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LETTER TO THE EDITOR

A new magnetic structure for Mn_3Pt

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Abstract. We suggest a new magnetic structure for the high-temperature antiferromagnet found in the Mn_3Pt system. Our structure may be interpreted as a 'multiple-q' state and is unique up to a discrete set of analogous states. The exotic features of the previous description; atoms with vanishing moments, unobserved tetragonal symmetry and reduction in spin dimensionality as the temperature is increased, are all absent from our proposed state. Our state is consistent with published scattering experiments.

The Mn_3Pt alloy has been much studied because of its first-order phase transition between two ordered antiferromagnetic phases [1]. The alloy crystallises in the Cu_3Au structure with moments of about $3\mu_B$ sitting on the Mn atoms. The Pt atoms have a much smaller moment that we will ignore. At low temperatures the moments order in the triangular lattice ground state depicted in figure 1. Each moment is at a relative angle of 120° to each of its nearest neighbours and all the spins are coplanar. At intermediate temperatures there is a second antiferromagnetic phase characterized by a doubling of the unit cell. The previous works on this material have all assigned the quite exotic collinear state depicted in figure 2 to this phase [2].

There are several surprising features of this collinear state: firstly, there are Mn atoms with no ordered moment. It is supposed that the moments on these sites fluctuate in direction, and furthermore it has been proposed that the associated entropy might be used as an explanation for the possibility of this phase becoming stable [3]. Secondly, it is clear that the crystal symmetry of the phase is tetragonal. The energy scale of the exchange in this type of system ensures that there would be a corresponding structural distortion which ought to be experimentally measurable. Indeed, the analogous distortion in type I γ-Mn antiferromagnets is of the order of a few percent [4], which is easily measurable. Such a structural distortion is conspicuous by its absence in the experimental data. Thirdly, the previously proposed phase transition, as the temperature is raised, involves a *reduction* in the number of spin dimensions required to describe the order, decreasing from two dimensions to one dimension. This is opposite behaviour to that predicted quite generally from considerations of disorder and thermal fluctuations [5]. The magnetic structure that we are proposing does not suffer from any of these three 'deficiencies'.

We believe that the physically realized magnetic structure may be essentially uniquely defined by a collection of antiferromagnetic symmetries; simultaneous spatial translations and spin reflections. These symmetry operations involve translations through the three primitive-unit-cell basis vectors (the three vectors having the magnitudes of the

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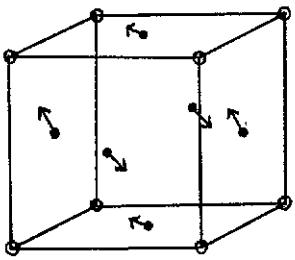

Figure 1. A representation of the triangular lattice antiferromagnetic ground state to the Mn_3Pt alloy. All the moments lie in the plane for which the Mn atom (full circle) is a face centre and point directly at a nearest neighbour Pt atom (open circle).

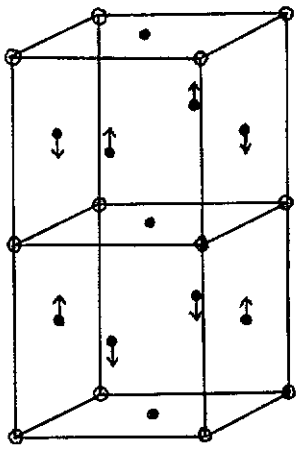

Figure 2. The magnetic state previously assigned to the antiferromagnetic state of Mn_3Pt found at intermediate temperature. The Mn atoms without moments may be thought of as having fluctuating direction.

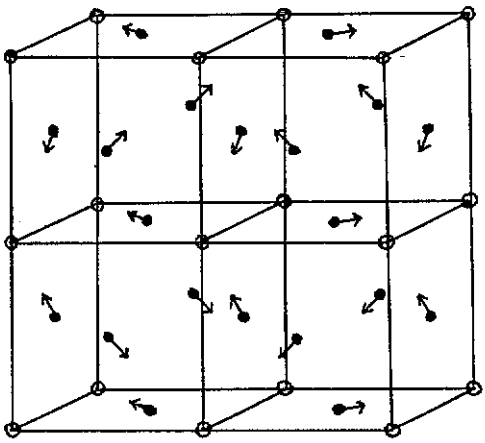

Figure 3. A representation of four unit cells of the magnetic state that we are proposing for the antiferromagnetic phase of Mn_3Pt at intermediate temperatures. There are twelve distinct orientations.

lattice spacing in the three Cartesian directions, ae_i) with *simultaneous* reflection of all the moments in the plane orthogonal to the relevant primitive translation vector. The simplest way to envisage our proposed state is to construct it from the unit cell depicted in figure 1, using these antiferromagnetic symmetries. All the moments may be found using a sequence of combined spatial translations and spin reflections. All nearest neighbour spins *remain* at a relative angle of 120°, but the magnetic structure becomes truely three dimensional. Four unit cells of the resulting magnetic structure are depicted in figure 3.

There are other possible magnetic structures that can be constructed using these symmetry operations; there are three independent cartesian directions and consequently three independent antiferromagnetic symmetries. We can construct states where either

one, two or three of the original pure translational symmetries is replaced by an antiferromagnetic symmetry. Obviously, the only case that retains cubic symmetry is the case where all three translations involve the spin reflections. A second consideration is that although all nearest neighbour spins are at a relative angle of 120° for all these magnetic structures, the next-nearest neighbour orientations vary wildly. Only for the two extremal cases, where all three translations are either pure translations or have the additional spin reflections, are the next-nearest neighbour orientations optimized. For the pure translation case all the next-nearest neighbours are parallel, and this is just the low temperature phase, while for the fully spin reflected phase, two next-nearest neighbours are parallel while the other four are orthogonal. The other two constructions yield inequivalent Mn atoms with intermediate numbers of parallel and orthogonal next-nearest neighbours.

The three 'deficiencies' previously referred to are not applicable to this new spin state with three independent antiferromagnetic symmetries. Obviously, all the Mn atoms have equal magnitudes for their moments. As the temperature is increased, the spin dimensionality would increase from two dimensions to three dimensions. This is quite generally to be expected on elementary grounds [5]. Put simply, moment orientation excitations are energetically less expensive in high dimensional spin states, because surrounding spins have more possible directions into which to relax. This argument applies equally well to static disorder or thermal fluctuations. The phase transition can now be viewed as being assisted by the increase in temperature. Finally, the underlying magnetic symmetry is pure cubic. There is no necessity for a structural distortion to accompany the magnetic phase transition.

The elementary comparison of the low-temperature triangular lattice antiferromagnet with our proposed intermediate temperature antiferromagnet shows that ferromagnetic next-nearest neighbour exchange favours the low-temperature phase while antiferromagnetic next-nearest neighbour exchange favours the intermediate temperature phase. This is in complete accord with previous explanations for the phase transition [1].

We close with a brief indication of a second method of understanding our proposed magnetic state as a 'multiple-q' state. The state depicted in figure 2 may be formally described by the Fourier component pair:

$$S_k = (S/2)(\mp i)e_3\delta_{k,\pm Q_3} \tag{1}$$

where $Q_3 = (\pi/a)(e_3 + 2e_1)$. This leads to:

$$S_i = Se_3 \sin(Q_3 \cdot R_i) \tag{2}$$

in real space. For alternate x–y planes the moments vanish, while for the x–y planes composed wholly of Mn atoms we find the desired Néel correlations. This component is conserved by the three combined translations and moment reflections and therefore has the symmetry that we believe *defines* the magnetic state. The difficulty with this spin component is that it is *not*, as previously suggested, a turning point of the relevant Heisenberg Hamiltonian. In fact, only the 'multiple-q' combination of:

$$S_i = (S/\sqrt{2})[e_1 \sin(Q_1 \cdot R_i) + e_2 \sin(Q_2 \cdot R_i) + e_3 \sin(Q_3 \cdot R_i)] \tag{3}$$

where $Q_1 = (\pi/a)[e_1 + 2e_2]$, $Q_2 = (\pi/a)[e_2 + 2e_3]$ and $Q_3 = (\pi/a)[e_3 + 2e_1]$, satisfy the relevant constraints. This is precisely the magnetic structure that we are proposing, which is depicted in figure 3.

One important consideration is that, as almost always happens with 'multiple-q' spin states, scattering experiments cannot easily distinguish between a 'multiple-q' state and a sample containing three equally populated domains of a 'single-q' state. Obviously, our proposed state has the magnetic Bragg spots in the correct places.

In conclusion, we believe that Mn_3Pt exhibits a cubic-to-cubic phase transition between the phases depicted in figures 1 and 3. Unlike more elementary 'multiple-q' structures, for Cu_3Au systems only an *equal* superposition of the three spin density waves satisfies the relevant constraints. The magnetic structure we propose has no non-trivial continuous degeneracy and is unique up to a few *discrete* permutations of the magnetic sublattices. The so-called collinear phase is *not* the classical solution to the problem as has previously been suggested.

This magnetic structure now awaits experimental confirmation.

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