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Magnetic Diffuse Scattering in the Frustrated Square Lattice

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Abstract

Some of the first transition metal alloys show first order antiferromagnetic phase transition as a function of alloy concentration [1-5]. The antiferromagnetism of materials can be classified into three different class: type I, type II and type III. Although the neutron diffraction studies give information about the type of magnetism, they do not show the fluctuations of magnetic moment from the original direction. So in order to have some information about this fluctuations one must perform the magnetic diffuse scattering. Recently, we proposed that the square lattice may have a double-Q structure as well as single-Q [6]. We showed that the alloying may cause a first order antiferromagnetic phase transition. In order to show that this is the case we perform the magnetic diffuse scattering and give more evidences that the alloying play the dominant role in this phase transition.

1. Introduction

First row transition metal alloys, especially γ-Mn, shows very interesting behavior. Manganese quenched into a face center cubic structure exhibits type-I antiferromagnetism. This is a sequence or ferromagnetic x-y layers that alternate in spin direction along the z-axis. The z-axis becomes inequivalent to the other cartesian directions and magnetism induces a huge tetragonal distortion of approximately six per cent parallel to the z-axis [7]. Neutron scattering [8] shows that the spins align parallel to the z-axis where they are held in place by spin-orbit coupling. As we have already mentioned, the structure of γ-Mn is face center cubic. Antiferromagnetism in f.c.c lattice is frustrated, with only a fraction of nearest neighbors being allowed to be antiparallel. Geometrically frustrated lattices provide some of the most sophisticated and interesting types of antiferromagnet. The fundamental cause is that the frustration forces some of the bands to gain less than

their optimum energy. There are usually a variety of ways in which this loss can be spread amongst the different bonds, often leading to ground state degeneracy to leading order. This degeneracy is usually lifted on a smaller energy scale than that promoting the magnetism, and this then leads to the possibility of phase transitions between different magnetic ground states caused by fairly small changes in the magnetic interactions. In particular, alloying a frustrated antiferromagnet can lead to such a phase transition at quite modest doping. The alloying is main focus in our model. When another transition metal is doped into the manganese there are quite dramatic changes in behavior. Fe [1], Ir [9], Ni [2] and Cu [10] all substantially reduce the tetragonal distortion and for Fe, Ir and Ni there is evidence of a cubic phase which is stabilized at doping concentrations of approximately a quarter.

In order to show that the alloying causes this phase transition we will use the Heisenberg model, which is quite appropriate when the magnetic moments are large enough.

2. Heisenberg Model

In our previous studies, we showed that the square lattice can be frustrated by longer-range bonds [6]. We used the classical limit of the Heisenberg model, which says that if the magnetic electrons were described by localized orbitals, then the resultant spins on the neighboring atoms would align either parallel or antiparallel depending on the sign of the exchange integral J.

We proposed that the square lattice has frustrated antiferromagnetic arrangement (double-Q) depending on the strength of the higher nearest neighbors' interactions, although it is commonly known as an unfrustrated system (single-Q).

Heisenberg Hamiltonian is

$$H = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S_i} \cdot \mathbf{S_j}, \tag{1}$$

where S_i and S_j are the resultant spins of neighboring atoms, which are quantum mechanical operators satisfying the commutation relations $(h/2\pi = 1)$

$$\left[\mathbf{S_i^\alpha}, \mathbf{S_j^\beta}\right] = \sum_\gamma i\varepsilon_{\alpha\beta\gamma}\delta_{ij}\mathbf{S_i^\gamma} \tag{2}$$

According to Hund' s third rule the spin is usually maximum [11] so there is a constraint on spin. The total spin constraint is

$$\mathbf{S_i} \cdot \mathbf{S_j} = S(S+1) \tag{3}$$

and applies to each atom independently. For the ground state, it must be minimized subject to constraint. This yields a very complicated quantum mechanical problem [12].

However, the spins may be considered as very large spins i.e, $S \to \infty$. In this limit the quantum mechanics goes away. If we normalize the spins by

$$\hat{\mathbf{S}}_{\mathbf{i}} = S^{-1}\mathbf{S}_{\mathbf{i}}. \tag{4}$$

Then

$$\left[\hat{\mathbf{S}}_{\mathbf{j}}^{\alpha}, \hat{\mathbf{S}}_{\mathbf{j}}^{\beta}\right] = \frac{1}{S}\sum_{j} i\varepsilon_{\alpha\beta\gamma}\delta_{ij}\hat{\mathbf{S}}_{\mathbf{i}}^{\gamma} = 0. \tag{5}$$

The remainder of the problem is simply to find relative directions in which each of the spins point in order that the Hamiltonian

$$H = JS^2 \sum_{ij} \hat{\mathbf{S}}_{\mathbf{i}} \cdot \hat{\mathbf{S}}_{\mathbf{j}} \tag{6}$$

is minimized. Including the second nearest neighbors interactions the Hamiltonian becomes

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_{\mathbf{i}} \cdot \mathbf{S}_{\mathbf{j}} + J_2 \sum_{[i,j]} \mathbf{S}_{\mathbf{i}} \cdot \mathbf{S}_{\mathbf{j}}. \tag{7}$$

Once this Hamiltonian is solved [6], we can get two different magnetic ordering known as collinear and non-collinear as shown in Figure 1 and Figure 2. We proposed that the alloying may cause an antiferromagnetic first order phase transition. We found some indications that this is really happening. In order to give more evidences that this is the case one must perform magnetic diffuse scattering.

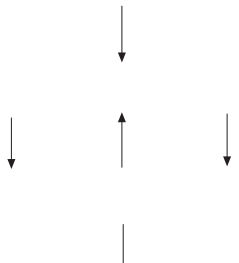

Figure 1. Collinear phase

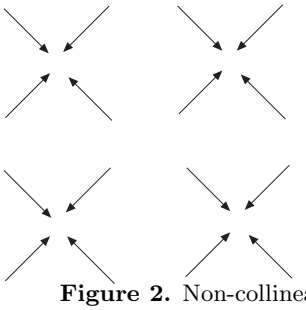

Figure 2. Non-collinear phase

3. Magnetic Diffuse Scattering

If there is any impurity in the system, then we can express the spins as

$$\mathbf{S_R} = \mathbf{S_R^0} + \delta\mathbf{S_R}, \tag{8}$$

where δS_R is spin fluctuations. Then one can calculate the Fourier transform of the spin as

$$\mathbf{S_k} = \sum_i (\mathbf{S_i^0} + \delta\mathbf{S_{Ri}}) \exp(i\mathbf{k}\cdot\mathbf{R_i}). \tag{9}$$

Thus the magnetic scattering intensity

$$I(\mathbf{k}) \propto \left|\hat{\kappa}\wedge(\mathbf{S_k}\wedge\hat{\kappa})\right|^2 \tag{10}$$

can be written as

$$I(\mathbf{k}) \propto \left|\hat{\kappa}\wedge\left[(\mathbf{S_k^0}+\delta\mathbf{S_k})\wedge\hat{\kappa}\right]\right|, \tag{11}$$

which is

$$I(\mathbf{k})_m + I(\mathbf{k})_{\text{diff}} \tag{12}$$

Here, we have omitted terms like $\mathbf{S}_\perp\mathbf{S}\mathbf{S}_\perp + \mathbf{S}\mathbf{S}_\perp\mathbf{S}_\perp$, since the contribution from this term is small. This leads to a sum of two scatterings. If we choose the propagating to be parallel to the spin direction, then the magnetic scattering intensity disappears. The only remaining thing is now diffuse, which is

$$I(\mathbf{k}) \propto \left|\hat{\kappa}\wedge(\delta\mathbf{S_k}\wedge\hat{\kappa})\right|^2. \tag{13}$$

This is the quantity that gives information about the fluctuations. We will use this approximation in order to investigate any non-collinear spin arrangement in the square lattice. Equation (13) can be rewritten as $\left|\delta\mathbf{S_k} - \left[\hat{\kappa}\delta\mathbf{S_k}\right]\hat{\kappa}\right|^2$. Compering with $\left|\delta\mathbf{S_k}\right|^2$, the other terms can be ignored. This means we can work with only $\left|\delta\mathbf{S_k}\right|^2$.

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

$$I(\mathbf{k}) = \left|\delta\mathbf{S}_\parallel\right|^2 + \left|\delta\mathbf{S}_\perp\right|^2 - \left|\delta\mathbf{S}_\parallel\hat{\kappa}_\mathbf{z} + \delta\mathbf{S}_\perp\hat{\kappa}_\mathbf{x}\right|^2 \tag{14a}$$

$$I(\mathbf{k}) = \left|\delta\mathbf{S}_\parallel\hat{\kappa}_\mathbf{x} - \delta\mathbf{S}_\perp\hat{\kappa}_\mathbf{z}\right|^2 + \hat{\kappa}_\mathbf{y}^\mathbf{2}\left(\left|\delta\mathbf{S}_\parallel\right|^2 + \left|\delta\mathbf{S}_\perp\right|^2\right) \tag{14b}$$

in terms of parallel $(\delta \mathbf{S}_{\parallel})$, and perpendicular $(\delta \mathbf{S}_{\perp})$, spin components.

Firstly, we will consider the collinear arrangement on the square lattice. Using equation (13) one can work out magnetic Bragg spots. We show the Bragg spots of three phases in the square lattice in Figure 3a to Figure 3c.

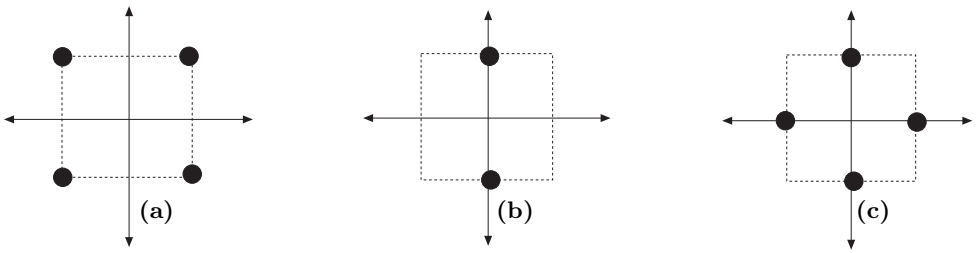

Figure 3. Three phases of square lattice

Figure 3a is the Bragg spots of the simple collinear arrangement. Figure 3b is the Bragg spots of the degenerate improved [12] collinear arrangement and Figure 3c shows the Bragg spots of the non-collinear arrangement in the first Brillioun zone.

The question is now whether or not the impurities cause a non-collinear arrangement in the collinear phase? In order to learn this, we will work with diffuse scattering.

4. Magnetic Diffuse Scattering In Chosen Clusters

Firstly, we will consider a small cluster with an impurity. Before the impurity addition in the cluster, the spin configuration is shown in Figure 4a. In the presence of the impurity we can represent the spin as

$$\mathbf{S_l} = \mathbf{S_l^0} + \delta \mathbf{S_{Rl}} \tag{15}$$

in which $\delta \mathbf{S_{Rl}}$ is the additional spin term due to impurity that forces the spin to have two components. This means the spins must rotate from the original direction with an angle θ which is pictured in Figure 4b.

We need δS_k for calculating the diffuse magnetic scattering. This is

$$\delta \mathbf{S_k} = \frac{1}{N} \sum_l \exp(i\mathbf{k}.\mathbf{R_i}) \mathbf{S_{Rl}} \tag{16}$$

In this cluster we assume that only four spins would be affected after an impurity. So $\delta \mathbf{S_R}$ are

$$\delta\mathbf{S}(\mathbf{R_0}) = -\mathbf{S_0} \tag{17a}$$
$$\delta\mathbf{S}(\mathbf{R_1}) = (\mathbf{T_1} - \mathbf{S_1}) \tag{17b}$$
$$\delta\mathbf{S}(\mathbf{R_2}) = (\mathbf{T_2} - \mathbf{S_2}), \tag{17c}$$

where $\mathbf{T_1}$ and $\mathbf{T_2}$ are

$$\mathbf{T_1} = \mathbf{S}\cos\theta + \mathbf{S}^{\imath}\sin\theta \tag{18a}$$
$$\mathbf{T_2} = \mathbf{S}\cos\theta - \mathbf{S}^{\imath}\sin\theta, \tag{18b}$$

and where $\mathbf{S}\cdot\mathbf{S^l} = 0$.

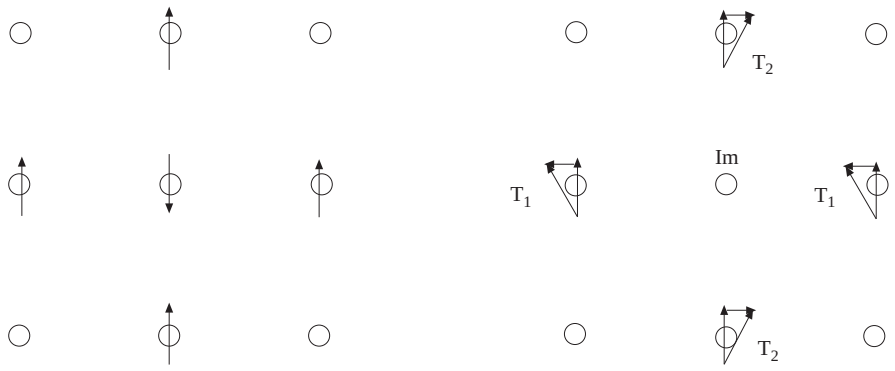

Figure 4. a) Before the impurity addition in a small cluster b) One impurity in a small cluster

Then the fourier transform of $\delta\mathbf{S_R}$ is

$$\delta\mathbf{S_k} = -\mathbf{S_0}(\exp i\mathbf{k}\cdot\mathbf{R_0}) + (\mathbf{T_1} - \mathbf{S_1})(\exp(i\mathbf{k}\cdot\mathbf{R_1}) + \exp(-i\mathbf{k}\cdot\mathbf{R_1}) + (\mathbf{T_2} - \mathbf{S_2})(\exp i\mathbf{k}\cdot\mathbf{R_2} + \exp(-i\mathbf{k}\cdot\mathbf{R_2}) \tag{19}$$
$$\delta\mathbf{S_k} = 2\cos\mathbf{k_X}(\mathbf{T_1} - \mathbf{S_1}(\mathbf{T_1} - \mathbf{S_1}) - \mathbf{S_0} + 2\cos\mathbf{k_Y}(\mathbf{T_2} - \mathbf{S_2}) \tag{20}$$

Using the equations (18.a) and(18.b) we can rewrite this equation as

$$\delta\mathbf{S_k} = \mathbf{S_0}\left[2\cos\theta(\cos\mathbf{k_X} + \cos\mathbf{k_Y}) - 1\right] + \mathbf{S_1}\left[2(1-\sin\theta)(\cos\mathbf{k_Y} - \cos\mathbf{k_X})\right] \tag{21}$$

and its square is

$$\left|\delta\mathbf{S_k}\right|^2 = S^2\left[2\cos\theta(\cos\mathbf{k_X}+\cos\mathbf{k_Y})-1\right]^2 + 4(1-\sin\theta)^2(\cos\mathbf{k_X}-\cos\mathbf{k_Y})^2. \quad (22)$$

Here we have used $\mathbf{S_0}\cdot\mathbf{S_1}=0$. This can be separated into two parts as

$$\left|\delta\mathbf{S}_\perp\right|^2 = 4(1-\sin\theta)^2(\cos\mathbf{k_X}-\cos\mathbf{k_Y})^2 \quad (23a)$$

$$\left|\delta\mathbf{S}_\parallel\right|^2 = \left[2\cos\theta(\cos\mathbf{k_X}+\cos\mathbf{k_Y})-1\right]^2. \quad (23b)$$

The first equation gives the scattering peaks when $\cos\mathbf{k_X} = -\cos\mathbf{k_Y} = 1$ as shown in Figune 5 by X.

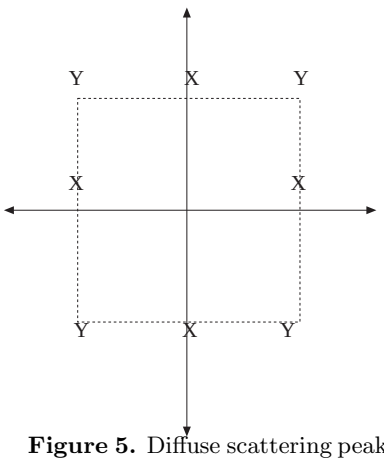

Figure 5. Diffuse scattering peaks of collinear phase after an impurity

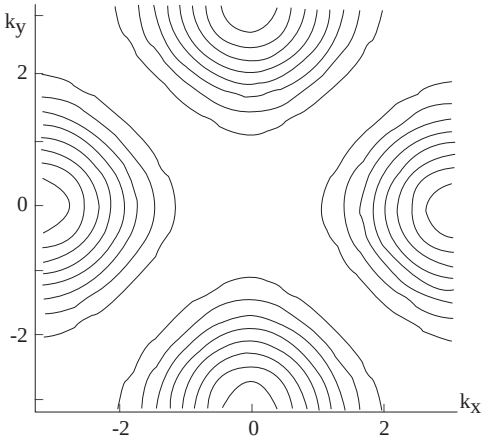

Figure 6. Magnetic diffuse scattering in a small cluster

However the second equation gives scattering peaks when $cos\mathbf{k_X} = cos\mathbf{k_Y}$=-1, which is indicated by Y in Figure 5. It is obvious that the peaks at Y points corresponds to the Bragg spots of the collinear state.

As we mentioned above, in the interval x_c¡z¡1/2 for the simple collinear phase the spins must be represented by two components after the impurity addition. In this interval these X peaks are observable. It is easy to see that the symmetry of the X peaks is the same as the symmetry of non-collinear phase. Thus it may be concluded that the impurity trap a local non-collinear arrangement of spins.

Furthermore, we looked at the variation of $|\delta\mathbf{S}_\perp|$ as a function of the cluster size. We observed that this peak spreads out towards the origin by increasing the size of cluster as shown from Figure 7a to Figure 7d. These results indicate that the scattering is converging towards the Green function calculations of the scattering [4], which says

that there is a long range correlation between the fluctuations. these results are the same as the results observed in MnCu alloys [13]. It means that the prediction of our model is not far away from the reality. If this long range correlation occurs, it is probable that the system picks up another ordering. So the impurities may causes a first order antiferromagnetic phase, transition.

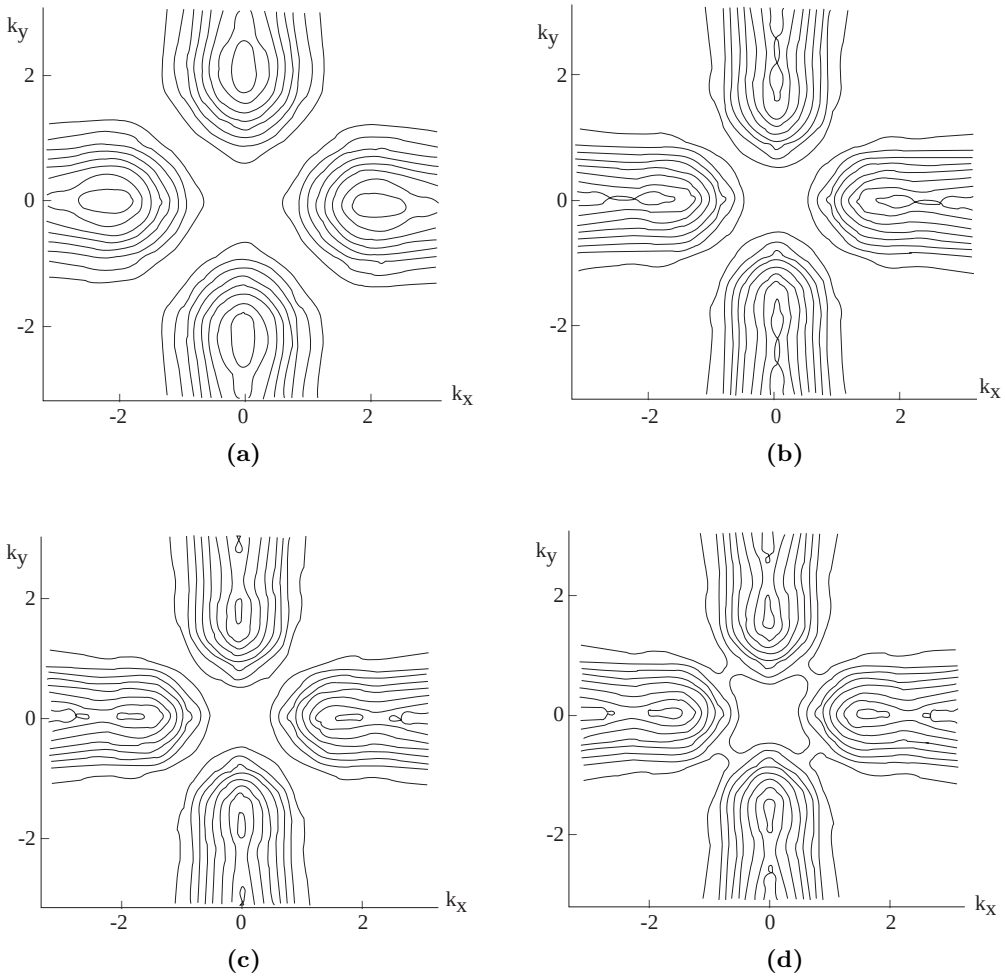

Figure 7. a) Magnetic diffuse scattering:9spins, b) Magnetic diffuse scattering:16spins, c) Magnetic diffuse scattering:36spins, d) Magnetic diffuse scattering:49spins.

5. Conclusions

In this study we investigated the magnetic diffuse scattering after impurity additions. We started with a small cluster of the simple collinear phase containing a single impurity. We saw that the symmetry of diffuse scattering pattern is the same as symmetry of non-collinear phase. By increasing the cluster size we realize that the results converge to the exact results of the Green' s function calculations which says that there may or may not be long rang ordering of the non-collinear phase depending on clustering or anti-clustering [14].

If there is a long range order of the non-collinear phase then one may say that the impurities cause a first order phase transition. We can finally conclude that the alloying cause a first order antiferromagnetic phase transition from one ordering to the another.

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