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Ginzburg–Landau Theory of Jahn–Teller Phase Transitions

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Jahn–Teller structural phase transitions are examined using a simple Ginzburg–Landau approach. The order parameter representing orbital order is complex for the case of doubly degenerate d-orbital states. We find tricritical behavior in ferromagnet-like orbital order under applied uniaxial stress and in antiferromagnet-like orbital order with varying the strength of the interaction between the two sublattices.

KEYWORDS: Jahn–Teller effect, Ginzburg–Landau theory, structural phase transition, orbital order

A representative microscopic origin of structural phase transitions occurring without large-scale composition changes is a coupling between electronic orbital states and lattice distortions, called the Jahn–Teller coupling.[1–6] For example, one electron occupies an electronic state spanned by doubly degenerate d-orbital states around Cu^{2+} or Mn^{3+} in the undistorted crystal structure. Such orbital states can order cooperatively at low temperatures, leading to a cubic-to-tetragonal transformation. Kanamori introduced pseudo-Pauli spin matrices $\hat{\sigma}_{nx}$ and $\hat{\sigma}_{nz}$ operating on electronic states at site n and expressed the orbit-lattice interaction energy in the form[3]

$$\mathcal{H}_{\rm JT} = g_{\rm K} \sum_n (\hat{\sigma}_{nz} Q_{n3} + \hat{\sigma}_{nx} Q_{n2}), \tag{1}$$

where Q_{n2} and Q_{n3} represent appropriate linear combinations of atomic displacements at site n. Their acoustic parts may be equated to e_2 and e_3, to be defined in (2) below. Then, an orbital order represented by $\langle \hat{\sigma}_{nz} \rangle \propto \cos\varphi$ and $\langle \hat{\sigma}_{nz} \rangle \propto \sin\varphi$ at low temperatures gives rise to a cubic-to-tetragonal structural change represented by $\langle Q_{n3} \rangle = \eta_0 \cos\varphi$ and $\langle Q_{n2} \rangle = \eta_0 \sin\varphi$. The tetragonal distortion is along the x, y, and z axes for $\varphi = 2\pi/3$, $-2\pi/3$, and 0, respectively. Hence, the orbital order is the order parameter of the phase transition in this problem.

We also note that the conventional continuum theory of proper martensitic transition[7–10] involves the strains only, including anharmonic terms. However, it is unclear under what conditions this approach is justified. The aim of this letter is hence to set up a Ginzburg–Landau theory for the orbital order parameter coupled to the elastic strains in the Jahn–Teller systems. This is analogous to a similar Ginzburg–Landau theory for binary alloys by the present author.[11–13] Such a coarse-grained theory can have a higher predicting power for complex phase transition behavior than microscopic theories and is indispensable for the study of phase ordering kinetics.[10–14]

We define the diagonal strains as

$$e_1 = \nabla_x u_x + \nabla_y u_y + \nabla_z u_z,$$
$$e_2 = \nabla_x u_x - \nabla_y u_y,$$
$$e_3 = \frac{1}{\sqrt{3}} (2\nabla_z u_z - \nabla_x u_x - \nabla_y u_y), \tag{2}$$

where ∇_j $(j = x, y, z)$ are the space derivatives. The off-diagonal components are written as

$$e_4 = \nabla_x u_y + \nabla_y u_x, \quad e_5 = \nabla_y u_z + \nabla_z u_y, \quad e_6 = \nabla_z u_x + \nabla_x u_z. \tag{3}$$

In the bilinear order, the elastic energy density is expressed as

$$f_{\rm el} = \frac{K}{2} e_1^2 + \frac{C_0'}{2} (e_2^2 + e_3^2) + \frac{C_{44}}{2} (e_4^2 + e_5^2 + e_6^2), \tag{4}$$

where K is the bulk modulus, C_{44} is the shear modulus, and C_0' is the background value of the modulus C' for the tetragonal strains. We shall see that C' is an effective modulus smaller than C_0'.

Before defining the order parameter, we give some discussion on the Jahn–Teller interaction for doubly degenerate d-orbital states,[3] whose wave functions are represented as linear combinations of the wave functions proportional to $2z^2 - x^2 - y^2$ and $x^2 - y^2$. The index n denoting the lattice site is dropped here for simplicity. The orbital states proportional to $2x^2 - y^2 - z^2$, $2y^2 - z^2 - x^2$, and $2z^2 - x^2 - y^2$ are written as $|x^2\rangle$, $|y^2\rangle$, and $|z^2\rangle$, respectively, while those with wave functions proportional to $x^2 - y^2$, $y^2 - z^2$, and $z^2 - x^2$, are written as $|x^2 - y^2\rangle$, $|y^2 - z^2\rangle$, and $|z^2 - x^2\rangle$, respectively. As orthogonal, complete bases, we define

$$|1\rangle = |x^2 - y^2\rangle = \frac{1}{\sqrt{3}} (|x^2\rangle - |y^2\rangle), \quad |2\rangle = |z^2\rangle. \tag{5}$$

Because the electronic orbit is stretched in the x, y, and z axes in the states $|x^2\rangle$, $|y^2\rangle$, and $|z^2\rangle$, respectively, the orbit-lattice coupling energy at each site in cubic solids should be of the form,

$$H_{\rm JT} = -\bar{g}_{\rm K} \big[Q_x |x^2\rangle\langle x^2| + Q_y |y^2\rangle\langle y^2| + Q_z |z^2\rangle\langle z^2| \big], \tag{6}$$

in the bra and ket representation of quantum mechanics, where $\bar{g}_{\rm K}$ is a positive coupling constant and Q_j represents the atomic displacements whose acoustic parts are $\nabla_j u_j$. The sum of $H_{\rm JT}$ over all the lattice sites becomes (1) with $g_{\rm K} = \sqrt{3}\bar{g}_{\rm K}/4$, $Q_2 = Q_x - Q_y \propto e_2$, and $Q_3 = (2Q_z - Q_x - Q_y)/\sqrt{3} \propto e_3$, if we neglect a constant

term proportional to $\sum_j Q_j$. The pseudo-Pauli matrices are defined at each lattice site and are expressed as $\hat{\sigma}_z = |1\rangle\langle 1| - |2\rangle\langle 2|$ and $\hat{\sigma}_x = |1\rangle\langle 2| + |2\rangle\langle 1|$.

First, we consider ferromagnet-like orbital order in spinel-type crystals such as $CuFe_2O_4$ and Mn_3O_4. We introduce a coarse-grained, two-component order parameter (ψ_1, ψ_2) as

$$\psi_1(\boldsymbol{r}) = -\ell^{-d}\sum_n \langle\hat{\sigma}_{nz}\rangle, \quad \psi_2(\boldsymbol{r}) = -\ell^{-d}\sum_n \langle\hat{\sigma}_{nx}\rangle, \tag{7}$$

where the summation is over lattice points in an appropriately defined cell with a lattice constant ℓ longer than the original lattice constant a, and $\boldsymbol{r}$ is a representative position within the new cell. The average $\langle\cdots\rangle$ is taken doubly over the quantum and thermal fluctuations. It is convenient to define the complex order parameter by $\psi = \psi_1 + \mathrm{i}\psi_2 = |\psi|\exp(\mathrm{i}\varphi)$. The free-energy density $f_{\rm JT}$ representing the orbit-lattice coupling reads

$$f_{\rm JT} = -g(e_3\psi_1 + e_2\psi_2) = -g\,\mathrm{Re}[(e_3 + \mathrm{i}e_2)\psi^*]. \tag{8}$$

Here, it is informative to consider how ψ is changed with respect to a rotation of the reference frame by $\pi/2$. The rotation about the x axis is equivalent to the replacement, $y \to z$, $z \to -y$, with x unchanged, which yields the transformation $\psi \to \exp(-2\pi/3)\psi^*$. The rotations about the y and z axes yield $\psi \to \exp(2\pi/3)\psi^*$ and $\psi \to \psi^*$, respectively. The complex strain defined by $e_3 + \mathrm{i}e_2$ is also changed in the same manner and $f_{\rm JT}$ is invariant for these rotations. Generally, in the presence of the cubic crystal symmetry in the disordered phase, the total free energy density $f = f(\psi, \boldsymbol{u})$ for ψ and the elastic strains should be invariant with respect to these rotations. We propose the Landau expansion $f = f_0(\psi) + f_{\rm JT} + f_{\rm el}$ close to the transition with

$$f_0(\psi) = \frac{r_0}{2}|\psi|^2 + \frac{u}{4}|\psi|^4 + \frac{C}{2}|\nabla\psi|^2 - B_0(\psi_1^3 - 3\psi_1\psi_2^2), \tag{9}$$

where u, C, and B_0 are positive constants. We also assume that r_0 depends on the temperature T as $r_0 = A_0(T - T_0)$, where A_0 is a positive constant and T_0 is a constant temperature. Due to the last third-order term, the ordered states with $\varphi = 0, 2\pi/3$, and $-2\pi/3$ are favored, and correspond to the tetragonally distorted states with the symmetry axes along the z, x, and y axes, respectively.

The real parts of $\psi^{3-k}(e_3 + \mathrm{i}e_2)^k$ $(k = 0, 1, 2, 3)$ constitute four third-order invariants. More explicitly, they are written as $I_{30} = \psi_1^3 - 3\psi_1\psi_2^2 = M^3\cos 3\varphi$, $I_{31} = (\psi_1^2 - \psi_2^2)e_3 - 2\psi_1\psi_2 e_2$, $I_{32} = \psi_1(e_3^2 - e_2^2) - 2\psi_2 e_2 e_3$, and $I_{33} = e_3^3 - 3e_3e_2^2$. We may assume a third-order term expressed as $\sum_{j=0}^{3} B_j I_{3j}$ in the free energy density, where B_j are the coefficients. In (9), for simplicity, we retain a third-order term proportional to I_{30} only. The invariant I_{33} has been of crucial importance in the conventional nonlinear strain theory.[7–9)]

If small e_3 is induced in the disordered phase, we have $\psi \cong ge_3/r_0$ in the static limit. Elimination of ψ yields the effective elastic modulus C' for the tetragonal strains,

$$C' = C_0' - g^2/r_0. \tag{10}$$

We next consider a tetragonal state stretched along the z axis under the stress-free condition, where $\psi_1 = M > 0$ and $\psi_2 = 0$. By setting $e_3 = gM/C_0'$, we obtain the equation of state, $r + uM^2 - 3B_0M = 0$, with

$$r = r_0 - g^2/C_0' = A_0(T - T_{\rm c0}), \tag{11}$$

where $T_{\rm c0} = T_0 + g^2/A_0C_0'$. At $r = r_{\rm tr}$, a first-order phase transition occurs, and M and e_3 change from 0 to $M_{\rm tr}$ and $e_{\rm tr}$, respectively, where

$$r_{\rm tr} = 2B_0^2/u, \quad M_{\rm tr} = 2B_0/u, \quad e_{\rm tr} = 2gB_0/C_0'u. \tag{12}$$

For $r < r_{\rm tr}$, the ordered phase is stable and M is expressed as

$$M = \frac{3}{4}M_{\rm tr}\Big[1 + \sqrt{1 - 8r/9r_{\rm tr}}\Big]. \tag{13}$$

The elastic modulus C' in (10) immediately *above* the transition is expressed as $C_0'w/(1 + w)$ with

$$w = 2C_0'B_0^2/g^2u. \tag{14}$$

In terms of the elastic properties, w represents weakness of the first-order phase transition.

The fluctuation effect is much more complicated in ordered states than in disordered states due to the two-component nature of ψ. The second-order contributions to f from the fluctuations of ψ and the strains are written as

$$\delta f = \frac{1}{2}r_{\rm L}(\delta\psi_1)^2 + \frac{1}{2}r_{\rm T}\psi_2^2 + \frac{C}{2}|\nabla\psi|^2 + \delta f_{\rm el}, \tag{15}$$

where $\delta\psi_1 = \psi_1 - M$, and $f_{\rm el}$ is obtained from (4) if e_3 and e_2 are replaced by $e_3 - g\psi_1/C_0'$ and $e_2 - g\psi_2/C_0'$, respectively. The longitudinal and transverse inverse susceptibilities, $r_{\rm L}$ and $r_{\rm T}$, are given by

$$r_{\rm L} = 2uM^2 - 3B_0M, \quad r_{\rm T} = 6B_0M. \tag{16}$$

It is worth noting that $r_{\rm T}$ is positive owing to the third-order term, while it vanishes in the (isotropic) many-component spin systems.[14,15)] Considering homogeneous deviations, we eliminate $\delta\psi_1$ and ψ_2 to obtain effective elastic moduli, $C_{\rm L}'$ and $C_{\rm T}'$, for the strains e_3 and e_2, respectively. In terms of w in (14) and $m \equiv M/M_{\rm tr}$, they are expressed as

$$\frac{C_{\rm L}'}{C_0'} = \frac{w(4m^2 - 3m)}{1 + w(4m^2 - 3m)}, \quad \frac{C_{\rm T}'}{C_0'} = \frac{9wm}{1 + 9wm}. \tag{17}$$

In Fig. 1, we display these elastic moduli near the transition for $w = 0.04$. The modulus $C_{\rm L}'$ below the transition is continuously connected to C' in (11) above the transition and is smaller (larger) than $C_{\rm T}'$ for r larger (smaller) than $-(45/16 + 3\sqrt{45/32})r_{\rm tr}$.

For inhomogeneous deviations, we express the strains in terms of the Fourier components assuming the mechanical equilibrium.[6,11,14)] Then, the space integral of $\delta f_{\rm el}$ is expressed as

$$\int \mathrm{d}\boldsymbol{r}\delta f_{\rm el} = \frac{1}{2}\sum_{\alpha,\beta=1,2}\sum_{\boldsymbol{k}} A_{\alpha\beta}(\hat{\boldsymbol{k}})\psi_{\alpha\boldsymbol{k}}\psi_{\beta\boldsymbol{k}}^*, \tag{18}$$

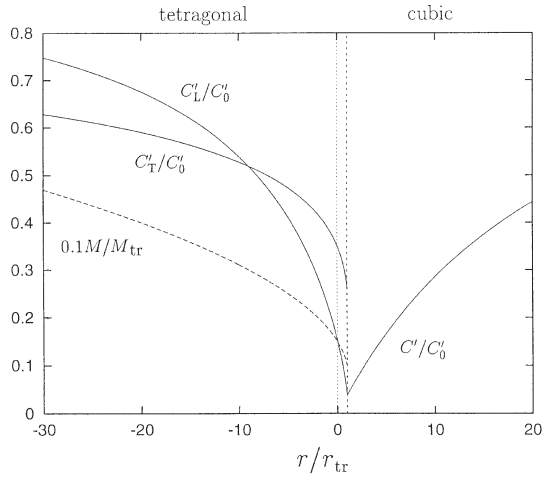

Fig. 1. Normalized elastic moduli for tetragonal strains vs normalized reduced temperature ($\propto T - T_{c0}$) for $w = 0.04$. Here, C'_L and C'_T are the moduli for e_3 and e_2, respectively, in the tetragonal phase stretched along the z axis. Normalized order parameter M/M_{tr} (divided by 10) is also plotted.

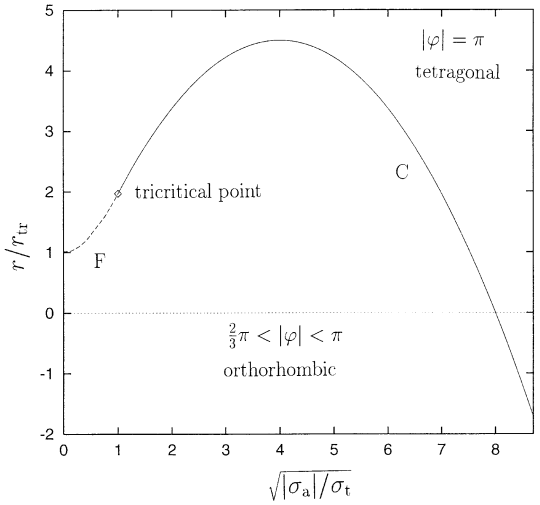

Fig. 2. Phase diagram under uniaxial compression ($\sigma_a < 0$) with the free energy density (19). The horizontal and vertical axes represent $|\sigma_a/\sigma_t|^{1/2}$ and r/r_{tr}, respectively. The line of the first-order phase transition (dashed line) starts from the point where $r = r_{tr}$ and $\sigma_a = 0$, and meets the critical line (solid line) at the tricritical point, where $r = r_{tr}$ and $\sigma_a = -\sigma_t$. Above the curves, we have a tetragonal phase ($\varphi = \pm\pi$), while below them, we have two stable orthorhombic variants ($2\pi/3 < |\varphi| < \pi$) as expressed by (21).

where $A_{\alpha\beta}(\hat{\boldsymbol{k}})$ is a non-negative-definite matrix dependent on the direction of the wave vector, $\hat{\boldsymbol{k}} \equiv k^{-1}\boldsymbol{k}$. Note that we include $g^2|\delta\psi|^2/2C'_0$ in the first two terms in (15). For $\hat{\boldsymbol{k}}$ along [110], we obtain $A_{22} = A_{12} = 0$ and hence $\lim_{k\to 0}\langle|\psi_{2\boldsymbol{k}}|^2\rangle = 1/r_T$. As a result, the velocity of the transverse sound propagating along [110] and polarized along [1$\bar{1}$0] is given by $c_t[110] = (C'_T/\rho)^{1/2}$, ρ being the mass density. In passing, let us consider the limit $B_0 \to 0$ with $r(<0)$ and g fixed. Here, $C'_T \to 0$, while $C'_L/C'_0 \to 1 - 1/[1 + 2C'_0|r|/g^2]$. In accordance with this result, Pytte[5] found that $c_t[110]$ vanishes for all temperatures below the transition in the absence of the third-order terms.

Furthermore, some new effects can be predicted if a uniaxial stress σ_a is applied along the z axis while the stress is free in the xy plane. Setting $C'_0 e_2 = g\psi_2$ and $Ke_1 = \sqrt{3}(C'_0 e_3 - g\psi_1) = \sigma_a$, we minimize

$$\tilde{f} = \frac{r}{2}|\psi|^2 + \frac{u}{4}|\psi|^4 - B_0(\psi_1^3 - 3\psi_1\psi_2^2) - \frac{g}{\sqrt{3}C'_0}\sigma_a\psi_1. \tag{19}$$

In the case of stretching $\sigma_a > 0$, the tetragonal variant with $\psi_1 > 0$ and $\psi_2 = 0$ is favored below the transition. However, the phase behavior becomes more interesting in the case of compression $\sigma_a < 0$, so we limit ourselves to this case. As shown in Fig. 2, the temperature-stress plane is divided into two regions by a line of first-phase transition (line F) and a critical line (line C). These two lines meet at a tricritical point, where r, $|\sigma_a|$, and $M = |\psi_1|$ assume the following tricritical values,

$$r_t = \frac{63}{32}r_{tr}, \quad \sigma_t = \frac{81\sqrt{3}}{64}wC'_0 e_{tr}, \quad M_t = \frac{3}{8}M_{tr}, \tag{20}$$

respectively. On line C, we obtain $4r/9r_{tr} = |\sigma_a/\sigma_t|^{1/2} - |\sigma_a/8\sigma_t|$ and $M/M_{tr} = (3/8)|\sigma_a/\sigma_t|^{1/2}$. Above the lines, the stable phase consists of a single tetragonal state expressed as $\psi_1 = -M < 0$ and $\psi_2 = 0$. Below the lines, there are two stable orthorhombic variants expressed as

$$\psi_1 = -M\cos\varphi_a, \quad \psi_2 = \pm M\sin\varphi_a, \tag{21}$$

where the angle $\varphi_a = \pi \mp \varphi$ is in the range $0 < \varphi_a < \pi/3$ and satisfies $\sin^2\varphi_a = (3/4)[1 - |\sigma_a/\sigma_t|/(8M/3M_{tr})^2]$. Here, $\varphi_a \to \pi/3$ as $\sigma_a \to 0$ for $r < r_{tr}$, $\varphi_a \to 0$ as line C is approached, and φ_a remains nonvanishing as line F is approached from below. The modulus C'_T for the strain e_2 tends to zero as line C is approached both from above and below. Near the critical line, ψ_1 is less singular than ψ_2 and we may set $\psi_1 = -M + c_2\psi_2^2 + c_4\psi_2^4 + \cdots$ from $\partial\tilde{f}/\partial\psi_1 = 0$. The Landau expansion for the primary order parameter ψ_2 then becomes $\tilde{f} = a_2\psi_2^2 + a_4\psi_2^4 + a_6\psi_2^6 + \cdots$, where elimination of ψ_1 yields the sixth-order term with $a_6 > 0$. We have $a_2 = 0$ and $a_4 > 0$ on line C, while a_2 and a_4 are written as linear combinations of $r - r_t$ and $|\sigma_a| - \sigma_t$ near the tricritical point.[16] The maximum point on line C is given by $r = r_{max} = 9r_{tr}/2$ and $|\sigma_a| = \sigma_{max} = 16\sigma_t$. If r is fixed in the region $r_{tr} < r < r_{max}$, the orthorhombic phase is realized in a window region of $|\sigma_a|$. This region is expressed as $a_2 \cong r - r_{max} + (9/8)r_{tr}(|\sigma_a|/\sigma_{max} - 1)^2 < 0$ slightly below the maximum point.

In MnF_3, the orbital order occurs alternatively in two sublattices with a symmetry axis along one of the cubic axes (say, the z axis). In such cases, it is convenient to introduce the two complex variables, $\psi_A = \psi_{A1} + i\psi_{A2}$ and $\psi_B = \psi_{B1} + i\psi_{B2}$, for sublattices, A and B.[3] Then, we have the antiferromagnet-like order parameter $\zeta = \psi_A - \psi_B$ in addition to the ferromagnet-like order parameter $\psi = \psi_A + \psi_B$. The orbital order in MnF_3 is represented by $\psi = -2M\sin\phi$ and $\zeta = \pm 2iM\cos\phi$, where ϕ is the canting angle of the sublattice order. The solid is uniaxially deformed along the z axis.

In the linear order with respect to the strains, the orbit-lattice coupling may be written as

$$f_{\rm JT} = -g\,{\rm Re}[(e_3 - {\rm i}e_2)\psi] - g'\,{\rm Re}[(e_3 + {\rm i}e_2)\zeta^2]. \quad (22)$$

The free energy density f for ψ, ζ, and the strains is even with respect to ζ and is of the form,

$$f = f_0(\psi_{\rm A}) + f_0(\psi_{\rm B}) + f_{\rm JT} + f_{\rm el} + \frac{1}{4}r'(|\psi|^2 - |\zeta|^2) - B'\,{\rm Re}[\psi_{\rm A}\psi_{\rm B}\psi] + u'(|\psi|^2 - |\zeta|^2)^2, \quad (23)$$

where f_0 is given by (9) and the last three terms arise from the interaction between the two sublattices since $|\psi|^2 - |\zeta|^2 = 4\,{\rm Re}[\psi_{\rm A}\psi_{\rm B}^*]$. In the bilinear order, we have $\delta f = (r + r_1)|\psi|^2/4 + (r - r_1)|\zeta|^2/4$ after elimination of the tetragonal strains, where

$$r = r_0 - g^2/C_0', \quad r_1 = r' - g^2/C_0'. \quad (24)$$

Thus, the disordered phase is linearly unstable for $r < -r_1$ with respect to the fluctuations of ψ and for $r < r_1$ with respect to those of ζ. However, first-order phase transitions are induced by the cubic terms ($\propto B_0,\ B'$) and the phase behavior becomes very complicated for general r', B', u', g', and applied stress.

Here, we consider the effect of r' assuming $B' = u' = g' = 0$ under the stress-free condition.[3] Let the phase angles of $\psi_{\rm A}$ and $\psi_{\rm B}$ be $\varphi_{\rm A}$ and $\varphi_{\rm B}$, respectively. Then the angle-dependent part in f is written as $-B_0[|\psi_{\rm A}|^3\cos(3\varphi_{\rm A}) + |\psi_{\rm B}|^3\cos(3\varphi_{\rm B})] + r_1|\psi_{\rm A}\psi_{\rm B}|\cos(\varphi_{\rm A} - \varphi_{\rm B})$. For $r_1 < 0$, we find $\varphi_{\rm A} = \varphi_{\rm B}$ leading to ferromagnet-like order ($\zeta = 0$), while for $r_1 > 0$, the skewed state with $\varphi_{\rm A} = -\varphi_{\rm B}$ and $\cos 2\varphi_{\rm A} < 0$ minimizes the free energy. For this antiferromagnet-like order, we set $\psi_{\rm A} = \psi_{\rm B}^* = {\rm i}M\exp({\rm i}\phi)$ in (23) to obtain

$$f = rM^2 + \frac{u}{2}M^4 - 2B_0M^3\sin 3\phi - r_1M^2\cos 2\phi. \quad (25)$$

In Fig. 3, we show the phase diagram in the plane of r and r_1. A line of first-order phase transition starts from the point where $r = r_{\rm tr}$ and $r_1 = 0$, and ends at a tricritical point where $r = r_1 = 9r_{\rm tr}/2$.[2] On this line we have $2r/r_{\rm tr} = 1 + (2r_1/3r_{\rm tr} + 1)^{3/2}$, $4\sin^2\phi = 2 - (2r_1/3r_{\rm tr} + 1)^{1/2}$, and $M = 2\sin 3\phi$. For $r_1 > 9r_{\rm tr}/2$, however, the phase transition becomes second-order with a critical line given by $r = r_1$ and the primary order parameter is ${\rm Im}\,\psi_{\rm A} = {\rm Im}\,\zeta/2$. The system is in the disordered phase ($M = 0$) above the two lines. For small r_1 or far below the critical line, the effect of r_1 becomes weak and $\phi \cong \pi/6$.

We have presented a Ginzburg–Landau theory for relatively simple Jahn–Teller models to find rich phase transition behavior. The relationship between the nonlinear strain theory[7–10] and the present theory (with the true order parameter retained) is not still clear. However, when the discontinuity of the first-order phase transition is weak ($w \ll 1$), the linear relation $\psi \cong g(e_3 + {\rm i}e_2)/r_0$ holds and the two schemes yield essentially the same results near the transition ($|r| \ll g^2/C_0'$). It is also of great interest how the phase ordering kinetics proceeds under the influence of the nonlocal interaction (18) and the cubic terms in the free energy.[10,12,13]

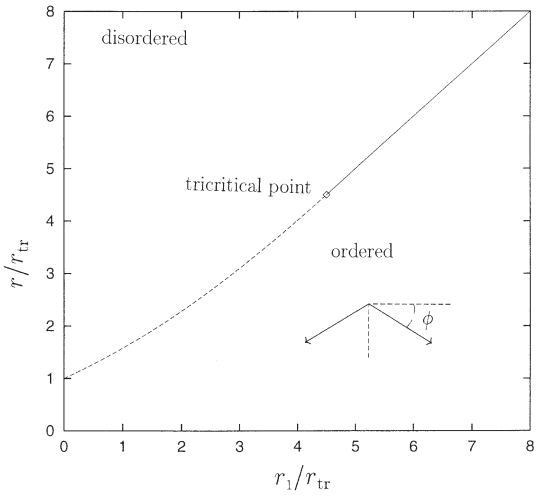

Fig. 3. Phase diagram of a Jahn–Teller system modelled using (25) with positive r_1 favoring canted antiferromagnet-like orbital order. The system is in the disordered phase ($M = 0$) above the line of first-order phase transition (dashed line) and the critical line (solid line).

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