol{T} \\ \boldsymbol{U} \\ \boldsymbol{V} \end{bmatrix} = \begin{bmatrix} \alpha(\kappa) \\ \beta(\kappa) \\ \gamma(\kappa) \end{bmatrix} \boldsymbol{S} \tag{B1}$$

for couplings A, B, C and α, β, γ which depend on the cluster type and Lagrange multipliers λ_X which are chosen to normalise the spins $\boldsymbol{X}$.

On the assumption that there is a unique orthogonal component, we may set

$$\boldsymbol{X} = \cos x \, \boldsymbol{S} + \sin x \, \bar{\boldsymbol{S}} \tag{B2}$$

where $\bar{\boldsymbol{S}}$ is perpendicular to the quantisation direction $\boldsymbol{S}$. The equations then reduce to

$$\begin{aligned} &\alpha(\kappa) \sin t + A \sin(u - t) + B \sin(v - t) = 0 \\ &\beta(\kappa) \sin u + A \sin(t - u) + C \sin(v - u) = 0 \\ &\gamma(\kappa) \sin v + B \sin(t - v) + C \sin(u - v) = 0 \end{aligned} \tag{B3}$$

which are easily solved.

Note that the matrix in (B1) is *singular* with a zero eigenvalue and with an eigenvector corresponding to the bound state.

For 'balanced' systems there is a complication because only the non-collinear component is 'balanced'. The coupling between $\boldsymbol{X}$ and $\boldsymbol{X}'$ has opposite signs for the two components and we find

$$\begin{bmatrix} \lambda_T + c & A & B \\ A & \lambda_U + b & C \\ B & C & \lambda_V + a \end{bmatrix} \begin{bmatrix} \cos t \\ \cos u \\ \cos v \end{bmatrix} = \begin{bmatrix} \alpha(\kappa) \\ \beta(\kappa) \\ \gamma(\kappa) \end{bmatrix} \tag{B4}$$

$$\begin{bmatrix} \lambda_T - c & A & B \\ A & \lambda_U - b & C \\ B & C & \lambda_V - a \end{bmatrix} \begin{bmatrix} \sin t \\ \sin u \\ \sin v \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \\ 0 \end{bmatrix} \tag{B5}$$

which reduce to

$$\begin{aligned} &\alpha(\kappa) \sin t + A \sin(u - t) + B \sin(v - t) + c \sin 2t = 0 \\ &\beta(\kappa) \sin u + A \sin(t - u) + C \sin(v - u) + b \sin 2u = 0 \\ &\gamma(\kappa) \sin v + B \sin(t - v) + C \sin(u - v) + a \sin 2v = 0 \end{aligned} \tag{B6}$$

which are again easily solved.

References

[1] Cowlam N, Bacon G E and Gillott L 1977 *J. Phys. F: Met. Phys.* **7** L315

[2] Hennion B, Hutchings M T, Lowde R D, Stringfellow M W and Tochetti D 1976 *Proc. 2nd Int. Conf. on Neutron Scattering (Gatlinberg, 1975) (Publication CONS-760601-P2)* ed R M Moon (New York: US Department of Commerce) p 825

[3] Mikke K, Jankowski J and Jaworska E 1983 *Physica* B+C **120** 156

[4] Endoh Y and Ishikawa Y 1971 *J. Phys. Soc. Japan* **30** 1614
[5] Yamaoka T, Mekata Y and Takaki H 1974 *J. Phys. Soc. Japan* **36** 438
Yamaoka T 1974 *J. Phys. Soc. Japan* **36** 445
[6] Honda N, Tanji Y and Nakagawa Y 1976 *J. Phys. Soc. Japan* **41** 1931
[7] Coles B R 1977 *Physica* B+C **91** 167
Makhurane P and Gaunt P 1969 *J. Phys. C: Solid State Phys.* **2** 959
[8] Umebayashi H and Ishikawa Y 1966 *J. Phys. Soc. Japan* **21** 1281
Jo T and Hirai K 1986 *J. Phys. Soc. Japan* **55** 2017
Long M W and Yeung W 1987 *J. Phys. C: Solid State Phys.* **20** 5839
[9] Long M W and Steigenberger U 1989 *Phys. Scr.* T **25** 220
[10] Rossat-Mignod J, Burlet P, Quezel S and Vogt O 1980 *Physica* B **102** 237
Kouvel J S and Kasper J S 1963 *J. Phys. Chem. Solids* **24** 529
[11] Jo T 1983 *J. Phys. F: Met. Phys.* **13** L211
Long M W and Yeung W 1986 *J. Phys. F: Met. Phys.* **16** 769
[12] Moze O and Hicks T J 1989 private communication
[13] There seems to be no good experimental review for disordered ferromagnets.
[14] Long M W and Yeung W 1987 *J. Phys. F: Met. Phys.* **17** 1175
[15] Moze O 1989 private communication
[16] Gaulin B D, Spooner S and Morii Y 1987 *Phys. Rev. Lett.* **59** 668
[17] Lowde R D 1986 private communication, and [18]
[18] Moze O and Hicks T J 1982 *J. Phys. F: Met. Phys.* **12** 1
[19] Long M W 1989 *J. Phys.: Condens. Matter* **1** 2857
Henley C L 1987 *J. Appl. Phys.* **61** 3962
[20] Lovesey S W 1984 *Theory of Neutron Scattering from Condensed Matter* vol 2 (Oxford: Clarendon)
[21] Rainford B D 1982 *J. Physique Coll. Suppl.* 12 **43** C7