Theory of multiple spin density wave and lattice distortion in FCC antiferromagnet

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Abstract. The phase diagram including the cubic, two kinds of tetragonal and the orthorhombic phases of the antiferromagnets Mn$_{1-x}$A$_x$ (A = Ni, Ga, Rh and Au) which are known as the first-kind antiferromagnets, is explained theoretically. Our Hamiltonian is composed of the polynomial of the variables describing multiple spin density wave (MSDW) states, the coupling between MSDW states and lattice distortion and the elastic energy. The polynomial is derived from symmetry consideration. By calculating the partition function and the free energy, we show that the phase diagram is reproduced and elucidate the structure of MSDW at each phase and the condition of the appearance of the orthorhombic phase.

1. Introduction
In FCC antiferromagnets, all the nearest neighboring magnetic moments cannot be antiparallel with each other. As a result, antiferromagnetic ordering generally induces lattice distortions. In Mn$_{1-x}$A$_x$ (A = Ni [1], Ga [2], Rh [3] and Au [4]), which shows the first-kind FCC antiferromagnetic structure, the phase diagram including the cubic, two kinds of tetragonal and the orthorhombic phases is reported in the temperature ($T$) and the concentration ($x$) plane; the schematic phase diagram is shown in Fig. 1. Although a phenomenological theory to explain the phase diagram, which takes into account the magnetoelastic coupling, is reported [5], no microscopic theory exists. The purpose of the present work is to propose a microscopic theory of lattice distortion for the purpose of explaining the phase diagram observed in the antiferromagnetic region of some FCC alloys.

Figure 1. The schematic phase diagram of the concentration ($x$) dependence of Mn$_{1-x}$A$_x$ (A = Ni, Ga, Rh and Au) below the Néel temperatures ($T_N$).
In the first-kind antiferromagnet there are three wave vectors to describe the modulation of atomic magnetic moment \( Q_x = (2\pi/a)(1, 0, 0) \), \( Q_y = (2\pi/a)(0, 1, 0) \) and \( Q_z = (2\pi/a)(0, 0, 1) \) with the lattice constant \( a \). A general magnetic structure is described by a superposition of the modulations with \( Q_x \), \( Q_y \) and \( Q_z \), which is called multiple spin density wave (MSDW). In the Single-\( Q \) state described by \( Q_z \), for example, the magnetic moments align parallel in the \( x-y \) plane and antiparallel along the \( z \) axis. If the interaction between the nearest neighboring pair prefers the antiferromagnetic coupling, it is expected that the lattice constant along the \( z \) axis decreases and the lattice constants along the \( x \) and \( y \) axes increase, i.e., the tetragonal distortion with \( c/a < 1 \) in the Single-\( Q \) state. In fact, the first-principles calculation for \( \gamma \)Mn \([6]\) which optimizes the lattice distortion, gives the Single-\( Q \) state with the tetragonal \( (c/a < 1) \) lattice structure.

In the present work we introduce "microscopic" mechanical variables to describe the MSDW states. We assume the Hamiltonian is composed of the polynomial of the variables, the coupling between the variables and the lattice strain and the elastic energy, all of which are derived from symmetry consideration of our system. The Mn alloys of our interest are itinerant electron systems, where the electronic structure is to be taken into account explicitly. The explanation of the phase diagram (see Fig. 1) based on the electronic state seems to be beyond the tractable limit at present. We expect our approach gives some insights into more realistic ones based on the electronic structure.

2. Hamiltonian

In MSDW states where the component of \( Q \) is \( A_i \) \((i = x, y, z)\), the spin at the \( i \)-th lattice point \( \mathbf{R}_i \) with magnitude \( S \) is represented as \( \mathbf{S}_i = S (A_x e^{iQ_x \mathbf{R}_i} + A_y e^{iQ_y \mathbf{R}_i} + A_z e^{iQ_z \mathbf{R}_i} \mathbf{k}) \), where \( A_x^2 + A_y^2 + A_z^2 = 1 \) and \( i, j \) and \( k \) are the unit vectors toward the \( x, y \) and \( z \) axes, respectively.

We define quantities \( X_2 \equiv (A_x^2 - A_y^2)/\sqrt{2} \) and \( X_3 \equiv (2A_x^2 - A_x^2 - A_y^2)/\sqrt{6} \) to describe the MSDW states in FCC Mn alloys, and define symmetry strains \( \varepsilon_2 \equiv (e_{xx} - e_{yy})/\sqrt{2} \) and \( \varepsilon_3 \equiv (2e_{zz} - e_{xx} - e_{yy})/\sqrt{6} \), where \( e_{xx} \), \( e_{yy} \) and \( e_{zz} \) represent the strains along \( x \), \( y \) and \( z \) axes, respectively.

We imagine the Hamiltonian of the spin system described in terms of \( \mathbf{S}_i \)'s, \( \mathcal{H}_{\text{electron}} \). Then the lowest order part is the classical Heisenberg interaction, but this term leads to a constant with the relation \( A_x^2 + A_y^2 + A_z^2 = 1 \). The next order part is the so-called four-spin interaction and can be represented as \( X_2^2 + X_3^2 \) apart from a constant term. In the same way, we consider higher order interactions. By symmetry consideration, we tentatively adopt the Hamiltonian

\[
\mathcal{H} = \mathcal{H}_{\text{electron}} + \mathcal{H}_{\text{coupling}} + \mathcal{H}_{\text{elastic}},
\]

with

\[
\mathcal{H}_{\text{electron}} = A(X_2^2 + X_3^2) + B(X_3^2 - 3X_2X_3^2) + C(X_2^2 - 3X_2X_3^2)^2, \tag{2}
\]

\[
\mathcal{H}_{\text{coupling}} = g(\varepsilon_2X_2 + \varepsilon_3X_3), \tag{3}
\]

\[
\mathcal{H}_{\text{elastic}} = \frac{\kappa}{2}(\varepsilon_2^2 + \varepsilon_3^2), \tag{4}
\]

where \( \mathcal{H}_{\text{electron}}, \mathcal{H}_{\text{coupling}} \) and \( \mathcal{H}_{\text{elastic}} \) correspond to MSDW states, the coupling between MSDW states and lattice distortion with the positive coupling constant \( g \) and the elastic energy with the elastic constant \( \kappa \), respectively. In the ground state, by using the condition \( \partial \mathcal{H}/\partial \varepsilon_2 = 0 \) and \( \partial \mathcal{H}/\partial \varepsilon_3 = 0 \), which minimizes the Hamiltonian, we obtain \( \varepsilon_2 = -gX_2/\kappa \) and \( \varepsilon_3 = -gX_3/\kappa \), and the Hamiltonian is expressed as

\[
\mathcal{H} = A'(X_2^2 + X_3^2) + B(X_3^2 - 3X_2X_3^2) + C(X_2^2 - 3X_2X_3^2)^2, \tag{5}
\]
with $A' = A - g^2/(2\kappa)$. In our system, the Triple-$Q$ state with $A_x^2 = A_y^2 = A_z^2$ corresponds to the cubic phase, the state with $A_x^2 = A_y^2 < A_z^2$ etc. to the tetragonal one with $c/a < 1$ ($\varepsilon_3 < 0$, $\varepsilon_2 = 0$ etc.), that with $A_x^2 = A_z^2 > A_y^2$ etc. to the tetragonal one with $c/a > 1$ ($\varepsilon_3 > 0$, $\varepsilon_2 = 0$ etc.) and that with $A_x^2 < A_y^2 < A_z^2$ etc. to the orthorhombic one ($\varepsilon_3 \neq 0$, $\varepsilon_2 \neq 0$ etc.). In the absence of the magnetoelastic coupling ($g = 0$), the coefficient $A$ of the leading term $X_x^2 + X_y^2$ is inferred to be positive and the Triple-$Q$ state is expected to be realized for $\gamma$Mn [7]. We therefore set $A = 1.0$, i.e., the unit of energy. In Fig. 2, we show the calculated phase diagram in the $A'$-$B$ plane. The positive (negative) $A'$ prefers the cubic (tetragonal with $c/a < 1$) phase and the positive $B$ can stabilize the tetragonal phase with $c/a > 1$. The positive $C$ is found to be needed to stabilize the orthorhombic phase between the $c/a < 1$ and $c/a > 1$ phases.

Our scenario of the concentration dependence of the position in $A'$-$B$ plane is the following (see Fig. 2). We assume $B > 0$, and note that $g$ is proportional to the square of the sublattice magnetization $S$ ($g^2$ is quartic of $S$). For $x = 0$, due to the large $g^2/(2\kappa)$, $A'$ is expected to be negative, if we note the first-principles electronic structure calculation [6]. With the increase of $x$, $S$ is decreased by the dilution effect and reaches $\sim 0.75S$ for $x \sim 0.25$, where FCC is realized for $T = 0$ K. If we note $g^2 \propto S^4$, $g^2/(2\kappa)$ is reduced to about one-third as $x$ varies from $x = 0$ to $x \sim 0.25$. We may therefore expect the positive $A'$ at $x \sim 0.25$, the change from P($x = 0$) to Q($x \sim 0.25$) in Fig. 2(b) and the successive transition from $c/a < 1$ to FCC passing through the orthorhombic and the $c/a > 1$ phases.

### 3. Phase Diagram

We calculate the partition function $z(\varepsilon_2, \varepsilon_3)$ given by

$$z(\varepsilon_2, \varepsilon_3) = \int_{A_x^2 + A_y^2 + A_z^2 = 1} dA_x dA_y dA_z e^{-\mathcal{H}/(k_BT)}$$

and the free energy $F(\varepsilon_2, \varepsilon_3) = -k_BT \ln z$, and determine the stable structure in the $\sqrt{g}$-$k_BT$ plane. In Fig. 3(a) we show the obtained phase diagram. With the decrease of $\sqrt{g}$, i.e., the increase of $x$, the phase diagram of Mn$_{1-x}$A$_x$ (A = Ni, Ga, Rh and Au) is found to be reproduced at least qualitatively. Although the orthorhombic phase is extremely narrow at $k_BT \approx 0.08$, there exists the orthorhombic phase between the two tetragonal phases over the all range of the temperatures. The magnitude of $X_2$ and $X_3$ is $\sim 1$, while that of $\varepsilon_2$ and $\varepsilon_3$ is $\sim 0.01$. We assume the $g$ and $\kappa$ values so that $\mathcal{H}_{\text{electron}}$, $\mathcal{H}_{\text{coupling}}$ and $\mathcal{H}_{\text{elastic}}$ are comparable to one another.
The temperature dependence of the symmetry strains $\varepsilon_2$ and $\varepsilon_3$ at $p_g = 11.2$ is shown in Fig. 3(b). The transition from the cubic to the tetragonal phase is first order, i.e., the jump of $\varepsilon_3$. The cubic to the orthorhombic transition point, where the $c/a < 1$ and $c/a > 1$ phases meet, is second order. There exists the jump of the symmetry strain $\varepsilon_2$ at the transition temperature from the tetragonal ($c/a < 1$) phase to the orthorhombic one.

From the discussion in Sec. 2, the change of $A_z$ at the transition temperature from one phase to another one is continuous (discontinuous) if the symmetry strain $\varepsilon_3$ varies continuously (discontinuously).

We found that the term $C(X_3^3 - 3X_3X_2^2)^2$ in $H_{\text{electron}}$ (see eq. (2)) with the positive $C$ is necessary for the orthorhombic phase to appear, which corresponds to the 12-spin interaction. Here we examine another origin of the $(X_3^3 - 3X_3X_2^2)^2$ term. If we add the higher order coupling $g'\{\varepsilon_2^2 - \varepsilon_3^2\}X_3 - 2\varepsilon_2\varepsilon_3X_2$ in eq. (3) and the higher order term of the elastic energy $(\kappa'/2)(\varepsilon_2^2 + \varepsilon_3^2)^2$ in eq. (4) and remove $\varepsilon_2$ and $\varepsilon_3$ by using $\partial H/\partial \varepsilon_2 = \partial H/\partial \varepsilon_3 = 0$, we then obtain the $(X_3^3 - 3X_3X_2^2)^2$ term with the positive coefficient. The origin of this term is more likely from the higher order coupling and elastic energy than the 12-spin interaction. The Hamiltonian including the higher order coupling and elastic energy without the $(X_3^3 - 3X_3X_2^2)^2$ term in eq. (2) is found to give the phase diagram similar to Fig. 3(a).

4. Conclusions

In this study we showed that the phase diagram below the Néel temperatures of the first-kind antiferromagnets Mn$_{1-x}$A$_x$ (A = Ni, Ga, Rh and Au) is reproduced by using the Hamiltonian which takes into account the coupling between MSDW states and lattice distortion, and elucidated the relation between MSDWs and lattice distortion.

References

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