J. FAC. SCI., SHINSHU UNIVERSITY, Vol. 30, No. 1, pp. 7-23 1995

*The magnetic structure of the intermetallic compounds in the cubic Laves phase (C15) crystal**

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(Received July 17, 1995)

Abstract

Magnetic structure of intermetallic compounds of rare earth and 3d transition metal with the C15 structure is studied on the basis of the classical Heisenberg model. By making use of the Lyons-Kaplan method, magnetic phase diagram is calculated with respect to the states with the modulation wave vector $\boldsymbol{Q}$ equivalent to [0,0,0] to obtain seven types of spin structure, and their stability is compared with screw structures of $\boldsymbol{Q}$ parallel with [0,0,1], [1,1,0] and [1,1,1]. The stable region of the $\boldsymbol{Q} \equiv$ [0,0,0] states is limited most drastically by the modulation of $\boldsymbol{Q} \| [1,1,0]$.
Key words: magnetic structure, intermetallic compounds, the Laves phase, the classical Heisenberg model, the Lyons-Kaplan method

1 Introduction

The stable magnetic structure of a classical spin system is found out by minimizing the energy with respect to various spin configurations, but the actual calculations are usually complicated and almost impossible to be carried out. Luttinger and Tisza [1, 2] developed a method to make the problem tractable for the Bravais lattice. Lyons and Kaplan [3] generalized the method to treat some non-Bravais lattices and studied the spin configuration of oxide magnets with the spinel structure.

In this paper, we investigate the magnetic structure of the intermetallic compound AB_2 with the cubic Laves phase (C15) structure by making use of the Lyons-Kaplan method, where A is rare earth atom on the 8a site and B 3d-transition metal atom on the 16d site. It is noted that the the atoms on the 8a and 16d sites make up the same structure as the magnetic ions in the ferrimagnet of the normal spinel structure. We investigate the classical Heisenberg model with the exchange couplings between the nearest A-A pair as well as those between the nearest B-B and A-B pairs. Both the

* Based on the theses for M. Sc., Shinshu University, by T. K., 1990, and by Y. T., 1988.

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ferromagnetic and the antiferromagnetic couplings are considered.

Even with the aid of the Lyons-Kaplan method, it is difficult to carry out calculations to determine the characteristic modulation wave vector $\boldsymbol{Q}$ to specify the spin configuration at the ground state in general. So we concern ourselves only in the cases that the ground state is characterized by single pair of $\boldsymbol{Q}$ and $-\boldsymbol{Q}$. In section 3, the magnetic phase diagram for $\boldsymbol{Q}$ equivalent to [0,0,0] is calculated as a function of exchange parameters. The stability of the states with $\boldsymbol{Q}\equiv[0,0,0]$ is examined for the modulations of $\boldsymbol{Q}$ parallel with [0,0,1], [1,1,1] and [1,1,1] in section 4. A brief introduction of the theory is given in section 2 and conclusions are summarized in section 5.

2 The Lyons-Kaplan Method

The energy of the classical Heisenberg model is

$$E_{\mathrm{ex}}=-\sum_{(n\nu,m\mu)} J_{n\nu,m\mu}\boldsymbol{S}_{n\nu}\cdot\boldsymbol{S}_{m\mu},$$

where $\boldsymbol{S}_{n\nu}$ is the classical spin vector on the site $n\nu$ specified by the position vector

$$\boldsymbol{R}_{n\nu}=\boldsymbol{R}_n+\boldsymbol{R}_\nu,\ n=1\sim N,\ \nu=1\sim 6.$$

Here N is the number of unit cells and n and ν run over unit cells and over atoms in the unit cell, respectively, as shown in fig. 1 and table I. The exchange coupling constant $J_{n\nu,m\mu}$ is taken into account for the pairs between the nearest A-A, A-B and B-B atoms, and denoted by J_{AA}, J_{AB} and J_{BB}, respectively.

Our problem is to minimize the energy subject to constraints

$$\boldsymbol{S}_{n\nu}\cdot\boldsymbol{S}_{n\nu}=S_\nu^2 \quad \text{for all } n. \tag{1}$$

Table I. Positions of atoms in a unit cell. a is the length of the edge of the cubic cell.

ν	$\boldsymbol{R}_\nu$	ν	$\boldsymbol{R}_\nu$
1	(0, 0, 0,)	5	(3a/8, 3a/8, 3a/8)
2	(0, a/4, a/4)	6	(5a/8, a/8, a/8)
3	(a/4, a/4, 0)		
4	(a/4, 0, a/4)		

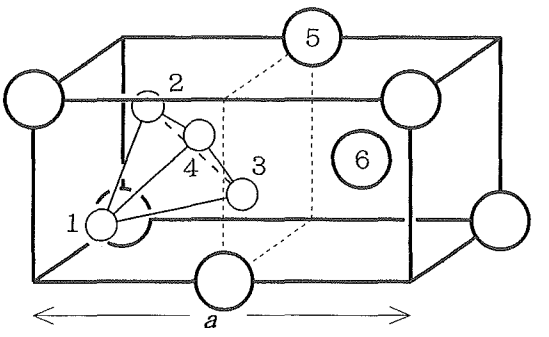

Fig. 1. Unit cell of the C15 structure. The small light circles (Nos. 1 through 4) are the 3d transition atoms on the 16d site, the large heavy circles (Nos. 5, 6) the rare earth atoms on the 8a site. The edge of the cubic cell is denoted by a.

In terms of the Fourier components, $\boldsymbol{S}_{n\nu}$ is represented as

$$\boldsymbol{S}_{n\nu} = S_\nu \sum_{\boldsymbol{q}} \boldsymbol{\delta}_\nu(\boldsymbol{q}) e^{i\boldsymbol{q}\cdot\boldsymbol{R}_{n\nu}},$$

and the exchange energy is transformed into

$$\mathscr{E} = \frac{E_{\mathrm{ex}}}{3N|J_{\mathrm{AB}}|S_{\mathrm{A}}S_{\mathrm{B}}} = \sum_{\boldsymbol{q}} \sum_{\mu\nu} C_{\mu\nu}\boldsymbol{\delta}_\mu(\boldsymbol{q}) \cdot \boldsymbol{\delta}_\nu^*(\boldsymbol{q}),$$

where

$$C_{\mu\nu}(\boldsymbol{q}) = -\frac{S_\mu S_\nu \sum_m J_{m\mu,n\nu} \exp[i\boldsymbol{q}\cdot(\boldsymbol{R}_{m\mu}-\boldsymbol{R}_{n\nu})]}{3|J_{\mathrm{AB}}|S_{\mathrm{A}}S_{\mathrm{B}}},$$

which is independent of $\boldsymbol{R}_n$ because of the translation invariance. The constraints eq. (1) become

$$\sum_{\boldsymbol{q}} \boldsymbol{\delta}_\nu(\boldsymbol{q}) \cdot \boldsymbol{\delta}_\nu^*(\boldsymbol{q}-\boldsymbol{q}') = \begin{cases} 1 & \text{for } \boldsymbol{q}' = 0, \\ 0 & \text{for } \boldsymbol{q}' \neq 0. \end{cases}$$

In this paper, we consider only the case where the spin configuration is characterized by a single pair of modulation wave vectors $\boldsymbol{Q}$ and $-\boldsymbol{Q}$. Then, the energy becomes

$$\mathscr{E} = \sum_{\mu,\nu} C_{\mu\nu}\boldsymbol{\delta}_\mu(\boldsymbol{Q}) \cdot \boldsymbol{\delta}_\nu^*(\boldsymbol{Q}) + c.c., \tag{2}$$

where *c.c.* stands for complex conjugate. The coefficients $C_{n\nu}$ in eq.(2) are expressed in a matrix from

$$\mathscr{C}(\boldsymbol{k}) = \begin{bmatrix} 0 & C_{12} & C_{13} & C_{14} & C_{15} & C_{15}^* \\ C_{12} & 0 & C_{23} & C_{24} & C_{25} & C_{25}^* \\ C_{13} & C_{23} & 0 & C_{34} & C_{35} & C_{35}^* \\ C_{14} & C_{24} & C_{34} & 0 & C_{45} & C_{45}^* \\ C_{15}^* & C_{25}^* & C_{35}^* & C_{45}^* & 0 & C_{56} \\ C_{15} & C_{25} & C_{35} & C_{45} & C_{56}^* & 0 \end{bmatrix}, \tag{3}$$

where the matrix elements are as follows [4]:

$$C_{12} = s\cos(k_y + k_z),\ C_{13} = s\cos(k_x + k_y),\ C_{14} = s\cos(k_z + k_x),$$
$$C_{23} = s\cos(k_z - k_x),\ C_{24} = s\cos(k_x - k_y),\ C_{34} = s\cos(k_y - k_z),$$
$$C_{15} = \mp\frac{1}{3}\{\exp[(\frac{i}{2})(k_x - 3k_y + k_z)] + \exp[(\frac{i}{2})(-3k_x + k_y + k_z)]$$
$$+ \exp[(\frac{i}{2})(k_x + k_y - 3k_z)]\},$$
$$C_{25} = \mp\frac{1}{3}\{\exp[(\frac{i}{2})(k_x + 3k_y - k_z)] + \exp[(\frac{i}{2})(-3k_x - k_y - k_z)]$$
$$+ \exp[(\frac{i}{2})(-k_x - k_y + 3k_z)]\},$$
$$C_{35} = \mp\frac{1}{3}\{\exp[(\frac{i}{2})(3k_x - k_y + k_z)] + \exp[(\frac{i}{2})(-k_x + 3k_y + k_z)]$$

$$+\exp[(\frac{i}{2})(-k_x-k_y-3k_z)]\}$$

$$C_{45}=\mp\frac{1}{3}\{\exp[(\frac{i}{2})(3k_x+k_y-k_z)]+\exp[(\frac{i}{2})(-k_x-3k_y-k_z)]$$

$$+\exp[(\frac{i}{2})(-k_x+k_y+3k_z)]\},$$

$$C_{56}=2t\{\exp(ik_z)\cos(k_x-k_y)+\exp(-ik_z)\cos(k_x+k_y)\}.$$

The upper and the lower parts of the composite signs correspond to positive and negative values of J_{AB}, respectively. Here, we define $\boldsymbol{k}=a\boldsymbol{Q}/4$. a is the lattice parameter and

$$s=-\frac{2J_{BB}S_B^2}{3|J_{AB}|S_AS_B},\qquad t=-\frac{J_{AA}S_A^2}{3|J_{AB}|S_AS_B}.$$

The constraints in the present case are written as

$$\begin{cases}2\vec{\sigma}(\boldsymbol{Q})\cdot\vec{\sigma}^*(\boldsymbol{Q})=1,\\ \vec{\sigma}(\boldsymbol{Q})\cdot\vec{\sigma}(\boldsymbol{Q})=\vec{\sigma}^*(\boldsymbol{Q})\cdot\vec{\sigma}^*(\boldsymbol{Q})=0.\end{cases}$$

Introducing a pair of orthonormal vectors $\mathbf{I}_{\boldsymbol{Q}}$ and $\mathbf{J}_{\boldsymbol{Q}}$, we can represent $\vec{\sigma}_\nu(\boldsymbol{Q})$ as

$$\vec{\sigma}_\nu(\boldsymbol{Q})=\frac{1}{2}u_\nu(\boldsymbol{Q})(\mathbf{I}_{\boldsymbol{Q}}-i\mathbf{J}_{\boldsymbol{Q}}),$$

where $\mathbf{I}_{-\boldsymbol{Q}}=\mathbf{I}_{\boldsymbol{Q}}$ and $\mathbf{J}_{-\boldsymbol{Q}}=-\mathbf{J}_{\boldsymbol{Q}}$. Substituting this expression into eq.(2), we get the energy

$$\mathscr{E}=\frac{1}{2}\sum_{\mu,\nu}C_{\mu\nu}(\boldsymbol{Q})u_\mu(\boldsymbol{Q})u_\nu^*(\boldsymbol{Q})+c.c.$$

and the constraints

$$|u_\nu(\boldsymbol{Q})|^2=1,\qquad \nu=1\sim 6. \tag{4}$$

Lyons and Kaplan [3] treated the complicated variational problem by introducing a 'weak' constraint

$$\sum_\nu \boldsymbol{S}_{n\nu}\cdot\boldsymbol{S}_{n\nu}/\beta_\nu^2=\sum_\nu S_\nu^2/\beta_\nu^2,$$

instead of the 'strong' constraint eq.(1), where the values of β_ν are chosen so that the solution on the 'weak' condition is consistent with the 'strong' constraint [5, 6], The weak constraint is written in terms of the Fourier compoments as

$$\sum_\nu 2\vec{\sigma}_\nu(\boldsymbol{Q})\cdot\vec{\sigma}_\nu^*(\boldsymbol{Q})/\beta_\nu^2=\sum_\nu 1/\beta_\nu^2,$$

or

$$\sum_\nu u_\nu(\boldsymbol{Q})u_\nu^*(\boldsymbol{Q})/\beta_\nu^2=\sum_\nu 1/\beta_\nu^2, \tag{5}$$

By means of the weak constraint condition, our problem is reduced to minimizing

$$\frac{1}{2}\sum_{\mu\nu}C_{\mu\nu}(\boldsymbol{Q})u_\nu(\boldsymbol{Q})u_\nu^*(\boldsymbol{Q})-\lambda\sum_\nu u_\nu(\boldsymbol{Q})u_\nu^*(\boldsymbol{Q})/\beta_\nu^2,$$

where λ is the Lagrange's undetermined multiplier, i.e., we are to solve the eigenvalue proglem

$$\frac{1}{2}\sum_\nu C_{\mu\nu}(\boldsymbol{Q})u_\mu(\boldsymbol{Q})=\frac{\lambda}{\beta_\nu^2}u_\nu(\boldsymbol{Q}),$$

where the eigenvalue corresponds to $\mathscr{E}/2$.

We will refer by a zero-wave-vector ($\boldsymbol{Q}\equiv[0,0,0]$) mode to the state where the characteristic wave vector $\boldsymbol{Q}$ is equivalent to $-\boldsymbol{Q}$, i.e., $\boldsymbol{Q}$ equals to [0,0,0] or a half of a reciprocal lattice vector. It is noted that, when $\boldsymbol{Q}\equiv-\boldsymbol{Q}$, the energy is

$$\mathscr{E}=\sum_{\mu,\nu}C_{\mu\nu}\vec{\sigma}_\mu(0)\cdot\vec{\sigma}_\nu(0)=\sum_{\mu,\nu}C_{\mu\nu}u_\mu(0)u_\nu(0),$$

and the strong constraints are simply written as

$$\vec{\sigma}_\nu(0)\cdot\vec{\sigma}_\nu(0)=1,\qquad \nu=1\sim6,$$

or

$$\vec{\sigma}(0)=u_\nu(0)\mathbf{I}_0$$

with

$$|u_\nu(0)|^2=1.$$

The weak constraint is given by

$$\sum_\nu\vec{\sigma}_\nu(0)\cdot\vec{\sigma}_\nu^*(0)/\beta_\nu^2=\sum_\nu 1/\beta_\nu^2,$$

or

$$\sum_\nu u_\nu(0)u_\nu^*(0)/\beta_\nu^2=\sum_\nu 1/\beta_\nu^2.$$

3 The Zero-Wave-Vector Mode

The matrix $\mathscr{C}$ of eq.(3) for $\boldsymbol{k}=0$ is

$$\mathscr{C}(0)=\begin{bmatrix} 0 & s & s & s & \mp1 & \mp1 \\ s & 0 & s & s & \mp1 & \mp1 \\ s & s & 0 & s & \mp1 & \mp1 \\ s & s & s & 0 & \mp1 & \mp1 \\ \mp1 & \mp1 & \mp1 & \mp1 & 0 & 4t \\ \mp1 & \mp1 & \mp1 & \mp1 & 4t & 0 \end{bmatrix}.$$

By setting the values of β_ν to be 1 for $\nu=1\sim4$, and to be β for $\nu=5,6$ as done in ref. [6], we have the eigenvalue equation

$$\left[\frac{1}{2}\mathscr{C}-\lambda\mathscr{B}\right]\vec{u}(\boldsymbol{k})=0,$$

where $\vec{u}$ is the vector composed of $u_\nu(\boldsymbol{k})$, $\nu=1\sim6$, and

$$\mathscr{B}=\begin{bmatrix}1&0&0&0&0&0\\0&1&0&0&0&0\\0&0&1&0&0&0\\0&0&0&1&0&0\\0&0&0&0&\beta^{-2}&0\\0&0&0&0&0&\beta^{-2}\end{bmatrix}. \tag{6}$$

The secular equation is easily factorized as

$$\left(\lambda+\frac{s}{2}\right)^3\left(\frac{\lambda}{\beta^2}+2t\right)\begin{vmatrix}-\lambda+\frac{3s}{2}&\mp2\\ \mp1&-\frac{\lambda}{\beta^2}+2t\end{vmatrix}=0,$$

and the eigenvalues $\lambda^{(\alpha)}$ and eigenvectors $\vec{u}^{(\alpha)}$ are obtained as shown in table II, where $\vec{u}^{(\alpha)}$ is normalized according to the weak constraint eq. (5) and the abbreviation

$$A=\frac{3}{2}s-\lambda^{(1)}$$

is used.

We leave the eigenstate $\vec{u}^{(4)}$ out of cosideration, hereafter, because evidently $\lambda^{(1)}<\lambda^{(4)}$. The eigenvalues $\lambda^{(1)}$, $\lambda^{(2)}$ and $\lambda^{(3)}$ may degenerate into each other, depending on the values of β. If they are non-degenerate with each other, the strong constraints eq. (4) cannot be satisfied other than $\vec{u}^{(1)}$. In the non-degenerate case, we choose the value of β as

$$\beta^2=\frac{2-3s}{4(1-t)},$$

Table II. The eigenvalues and eigenvectors for the $\boldsymbol{Q}\equiv[0,0,0]$ mode.

α	$\lambda^{(\alpha)}$	$\vec{u}^{(\alpha)}$
1	$\beta^2t+\frac{3}{4}s-\sqrt{(\beta^2t-\frac{3}{4}s)^2}$	$\sqrt{\frac{2\beta^2+1}{2\beta^2+A^2}}(1,\ 1,\ 1,\ 1,\ \pm A,\ \pm A)$
2	$-2t\beta^2$	$\sqrt{2\beta^2+1}\ (0,\ 0,\ 0,\ 0,\ 1,\ -1)$
3	$-\frac{s}{2}$(triply degenerate)	$\sqrt{\frac{2\beta^2+1}{2\beta^2}}\begin{cases}(1,\ 1,\ -1,\ -1,\ 0,\ 0)\\(1,\ -1,\ 1,\ -1,\ 0,\ 0)\\(1,\ -1,\ -1,\ 1,\ 0,\ 0)\end{cases}$
4	$\beta^2t+\frac{3}{4}s+\sqrt{(\beta^2t-\frac{3}{4}s)^2}$	$\sqrt{\frac{2\beta^2+1}{2\beta^2+4/A^2}}(1,\ 1,\ 1,\ 1,\ \mp\frac{2}{A},\ \mp\frac{2}{A})$

and get $A=1$. Thus we obtain the eigenvalue

$$\lambda^{(1)}=\frac{3}{2}s-1$$

and the state vector

$$\vec{u}^{(1)}=(1,1,1,1,\pm 1,\pm 1),$$

which satisfy the strong condition and gives the collinear spin configurations:

$$S_{nv}=\begin{cases} S_B\mathbf{I}_0 & \text{for} \quad v=1\sim 4 \\ \pm S_A\mathbf{I}_0 & \text{for} \quad v=5,6 \end{cases},$$

i. e., the ferromagnetic or Néel type configurations according to the sign of J_{AB}. In fig. 2(a) the projection onto the (001) plain of the examples of these spin configurations are shown. We refer to the structures by FN, hereafter, and the values of β , λ by β_{FN} and λ_{FN}, respectively.

If $\lambda^{(1)}$ is forced to degenerate into $\lambda^{(2)}$,

$$\beta^2=1=\frac{1-3st}{4t^2},$$

$$A=\frac{1}{2t},$$

and

$$\lambda^{(1)}=\lambda^{(2)}=\frac{3st-1}{2t}.$$

The eigenvector may be written as

$$\vec{u}=N_1\vec{u}^{(1)}+N_2\vec{u}^{(2)}.$$

From the strong constraints , we have

$$|N_1|^2=\frac{2\beta^2+A^2}{2\beta^2+1},\quad |N_2|^2=\frac{1-A^2}{2\beta^2+1},$$

and

$$N_1N_2^*+N_1^*N_2=0.$$

Then the state vectors become

$$\vec{u}=(1,1,1,1,\pm e^{\pm i\theta},\pm e^{\mp i\theta})\equiv \vec{u}_{YKA},$$

where θ is defined by $\cos\theta=1/2t$. This state vector gives a kind of Yafet-Kittel's spin structure [8] with spin vectors

$$S_{nv}=S_\nu(1,1,1,1,\pm\cos\theta,\pm\cos\theta)\mathbf{I}_0+S_\nu(0,0,0,0,\sin\theta,-\sin\theta)\mathbf{J}_0.$$

In fig.2 (b) the examples of these spin configurations are shown. We refer to the structures by YKA, and denote the state vector $\vec{u}$ and the values of β and λ by $\vec{u}_{YKA}$,

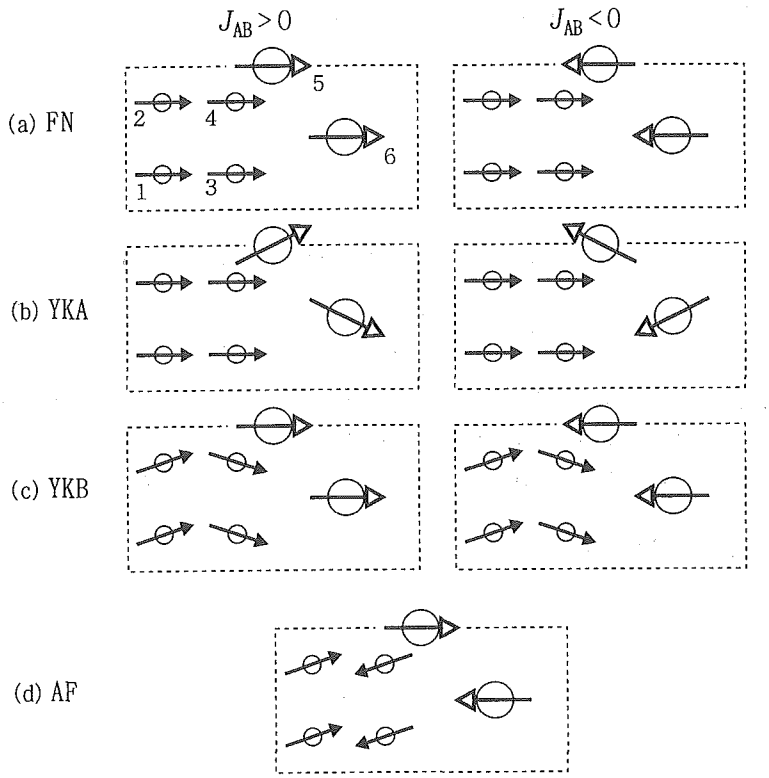

Fig. 2. Spin structures for the $\boldsymbol{Q}=[0, 0, 0]$ state. The number of sites are defined in fig. 1. The arrows show the spin direction, but the direction with respect to the crystal axis is indeterminate and tentative.

β_{YKA} and λ_{YKA}, respectively.

If $\lambda^{(1)}$ is forced to degenerate into $\lambda^{(3)}$, we obtain

$$\beta^2=\frac{s^2}{2(1-2st)},$$
$$A=2s$$

and

$$\lambda^{(1)}=\lambda^{(3)}=-\frac{s}{2}.$$

The state vector is obtained in a similar way to the YKA case as

$$\vec{u}=(e^{i\theta}, e^{i\theta}, -e^{i\theta}, -e^{i\theta}\pm 1, \pm 1)\equiv \vec{u}_{\mathrm{YKB}},$$

where

$$\cos\theta=1/2s.$$

We obtain alternative types of the Yafet-Kittel spin configuration:

$$\boldsymbol{S}_{n\nu} = (S_B\cos\theta, S_B\cos\theta, S_B\cos\theta, S_B\cos\theta, \pm S_A, \pm S_A)\mathbf{I}_0 + (S_B\sin\theta, S_B\sin\theta, -S_B\sin\theta, -S_B\sin\theta, 0, 0)\mathbf{J}_0,$$

which are illustrated in fig.2 (c). We refer to the structures by YKB and denote the state vector $\vec{u}$ and the values of β and λ by $\vec{u}_{YKB}$, β_{YKB} and λ_{YKB}, respectively.

If $\lambda^{(2)} = \lambda^{(3)}$, we choose

$$\beta^2 = \frac{s}{4t},$$

and get

$$\lambda^{(2)} = \lambda^{(3)} = -\frac{s}{2}.$$

The state vector is

$$\vec{u}_{AF} = N_2\vec{u}_2 + N_3\vec{u}_3.$$

In order to satisfy the strong constraints,

$$\left|N_2\sqrt{\frac{2\beta^2+1}{2}}\right| = 1,$$

$$\left|N_3\sqrt{2\beta^2+1}\right| = 1,$$

hence we get

$$\frac{|N_2|^2}{|N_3|^2} = \frac{2t}{s}.$$

Thus the state vector becomes

$$\vec{u} = (e^{i\theta}, e^{i\theta}, e^{-i\theta}, e^{-i\theta}, 1, -1) \equiv \vec{u}_{AF},$$

where θ is a phase difference between N_2 and N_3 and indeterminate within our treatments. The spin vector is

$$\boldsymbol{S} = (S_B\cos\theta, S_B\cos\theta, -S_B\cos\theta, -S_B\cos\theta, S_A, -S_A)\mathbf{I}_0 + (S_B\sin\theta, S_B\sin\theta, -S_B\sin\theta, -S_B\sin\theta, 0, 0)\mathbf{J}_0.$$

An example of this configuration is shown in fig.2 (d). We refer to this structure by AF, and denote the state vector $\vec{u}$ and the values of β and λ by $\vec{u}_{AF}$, β_{AF} and λ_{AF}, respectively.

For Lyons-Kaplan's parameter of eq. (6), we have obtained the seven configurations for $\boldsymbol{Q} \equiv [0,0,0]$ as shown in fig.2. It is also clear that the case of $\lambda^{(1)} = \lambda^{(2)} = \lambda^{(3)}$ does not take place, hence, the configurations YKA and YKB never coexist with each other.

Now we are going to discuss the phase diagram for the $\boldsymbol{Q} \equiv [0,0,0]$ mode. We represent one of the values β_{FN}, β_{YKA}, β_{YKB} or β_{AF} by β_0, and the corresponding eigenvalue by $\lambda_0\ (0, \beta_0)$. If the spin structure for $\lambda_0\ (0, \beta_0)$ is of the ground state, the following

conditions should be satisfied:

$$\lambda_0(0,\beta_0) \leq \lambda^{(\alpha)}(0,\beta_0), \qquad \alpha = 1 \sim 6,$$

where $\lambda^{(\alpha)}(0,\beta_0)$ are the eigenvalues $\lambda^{(\alpha)}$ in table II into which β_0 is substituted for β. And subsidiarily

$$\lambda_0(0,\beta_0) < 0,$$

and

$$\beta_0(s,t) \neq 0.$$

In the cases of YKA and YKB, we have to add

$$0 < A < 1.$$

After straightforward calculations, we obtain the field for each spin structure on the st-plain as follows:

$$\begin{cases} \text{(a)FN} & s < \frac{1}{2},\ t < \frac{1}{2}. \\ \text{(b)YKA} & st < \frac{1}{4},\ t > \frac{1}{2}. \\ \text{(c)YKB} & st < \frac{1}{4},\ s > \frac{1}{2}. \\ \text{(d)AF} & st > \frac{1}{4},\ s > 0, t > 0. \end{cases} \tag{7}$$

These fields are illustrated in fig.3.

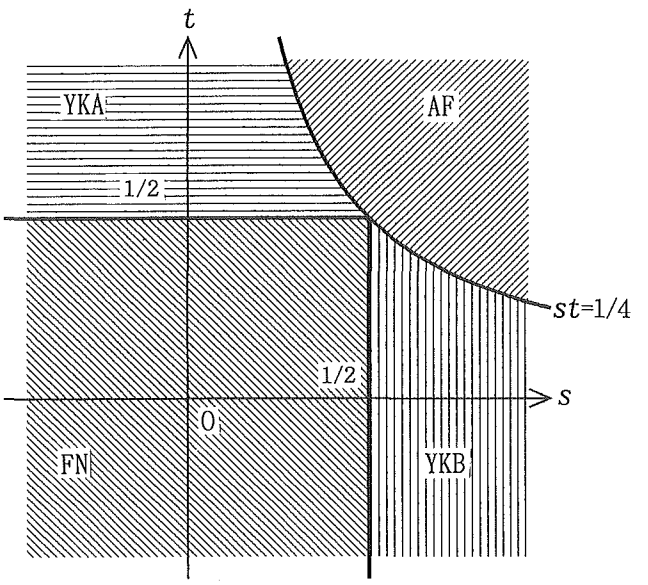

Fig. 3. Phase diagram for the $\boldsymbol{Q} \equiv [0, 0, 0]$ structures.

4 Stability Relative to Screw Structures

(1) $\boldsymbol{Q} \| [0,0,1]$ *modulation*

By substituting $\boldsymbol{k} = [0,0,\boldsymbol{k}]$ into eq. (3), we have

$$\mathscr{C}([0,0,k]) = \begin{bmatrix} 0 & a & b & a & f & f^* \\ a & 0 & a & b & f^* & f \\ b & a & 0 & a & f & f^* \\ a & b & a & 0 & f^* & f \\ f & f^* & f & f^* & 0 & e \\ f^* & f & f^* & f & e & 0 \end{bmatrix},$$

where

$$a = \frac{s}{2}\cos k, \quad e = 2t\cos k.$$

$$b = \frac{s}{2}, \qquad f = -\frac{1}{6}\left[2\exp(i\frac{k}{2}) + \exp(-i\frac{3k}{2})\right],$$

The secular equation is decomposed by the similarity transformation with

$$\frac{1}{2}\begin{bmatrix} 1 & 1 & 1 & 1 & 0 & 0 \\ 1 & 1 & -1 & -1 & 0 & 0 \\ 1 & -1 & 1 & -1 & 0 & 0 \\ 1 & -1 & -1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & \sqrt{2} & \sqrt{2} \\ 0 & 0 & 0 & 0 & \sqrt{2} & -\sqrt{2} \end{bmatrix}$$

into two submatrices:

$$\mathscr{A}(k)\vec{x} = \Lambda\vec{x},$$
$$\mathscr{B}(k)\vec{y} = \Lambda\vec{y},$$

where

$$\mathscr{A}(k) = \begin{bmatrix} a+b & c & 0 \\ c & e & 0 \\ 0 & 0 & -b \end{bmatrix}, \qquad \Lambda(k) = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & \lambda/\beta^2 & 0 \\ 0 & 0 & \lambda \end{bmatrix},$$

$$\mathscr{B}(k) = \begin{bmatrix} -a+b & -id & 0 \\ id & -e & 0 \\ 0 & 0 & -b \end{bmatrix},$$

$$\vec{x}=\begin{bmatrix} \frac{1}{2}(u_1+u_2+u_3+u_4) \\ \frac{1}{\sqrt{2}}(u_5+u_6) \\ \frac{1}{2}(u_1+u_2-u_3-u_4) \end{bmatrix}, \quad \vec{y}=\begin{bmatrix} \frac{1}{2}(u_1-u_2+u_3-u_4) \\ \frac{1}{\sqrt{2}}(u_5-u_6) \\ \frac{1}{2}(u_1-u_2-u_3+u_4) \end{bmatrix},$$

and

$$c=-\frac{\sqrt{2}}{3}\left(2\cos\frac{k}{2}+\cos\frac{3k}{2}\right), \quad d=-\frac{\sqrt{2}}{3}\left(2\sin\frac{k}{2}-\sin\frac{3k}{2}\right).$$

Because the secular equation with respect to $\mathscr{B}(k)$ is equivalent to that with respect to $\mathscr{A}(k+\pi)$, it is enough to study about $\mathscr{A}$. Three eigenvalues of $\mathscr{A}$ are easily calculated as

$$\lambda(k,\beta)_{\pm}=\frac{1}{2}\left(2\beta^2 t\cos k+s\cos k+\frac{1}{2}s\right)\pm\frac{1}{2}\left\{(2\beta^2 t\cos k+s\cos k+\frac{1}{2}s)^2 -4\beta^2\left[ts(2\cos k+1)\cos k-\frac{1}{9}(1+\cos k)(2\cos k+1)^2\right]\right\}^{1/2},$$

$$\lambda^{(3)}=-\frac{1}{2}s.$$

The stability of each phase of the $\boldsymbol{Q}\equiv[0,0,0]$ mode is examined with respect to the lowest eigenvalue $\lambda(k,\beta)_-$ as follows:

(a) *FN structure.* The necessary condition for the FN structure to be stable compared with $\boldsymbol{Q}\parallel[0,0,1]$ modulation is

$$\lambda_{\mathrm{FN}}<\lambda(k,\ \beta_{\mathrm{FN}})_-.$$

After straightforward but rather tedious calculations, the following boundary for the FN structure is obtained:

$$t<\frac{2}{9s^2}\left[(2-3s)(1-2s)^{1/2}+5s-2\right] \quad \text{for} \quad \frac{4}{9}\leq s<\frac{1}{2}$$

(b) *YKA structure.* The condition

$$\lambda_{\mathrm{YKA}}<\lambda(k,\ \beta_{\mathrm{YKA}})_-$$

shifts the boundary for the YKA structure to

$$ts<\frac{2}{9} \qquad \text{for } t>\frac{1}{2}.$$

(c) *YKB structure.* The condition

$$\lambda_{\mathrm{YKB}}<\lambda(k,\ \beta_{\mathrm{YKB}})_-$$

shifts the boundary for the YKB structure to

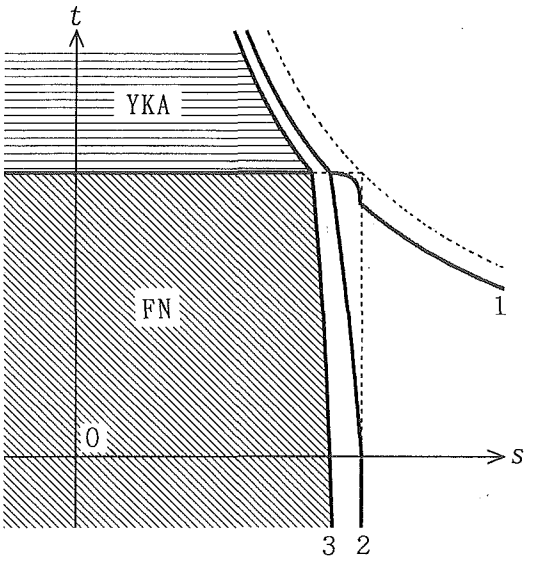

Fig. 4. Phase boundary limited by screw structures. The curves 1, 2, 3 show the boundaries in cases of $\boldsymbol{Q}\parallel[0,0,1]$, $\boldsymbol{Q}\parallel[1,1,1]$ and $\boldsymbol{Q}\parallel[1,1,0]$ modulation, respectively.

$$ts<\frac{2}{9} \qquad \text{for } s<\frac{1}{2}.$$

(d) *AF structure.* The condition

$$\lambda_{AF}<\lambda(k,\beta_{AF})_{-}$$

is broken over the whole region on st-plain, i. e., the AF structure is never realized for any values of s and t. The boundary limited by $[0,0,1]$ mode is shown by the curve 1 in fig.4.

(2) $\boldsymbol{Q}\parallel[1,1,1]$ *modulation*

The coefficient matrix eq. (3) for $\boldsymbol{k}=(\kappa,\kappa,\kappa)$, $\kappa=k/\sqrt{3}$, is

$$\mathbf{C}=\begin{pmatrix} 0 & a & a & a & f & f^* \\ a & 0 & b & b & g & g^* \\ a & b & 0 & b & g & g^* \\ a & b & b & 0 & g & g^* \\ f^* & g^* & g^* & g^* & 0 & e \\ f^* & g & g & g & e^* & 0 \end{pmatrix},$$

where

$$a=\frac{s}{2}\cos 2\kappa, \quad e=t(e^{ik}+e^{-ik}\cos 2k),$$
$$b=\frac{s}{2}, \qquad f=\mp\frac{1}{2}\exp(-i\kappa/2),$$
$$g=\mp\frac{1}{6}[2\exp(3i\kappa/2)+\exp(-5i\kappa/2)],$$

the upper and lower parts of the composite signs correspond to the positive and negative signs of J_{AB}. The secular equation is easily factorized as

$$(\lambda+b)^2\begin{vmatrix} -\lambda & a & f & f^* \\ 3a & -\lambda+2b & 3g & 3g^* \\ f^* & g^* & -\lambda/\beta^2 & e \\ f & g & e^* & -\lambda/\beta^2 \end{vmatrix}=0. \tag{8}$$

Thus the eigenvalues are

$$\lambda(\boldsymbol{k},\beta)=-\frac{s}{2}, \text{ doubly degenerate,}$$

and

$$\lambda'_\gamma(\boldsymbol{k},\beta), \qquad \gamma=1\sim 4,$$

the roots of 4×4 determinant. Because $\lambda(0,\beta_0)<-s/2$ for $\boldsymbol{Q}\equiv[0,0,0]$ mode, it is enough for our purpose to examine the condition

$$\lambda(0,\beta)<\lambda'_\gamma(\boldsymbol{k},\beta).$$

This condition is satisfied if the 4×4 determinant obtained by substituting β_0 and $\lambda(0,\beta_0)$ for β and λ in eq.(8) is positive definite, i. e., all the principal minors are positive. After straightforward but laborious calculations, we obtain the following results:

(a) *FN structure.* The first diagonal element is $-\lambda_{FN}>0$. The upperleft 2×2 principal minor is of a quadratic form for s and positive in the region bounded by eq. (7). From the condition that the full 4×4 determinant is positive, the FN structure is instable out of the region

$$t<\frac{3(1-2s)}{2-3s} \quad \text{for } \frac{4}{9}<s<\frac{1}{2}.$$

The 3×3 principal minor does not add any boundary.

(b) *YKA structure.* The same boundary as that for [0, 0, 1] modulation is obtained.

(c) *YKB structure.* YKB is instable out of the region

$$st<0 \quad \text{for } s>\frac{1}{2}.$$

(d) *AF structure.* The AF structure is completely eliminated by [1, 1, 1] modulation. The boundary limited by [1, 1, 1] is shown by curve 2 in fig.4.

(3) $\boldsymbol{Q} \| [1, 1, 0]$ *modulation*

The coefficient matrix for $\boldsymbol{k}=(\kappa, \kappa, 0)$, $\kappa = k/\sqrt{2}$, is

$$\mathscr{C}(\boldsymbol{k})=\begin{bmatrix} 0 & a/2 & b & a/2 & f & f^* \\ a/2 & 0 & a/2 & 0 & g & g \\ b & a/2 & 0 & a/2 & f^* & f \\ a/2 & c & a/2 & 0 & g & g \\ f^* & g & f & g & 0 & e \\ f & g & f^* & g & e & 0 \end{bmatrix},$$

where

$$a = s\cos\kappa, \qquad e = t(1+\cos 2\kappa),$$
$$b = \frac{s}{2}\cos 2\kappa, \qquad f = \mp\frac{1}{6}(2\exp(-i\kappa)+\exp i\kappa),$$
$$c = \frac{1}{2}s, \qquad g = \mp\frac{1}{6}(1+2\cos 2\kappa),$$

and the upper and lower parts of composite signs correspond to the positive and negative signs of J_{AB}, respectively. The secular equation is factorized [4] after a similarity transformation by

$$\frac{1}{\sqrt{2}}\begin{bmatrix} 1 & 0 & 1 & 0 & 0 & 0 \\ 1 & 0 & -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 1 \\ 0 & 0 & 0 & 0 & 1 & -1 \end{bmatrix}$$

into three factors as

$$(-c-\lambda)\begin{vmatrix} -b-\lambda & f-f^* \\ f^*-f & -e-\dfrac{\lambda}{\beta^2} \end{vmatrix}\begin{vmatrix} -b-\lambda & f+f^* & a \\ f+f^* & e-\dfrac{\lambda}{\beta^2} & 2g \\ a & 2g & c-\lambda \end{vmatrix}=0. \tag{9}$$

Because $-c-\lambda_0>0$ for $\lambda_0 \leq -s/2$, our problem is reduced to consider the condition that both the 2×2 and 3×3 determinants those are obtained by substituting β_0 and $\lambda(\boldsymbol{k}, \beta_0)$ are positive definite, as discussed in the case of [1, 1, 1] modulation. It is noted that the first 2×2 principal minor of the 3×3 determinant is always positive.

(a) *FN structure.* The stable region is given by

$$t<\frac{1}{2} \text{ for } s \leq s_0,$$

and

$$t<\frac{4\sqrt{s}\,(7-6s)-8(1+s)}{9s(1-2s)}, \quad \text{for } s_0<s<\frac{1}{2},$$

where $s_0=0.41251$, which is numerically calculated from the condition that the 3×3 determinant is positive definite.

(b) *YKA structure*. The stable region is limited by

$$s<(st)_0=0.20625 \text{ and } t>\frac{1}{2},$$

where $(st)_0$ is obtained numerically from the condition that the 3×3 determinant is positive definite.

The field of of YKB and AF structures is eliminated by $\boldsymbol{Q}\|[1,1,0]$ modulation. The boundary limited by $\boldsymbol{Q}\|[1,1,0]$ modulation shown by the curve 3 in fig.4.

5 Summaries

In this paper, the seven types of spin structure for the intermetallics with the C15 crystal are deduced on the basis of the classical Heisenberg model by making use of the Lyons-Kaplan method in the case of $\boldsymbol{Q}\equiv[0,0,0]$. The $\boldsymbol{Q}\equiv[0,0,0]$ spin structure becomes unstable compared with the screw spin modulation along $[0,0,1]$,$[1,1,1]$ and $[1,1,0]$ direction in some regions on st-plane. The $[1,1,0]$ mode gives the most drastic limitation. It should be emphasized that, at most, the FN and YKA structures are expected for the $\boldsymbol{Q}\equiv[0,0,0]$ states as shown in fig.4. The remaining fields for $\boldsymbol{Q}\equiv[0,0,0]$ states are limitted as follows:

(a) *FN structure*

$$t<\frac{1}{2} \quad \text{for } s\leq s_0=0.41251$$

$$t<\frac{4\sqrt{s}\,(7-6s)-8(1+s)}{9s(1-2s)}, \quad \text{for } s_0<s<\frac{1}{2},$$

(b) *YKA structure*

$$t>\frac{1}{2} \quad \text{for } st<(st)_0=0.20625.$$

Acknowledgment. The authores would like to thank Professor H. Yamada for his critical reading of the manuscript.

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