A Model for Supersymmetry — An Extension of the Chen-Walker's Model —

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A model for incommensurate transitions proposed by Chen and Walker (Phys. Rev. B **43** (1991) 5634) has been extended to describe a sub-lattice model which possesses supersymmetry. If a zone boundary mode freezes, a superstructure with supersymmetry can appear and may transform into a superstructure without supersymmetry at lower temperature. Although both superstructures belong to the same space group, the transition is either second or first order one with accompanying a definite anomaly in specific heat and susceptibility.

KEYWORDS: free energy model, structural transition, superstructure, supersymmetry

§1. Introduction

It is well known that a modulated structure can be stabilized if the nearest-neighbor interaction is competing with the second-neighbor interaction in an Ising spin model.[1] In general, the modulated structure has an incommensurate period against the basic structure, however, it becomes commensurate at lower temperature. From the numerical solution of the so-called ANNNI model,[2] it has been demonstrated that high-order commensurate phases appear successively and the modulation wave number changes as if a devil's staircase.[3] The symmetry property of the incommensurate phase has been successfully analyzed by the idea of 3+d dimensional superspace group.[4] Although the character of the commensurate phase is believed to be understood within the framework of the ordinal 3-dimensional space group, the symmetry of the superstructure must be considered carefully.

For the ANNNI model or in some extended models, one Ising spin is located in a unit cell of the basic structure. On the other hand, one-degree of freedom of lattice distortion is assigned in the ϕ^4-model describing the modulated structures.[5] In any way, a simplified theoretical model with one-degree of freedom is easy to treat theoretically and is tutorial to get an essence of the incommensurate-commensurate phase transition. But a real crystal may consist of two or more sub-lattices, and may exhibit a complicated structural change at low temperature. For example, two distinct commensurate structures with the same periodicity can appear, and the transformation between them may take place.

In studying a twofold commensurate structure, we have explained the additional extinction rule with introducing supersymmetry:[6] an extra symmetry operation that does not belong to a three-dimensional space group.[7,8] In this report we discuss whether a structure which possesses supersymmetry is stable or not on the basis of a thermodynamic potential for a sub-lattice model.

Since supersymmetry is additional symmetry between two sub-lattices within a unit cell, a more realistic model for the sub-lattice is needed in further discussions. A sub-lattice model which explains modulated structure has been proposed by Chen and Walker.[9] They considered A_2BX_4-type crystals which perform a successive transition from the normal phase (space group $Pmcn$: Z=4) to the incommensurate phase, followed by some commensurate phases. In the incommensurate phase, atomic displacements that belong both to Λ_2 and Λ_3 normal modes are responsible to the modulated structures.[10] Chen and Walker assigned two variables v_l and w_l to represent the atomic displacement of these two modes on the l-th layer; a unit cell of the normal phase consists of two layers: even-l and odd-l. The modulation vector is perpendicular to the layer and the interaction energy is given as the following:[9]

$$U = \sum_l [\frac{a}{2}v_l^2 + \frac{a'}{2}w_l^2 + \frac{b}{4}v_l^4 + \frac{b'}{4}w_l^4 + \frac{\zeta}{2}v_l^2 w_l^2 + \frac{J}{2}v_l v_{l+1} + \frac{J'}{2}w_l w_{l+1} + \frac{\xi}{2}(v_l w_{l+1} - v_{l+1}w_l)]. \quad (1.1)$$

Here, a, a', b, b', ζ, J, J' and ξ are constant parameters.

Although it is possible in principle to calculate the free energy from the above interaction energy by means of statistical mechanics, such an approximation as the mean field theory would be necessary to get a concrete expression for the free energy. Finally the each coefficient in eq. (1.1) contains a temperature dependence if the expression is considered as the free energy expansion. The Landau theory requires a critical temperature dependence on the binary terms as

$$a = a_0(T - T_0),\ a' = a'_0(T - T'_0), \quad (1.2)$$

if a second-order phase transition takes place, as well as a week first-order transition. Other coefficients are expected to depend little on temperature. If the variables u_l, w_l are considered as order parameters on the l-th layer and the coefficients a and a' have the temperature dependence of eq. (1.2), then eq. (1.1) is the phenomenological free energy investigated by Chen and

Walker.[9] They minimized the free energy numerically with respect to u_l and w_l, and obtained a set of phase diagrams explaining a number of commensurate phases realized in A_2BX_4-type crystals.

§2. Model Free Energy

Now we take into account of quadratic invariant ignored in the model of eq. (1.1). The following terms should be added:

$$V = \sum_l [\frac{\beta}{4}(v_l^2 v_{l+1}^2 + w_l^2 w_{l+1}^2) + \frac{\zeta'}{2} v_l^2 w_{l+1}^2 + \frac{\eta}{2}(v_l^3 w_{l+1} - v_{l+1}^3 w_l + v_l w_{l+1}^3 - v_{l+1} w_l^3) + \frac{\gamma}{6}(v_l^6 + w_l^6)]. \quad (2.1)$$

Here a sixth order term is also added with $\gamma > 0$. In general all the coefficients for fourth and sixth order terms may depend on temperature, it is ignored here. By the way, the order parameters v_l and w_l can be scaled so as to put $b = b' = 1$. The interaction energy itself can also be scaled and the ξ may be either 1 or -1.

Let's consider a superstructure whose lattice parameter perpendicular to the layer is double the prototype phase; the superstructure consists of four sub-layers of $l,\ l+1,\ l+2,\ l+3$. Then the order parameters can be represented by Fourier components as

$$v_l = v\cos(\frac{\pi}{2}l + \psi), \quad (2.2a)$$

$$w_l = w\sin(\frac{\pi}{2}l + \theta). \quad (2.2b)$$

Substituting eq. (2.2) into eq. (1.1) and eq. (2.1), we get the free energy per one layer as

$$f = \frac{a}{4}v^2 + \frac{a'}{4}w^2 + \frac{\xi}{2}\cos(\psi-\theta)vw + \frac{3+\beta}{32}(v^4+w^4) + \frac{1-\beta}{32}(v^4\cos 4\psi + w^4\cos 4\theta) + \frac{1}{8}[\zeta + \zeta' + (\zeta-\zeta')\cos 2\psi\cos 2\theta]v^2w^2 + \frac{\eta}{8}[\cos(3\psi+\theta) + 3\cos(\psi-\theta)]v^3w + \frac{\eta}{8}[\cos(\psi+3\theta) + 3\cos(\psi-\theta)]vw^3 + \frac{\gamma}{96}[5(v^6+w^6) + 3(v^6\cos 4\psi + w^6\cos 4\theta)]. \quad (2.3)$$

The order parameters v_l and w_l, introduced by Chen and Walker, belong to the irreducible representations Λ_2 and Λ_3, respectively. Since these two representations degenerate at the Brillouin zone boundary $k_z = \frac{1}{2}c^*$, we may expect as $a = a'$ in the superstructure case. Then the phases ψ, and θ for the modulation can be determined so that the free energy should be minimized. We can classify the following cases:

[i] $\psi = \theta = \frac{\pi}{4}, \frac{3\pi}{4}, \frac{5\pi}{4}, \frac{7\pi}{4}$:

$$f = \frac{a}{4}(v^2+w^2) + \frac{1+\beta}{16}(v^4+w^4) + \frac{\zeta+\zeta'}{8}v^2w^2 + \frac{\gamma}{48}(v^6+w^6) + \frac{\xi}{2}vw + \frac{\eta}{4}vw(v^2+w^2) \quad (2.4)$$

[i'] $\psi = \theta - \pi = \frac{\pi}{4}, \frac{3\pi}{4}, \frac{5\pi}{4}, \frac{7\pi}{4}$: In the above expression, ξ and η have the alternated sign.

[ii] $\psi = \theta = 0, \frac{\pi}{2}, \pi, \frac{3\pi}{2}$:

$$f = \frac{a}{4}(v^2+w^2) + \frac{1}{8}(v^4+w^4) + \frac{\zeta'}{4}v^2w^2 + \frac{\gamma}{12}(v^6+w^6) + \frac{\xi}{2}vw + \frac{\eta}{2}vw(v^2+w^2) \quad (2.5)$$

[ii'] $\psi = \theta - \pi = 0, \frac{\pi}{2}, \pi, \frac{3\pi}{2}$: In the above expression, ξ and η have the alternated sign.

As noted at eq. (1.2), a and a' depend linearly on temperature, and the other expansion coefficients in the free energy are considered to be constant. At high temperature, stable is the prototype phase (it is called as para-state, hereafter): $v = 0$ and $w = 0$. At low temperature, the ordered phase is either $v = -w \neq 0$ for $a < \xi$ or $v = w \neq 0$ for $a < -\xi$; the former is nominated as anti-state and the latter ferro-state in the following.

§3. Ferri-State Solution

The last free energy eq. (2.4) or eq. (2.5) is identical to the free energy demonstrating semi-polar phase ($v \neq w \neq 0$; it is called as ferri-state, hereafter) discussed by Suzuki and Okada two decades ago.[11] The model was originally introduced to extend the Kittel's sub-lattice model for antiferroelectricity. Suzuki and Okada found the following sequence of transitions with decreasing temperature;

para →	ferri →	anti or ferro
$v = w = 0$	$v \neq w \neq 0$	$\lvert v\rvert = \lvert w\rvert \neq 0$

It is easy to get other transition sequences

para → anti(or ferro) → ferri or ferro(or anti),
or → ferri → ferro(or anti),

with choosing the coefficients of the free energy appropriately.[12] A typical temperature dependence of the order parameters v and w is shown in Fig. 1.

We assume that the space group of the prototype phase is $Pmcn$, and that the modulation structure has the wave number along the c-axis, as same as Chen and Walker have discussed about A_2BX_4-type crystals. Then the modulation of the k-th atom on the l-th sublayer can be expressed as

$$\boldsymbol{u}_{[l,k]} = v_l \boldsymbol{e}_k(\Lambda_2) + w_l \boldsymbol{e}_k(\Lambda_3),$$

where $\boldsymbol{e}_k(\Lambda_i)$ is the eigen-vector of the Λ_i representation. There are two kind of superstructures that can be induced by the freezing of the zone boundary modes:

$$\text{Type I}: P2_1/c11 \ : \ v_l : \uparrow\downarrow\downarrow\uparrow \qquad w_l : \uparrow\uparrow\downarrow\downarrow$$

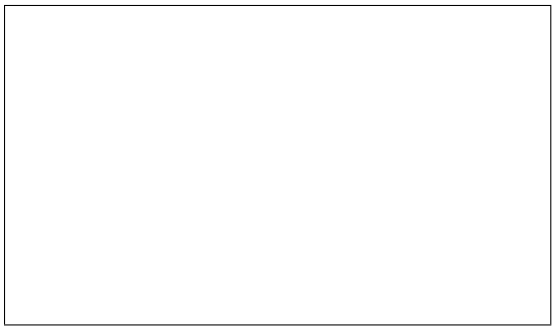

Fig. 1. The temperature dependence of the order parameters v and w in free energy eq. (2.5). The coefficients of the free energy are given by $\beta = 4,\ \gamma = 0.1,\ \xi = 0.5,\ \eta = 0.1,\ \zeta = 0,\ \zeta' = 2$, for which eq. (2.5) gives more stable solution than eq. (2.4).

$$\text{Type II}: Pc2_1n \qquad : v_l : \cdot \uparrow \cdot \downarrow$$
$$w_l : \uparrow \cdot \downarrow \cdot$$

Here arrows indicate a translation along the x-axis or a rotation of BX_4 tetrahedra about the axis nearly parallel to the c-axis, and $\cdot$ means the magnitude of the modulation is vanishing. It is clear that Type I and Type II correspond to [i] and [ii] of eq. (2.4) and eq. (2.5), respectively.

The crystal structure of $LiRbSO_4$ is essentially similar to A_2BX_4-type crystal. The prototype phase in high temperature belongs to the space group of $Pmcn$. Through the phase transition around 475K, the crystal takes a superstructure whose c-axis is double the prototype phase. The superstructure of the space group of $P2_1/c11$ is schematically drawn in Fig. 2; the type I structure. The rotation senses of a SO_4 tetrahedron corresponding to v_l and w_l are indicated by black and white arrows, respectively. The numbering on the tetrahedron is the same as Fig. 1 in the previous report.[6)]

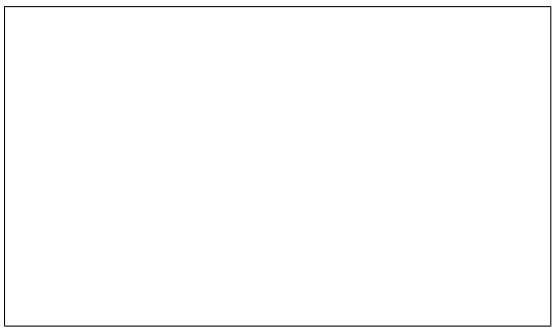

Fig. 2. Projection of the superstructure (phase III) of $LiRbSO_4$ along the b-axis. The normal modes Λ_2 and Λ_3 for the rotation of tetrahedra SO_4 are indicated by black and white arrows, respectively.

If the type I, $P2_1/c11$ symmetry, is realized, then the rotation angles of tetrahedra 3, 4, 5, and 6 should be vanishing because of $|v| = |w| \neq 0$. But according to the crystal structure analysis for the phase III of $LiRbSO_4$, [13)] $v \simeq 0$ and $w \neq 0$. Such a solution as $v = 0$ and $w \neq 0$ is possible, if $a \neq a'$ in eq. (1.2). Indeed, $v = 0$ and $w \neq 0$ should be a solution in a temperature range of $T_0' > T > T_0$. In $LiRbSO_4$, it is considered that a mode belonging to a general point near the zone boundary on the Λ_3 branch freezes at first, and that an incommensurate structure is realized.[14)] With further cooling, the zone boundary modes freezes to realize the superstructure, where the atomic displacement of Λ_3 mode is dominant. Therefore the realized structure of $P2_1/c11$ is considered to satisfy $v \simeq 0$ and $w \neq 0$, reasonably.

§4. Supersymmetry

Let's discuss about the relation between the frozen mode and supersymmetry. As a concrete model, we consider the space group $P2_1/c$ (type I structure) in $LiRbSO_4$. Four tetrahedra 1, 2, 7 and 8 in Fig. 2 are related with each other by the proper symmetry operations of $P2_1/c$, and the rotation angle of the tetrahedra is ψ_1. On the other hand, other tetrahedra 3, 4, 5 and 6 are related to have the same rotation angle ψ_2. In general these two roation angles ψ_1 and ψ_2 are different a little. That is, the white and black arrows in Fig. 2 have different magnitude; the superposed angle is the real rotation angle of the tetrahedron. Therefore, a sub-lattice composed of tetrahedra 1, 2, 7 and 8 is not equivalent to another sub-lattice composed of terahedra 3, 4, 5 and 6. It is hard to extinguish scattered X-ray from these two non-equivalent sub-lattices .

However, imagine that $T_0' > T_0$ and the solution of $v = 0$ and $w \neq 0$ exists in a temperature range of $T_0' > T > T_0$. Then all eight tetrahedra in Fig. 2 rotate as white arrows (the black arrows are vanishing) with the same rotation angle ψ. In this case, there is the following *symmetry operation* between these two sub-lattices:

$$(m_y|\frac{b}{2} + \frac{c}{4}).$$

It is easy to show the systematic absence of scattered X-ray with $h0l : \ l = 4m + 2$. This extra extinction rule is not found within the framework of the three-dimensional space group. In order to represent such a *symmetry operation* to impose the extra extinction rule, we call the additional symmetry as supersymmetry. In the present case, $v = 0$ and the phase θ of w is fixed to be $\cos 4\theta = -1$. These conditions satisfy the supersymmetry between the frozen atomic displacements. On further decreasing the temperature, the solution of $v \neq 0$ appears for $T < T_0$. Then the black angles are not vanishing and the supersymmetry disappears.

It should be noted that there is another supersymmetry in the case of the $P2_1cn$ space group(type II). If $|v| = |w| \neq 0$ in eq. (2.5), then tetrahedra 1, 3, 2 and 4 rotate as black, white, black and white arrows, and tetrahedra 5, 8, 6 and 7 rotate as black, white, black and white in Fig. 2. Since all the rotation angles have the same magnitude, the sub-lattice constituted with the tetrahedra 1, 2, 5 and 6 is related with another sub-lattice with 3, 4, 7 and 8; supersymmetry exists between 1 and 3, and others. The type II space group emerges

in the case of $a = a'$, when the mode freezes just at the zone boundary. With decreasing temperature, the anti- or ferro-state appears at first, which is followed by the ferri-state where the supersymmetry does not hold any more as indicated in Fig. 1 The order parameter changes with a kink at the transition point. The space group is the same both above and below the transition temperature, within the framework of the traditional space group theory. However, the high-temperature phase has the supersymmetry and the breakdown of the supersymmetry takes place through the transition.

Here it should be noted that the loss of supersymmetry accompanies an anomaly in the specific heat, as demonstrated in Fig. 3. A susceptibility will also indicate an anomaly.

Fig. 3. The temperature dependence of a specific heat c. The c/T is proportional to $-\frac{d(v^2+w^2)}{d\alpha}$ in the phenomenological theory, where the coefficients are the same as given in Fig. 1.

§5. Conclusion

In this article, we have taken fourth-order invariants, which have not been considered by Chen and Walker, into account. It has been shown that the transition from the anti-state (or ferro-state) to the ferri-state can take place. The free energy model is equivalent to the sublattice model investigated by Suzuki and Okada, who added some fourth-order terms into the famous Kittel's model for antiferroelectricity.

When a twofold commensurate phase appears, with doubling the c-axis, from the prototype phase of $Pmcn$ in A_2BX_4-type crystals, there are two possible cases for supersymmetry. First, a Λ_3-mode freezes at the zone boundary under the condition of $a > a'$. The modulation amplitude is given by $v = 0$ and $w \neq 0$, and the phase is fixed through the fourth-order terms as $\theta = \frac{\pi}{4} \bmod \frac{\pi}{2}$. The space group of the commensurate phase is $P2_1/c$: type I.

Second, two modes of Λ_3 and Λ_2 symmetry freeze at the zone boundary simultaneously. The modulation amplitude satisfies $|v| = |w| \neq 0$ and the phases are θ, $\phi = \frac{\pi}{2} \bmod \frac{\pi}{2}$. The structure with the supersymmetry has the space group $P2_1cn$: type II.

In both cases, it is expected that the transition from the anti-state to ferri-state occurs at T_c with accompanying the breaking of the supersymmetry. The transition is either the second- or first-order one; the space groups of both above and below T_c are the same within the framework of the three dimensional space group. Therefore it is a kind of symmetry-invariant transition, however, the loss of supersymmetry happens at T_c. The order parameter changes as if usual structural phase transitions between two modulated structures. We can expect a specific heat anomaly as shown in Fig. 3, as well as some critical change of a susceptibility.

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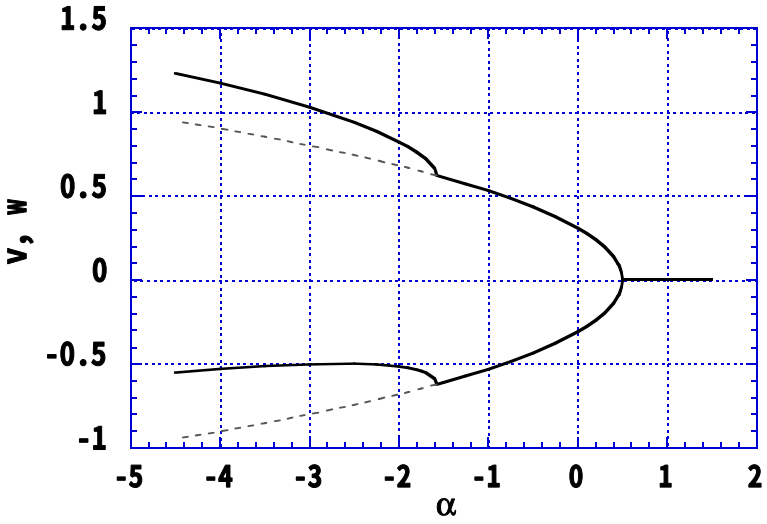

Fig. 1

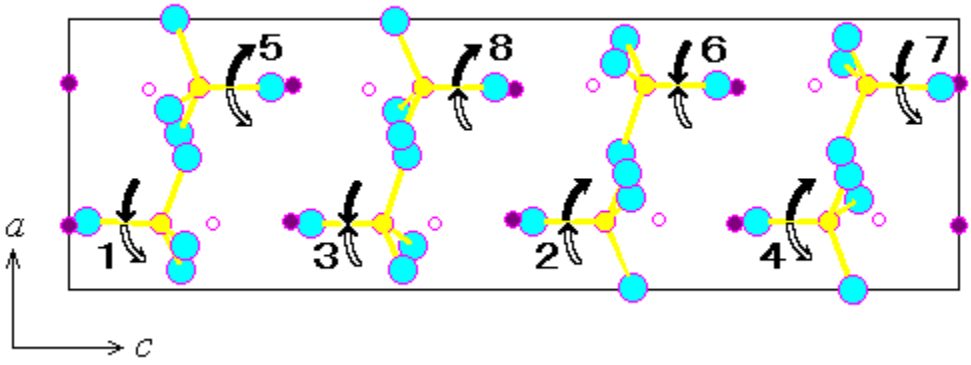

Fig. 2.

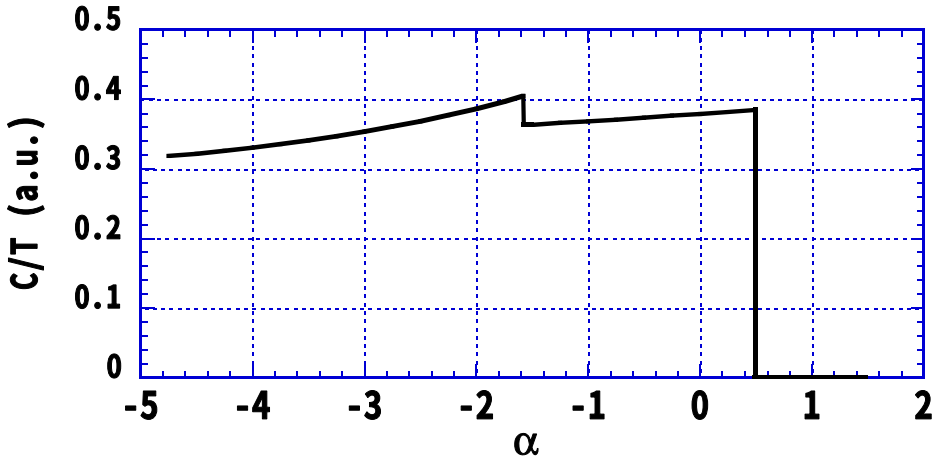

Fig. 3.