Stripe Structures and the Berry-Phase Connection: Concept of Geometric Energy

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(June 9, 1999)

Electronic states of an e_g electron are calculated in the system composed of two MnO_6 octahedra with the inclusion of the Berry phase acquired by parallel transport. Based on this calculation, a comment is made on the controversy between "Wigner-crystal" and "paired-stripe" models for the the insulating charge-ordered manganese oxides.

I. INTRODUCTION

In quantum mechanics the Hamiltonian H alone does not determine energy eigenstates of the system. We need information on the wavefunction φ which includes the boundary condition in general and for the case of a many-electron system, antisymmetry due to the Fermi-Dirac statistics. The antisymmetry may be regarded as addition of the phase factor $e^{i\pi}$ to φ in the interchange of any pair of electrons of the same spin. Note that this factor $e^{i\pi}$ brings about an important concept of "the exchange interaction" J which is just the energy difference between singlet ($S = 0$) and triplet ($S = 1$) states in an interacting two-electron system.

Addition of the phase factor to φ also occurs in the course of Berry-phase connection; the Berry phase is a geometric phase in φ acquired by parallel transport. [1] On the analogy of J, we propose a new concept of "the geometric energy" by considering an energy difference between the states characterized by different winding numbers w (the Chern integers) associated with the topological invariant in parallel transport.

In order to substantiate the above proposal, we calculate the geometric energy in this paper in the simplest possible system, namely, a system consisting of a single electron moving back and forth between two Jahn-Teller(JT) centers. In Sec. 2 we specify our system precisely by writing H and a prescription for parallel transport. The lowest-energy state for each w is given in Sec. 3 and Sec. 4 deals with our perspective into the stripe structures observed in $La_{1-x}Ca_xMnO_3$ with $x \geq 0.5$. [2]

II. TWO-JAHN-TELLER-CENTER SYSTEM AND PARALLEL TRANSPORT

Consider an e_g electron in the system composed of two $Mn^{4+}O_6$ octahedra with one oxygen ion common to both octahedra to provide a transfer path with t the transfer integral for the e_g electron in z-direction. We assume that the spins of t_{2g} electrons are aligned in one direction. Because of the strong Hund's rule coupling, spin of the e_g electron also aligns in the same direction. In this sense, there are no spin degrees of freedom and we may write H in second quantization as

$$H = -t(b_1^+ b_2 + b_2^+ b_1) + E_{\rm JT} \sum_{j=1,2} \Big[2\mathbf{d}_j^+ (q_j^{(2)} \tau_x + q_j^{(3)} \tau_z) \mathbf{d}_j + {q_j^{(2)}}^2 + {q_j^{(3)}}^2 \Big], \quad (1)$$

with the Pauli matrices, τ_x and τ_z, and $\mathbf{d}_j^+ \equiv (a_j^+, b_j^+)$, where a_j^+ and b_j^+ are, respectively, creation operators for electrons in $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals at Mn site $j(= 1$ or $2)$, $q_j^{(2)}$ and $q_j^{(3)}$ are dimensionless vibrational variables representing, respectively, $(\delta X_j - \delta Y_j)/\sqrt{2}$ and $(2\delta Z_j - \delta X_j - \delta Y_j)/\sqrt{6}$ modes around site j, and $E_{\rm JT}$ is the static JT energy. Since we shall treat static distortions in the adiabatic approximation, the kinetic-energy term of ions is not considered in H.

The Berry-phase effect at site j is best included by writing vibrational modes in polar coordinates as $q_j^{(2)} = q_j \sin\theta_j$ and $q_j^{(3)} = q_j \cos\theta_j$. Using θ_j, we transform the operators a_j and b_j into $\tilde{a}_j$ and $\tilde{b}_j$

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as $\tilde{a}_j = e^{i\theta_j/2}[a_j \cos(\theta_j/2) + b_j \sin(\theta_j/2)]$ and $\tilde{b}_j = e^{i\theta_j/2}[-a_j \sin(\theta_j/2) + b_j \cos(\theta_j/2)]$. In this representation, the second term (the site-energy term) in (1) is diagonalized. The molecular Aharonov-Bohm effect manifests itself in the phase factor $e^{i\theta_j/2}$ at each site j at which topology of the singularity is understood as a double-plane feature.

The transfer between the two sites connects θ_1 with θ_2. We make this connection by parallel transport, by which we mean that if we start with a state representing an electron at site 1, φ_1, we make the state evolve from φ_1 in the direction orthogonal to it, namely, along the direction of a state representing an electron at site 2, φ_2. This parallel transport indicates that an e_g electron picks up only a constant phase θ_0 by a trip from site 1 to site 2, namely, $\theta_2 = \theta_1 + \theta_0$. A further trip in the same direction from site 2 results in the total phase as $\theta_2 + \theta_0 = \theta_1 + 2\theta_0$. Now in general, a two-site system is equivalent to a corresponding periodic lattice system with periodicity two. Thus the phase $\theta_1 + 2\theta_0$ should represent the same physics at site 1 at which the phase is θ_1. This reasoning leads us to the conclusion that $2\theta_0$ should be equal to $2\pi w$ with an integer w. If we make a more general argument on topology, [3] it turns out that w is nothing but the winding number, a topologically conserved quantity which is useful to specify an eigenstate of the system. Note that w describes a geometric structure as to how the double-plane structure at each site is interconnected.

III. GEOMETRIC ENERGY

Let us calculate the lowest single-electron energy for each $w(= 0 \text{ or } 1)$, $E_0^{(w)}$. We adopt a variational procedure by considering a wavefunction $|\Psi_0^{(w)}\rangle$ as

$$|\Psi_0^{(w)}\rangle = \sum_{j=1,2} e^{i\theta_j/2}(\alpha_j^{(w)} \tilde{a}_j^+ + \beta_j^{(w)} \tilde{b}_j^+)|\text{vac}\rangle \otimes |q_1 = \bar{q}_1, q_2 = \bar{q}_2\rangle\,, \tag{2}$$

where $\alpha_j^{(w)}$ and $\beta_j^{(w)}$ are real numbers, $|\text{vac}\rangle$ is the vacuum for electron operators, and $|q_1 = \bar{q}_1, q_2 = \bar{q}_2\rangle$ is the phonon wavefunction representing the δ-function-like distribution of q_j around $\bar{q}_j$. We have optimized all the parameters involved in the problem except for θ_2 (which is related to θ_1 through $\theta_1 + \pi w$) in order to make $\langle\Psi_0^{(w)}|H|\Psi_0^{(w)}\rangle$ minimum under the condition of $\langle\Psi_0^{(w)}|\Psi_0^{(w)}\rangle = 1$ for given t and $E_{\rm JT}$. We have found that the lowest energy is obtained at $\theta_1 = 0$ irrespective of w. This is the condition for the e_g-electron orbital to polarize in the transfer direction. (If we start with a different configuration, say two sites in x-direction, we obtain completely the same physical results with the interchange of the role of z-axis with that of x.) However, $|\Psi_0^{(w)}\rangle$ is different for different w in an interesting way. For $w = 0$, we obtain

$$|\Psi_0^{(0)}\rangle = (\beta_1^{(0)} b_1^+ + \beta_2^{(0)} b_2^+)|\text{vac}\rangle \otimes |q_1 = {\beta_1^{(0)}}^2, q_2 = {\beta_2^{(0)}}^2\rangle\,, \tag{3}$$

with the coefficients $\beta_j^{(0)}$ shown in Fig. 1(a) as a function of $E_{\rm JT}$. As long as $E_{\rm JT}$ is smaller than t, symmetry exists between the sites, but for larger $E_{\rm JT}$ the site-symmetry is broken; an e_g electron tends to localize in site 1. (Of course, the state with the interchange of the sites is possible, but there is no overlap between these two states because of the orthogonality of phonon wavefunctions.)

For $w = 1$, on the other hand, we obtain

$$|\Psi_0^{(1)}\rangle = (\beta_1^{(1)} b_1^+ + \alpha_2^{(1)} b_2^+)|\text{vac}\rangle \otimes |q_1 = {\beta_1^{(1)}}^2, q_2 = 0\rangle\,, \tag{4}$$

with the coefficients $\beta_1^{(1)}$ and $\alpha_2^{(1)}$ shown in Fig. 1(b). In this case, the site-symmetry is broken even for an infinitesimally small positive value for $E_{\rm JT}$ due to the fact that the lattice at site 2 never deforms, because with this winding number, an e_g electron at site 1 in the lower adiabatic potential plane is parallel-transported to the upper adiabatic potential plane at site 2. In this sense, $w = 1$ describes "the inter-potential-plane connection", while $w = 0$ "the intra-potential-plane connection".

We plot the corresponding energies $E_0^{(w)}$ as a function of $E_{\rm JT}$ in Fig. 2 in which we give an analytic expression for $E_0^{(0)}$. (We can give $E_0^{(1)}$ only numerically.) The term $-t^2/2E_{\rm JT}$ clearly indicates the hopping nature of an electron localized by the JT stabilization energy $-E_{\rm JT}$ for $E_{\rm JT} > t$. We find that $E_0^{(0)}$ is always lower than $E_0^{(1)}$. The geometric energy, $E_0^{(1)} - E_0^{(0)}$, is negligibly small in both large- and small-$E_{\rm JT}$ regions, but it becomes as large as $0.1t$ for $E_{\rm JT} \approx t$.

Quite a similar situation occurs in the J-problem in a two-electron system in which the conserved quantity is the total spin S; the energy of the singlet state $E_0^{S=0}$ is always lower than that of the triplet one $E_0^{S=1}$. Note, however, that application of external magnetic fields can compensate the energy difference to stabilize the triplet state. Analogously, we may stabilize $|\Psi_0^{(1)}\rangle$ by compensating the geometric energy with application of external stresses to the system in z-direction, because the state $|\Psi_0^{(1)}\rangle$ is less distorted in the direction than $|\Psi_0^{(0)}\rangle$.

IV. DISCUSSION ON PAIRED-STRIPE STRUCTURES

Considerable accumulation of experimental data has been made as for the crystal structures of the insulating $La_{1-x}Ca_xMnO_3$ with $x \geq 0.5$. Everyone agrees the

existence of the charge and orbital ordering in these compounds, but a controversy continues about their detailed crystallographic superstructures; some claim the "paired-stripe" model [2] and others the "Wigner-crystal" model. [4]

In order to shed light on the argument, we have extended our calculation from the two-JT-center problem to a two-dimensional lattice system. [3] The key idea is to identify a zigzag conducting path of e_g electrons along which we define the winding number w. We have found that the state corresponding to the Wigner-crystal model provides the lower energy than the paired-stripe state, though the energy difference is quite small. This result is obtained without any consideration of the long-range Coulomb interaction V. We have included the effect of V recently and found that further stabilization is obtained for the Wigner-crystal state over the paired-stripe one. Thus, in terms of energies, it seems to be certain that the former state is favorable.

However, we have investigated the electronic state of e_g electrons in the observed paired-stripe structure, evaluated the corresponding winding number w, and found an interesting relation between w and the e_g-electron density specified by x as [3]

$$w = \frac{x}{1-x} = \frac{\text{Number of } \mathrm{Mn}^{4+} \text{ ions}}{\text{Number of } \mathrm{Mn}^{3+} \text{ ions}}. \qquad (5)$$

Since w is an integer, some specific values exist for $x[= w/(1+w)]$ such as 1/2, 2/3, 3/4, etc. This reminds us of the experimentally observed special values of x and the related lever rule. [2] Thus, if the crystal is placed in an "environment" to favor the state with this winding number w, we can expect that the paired-stripe state may be observed. At present, we do not know precisely how we can define the "environment" and this may be an important issue in the future not only from a viewpoint to clarify the crystal structures of $La_{1-x}Ca_xMnO_3$ but also from a fundamental aspect as to the identification of a physical variable to control the winding number or the geometric energy.

ACKNOWLEDGMENTS

YT is supported by the Mitsubishi Foundation as well as the Grand-in-Aid for Scientific Research (C) from the Monbusho of Japan.

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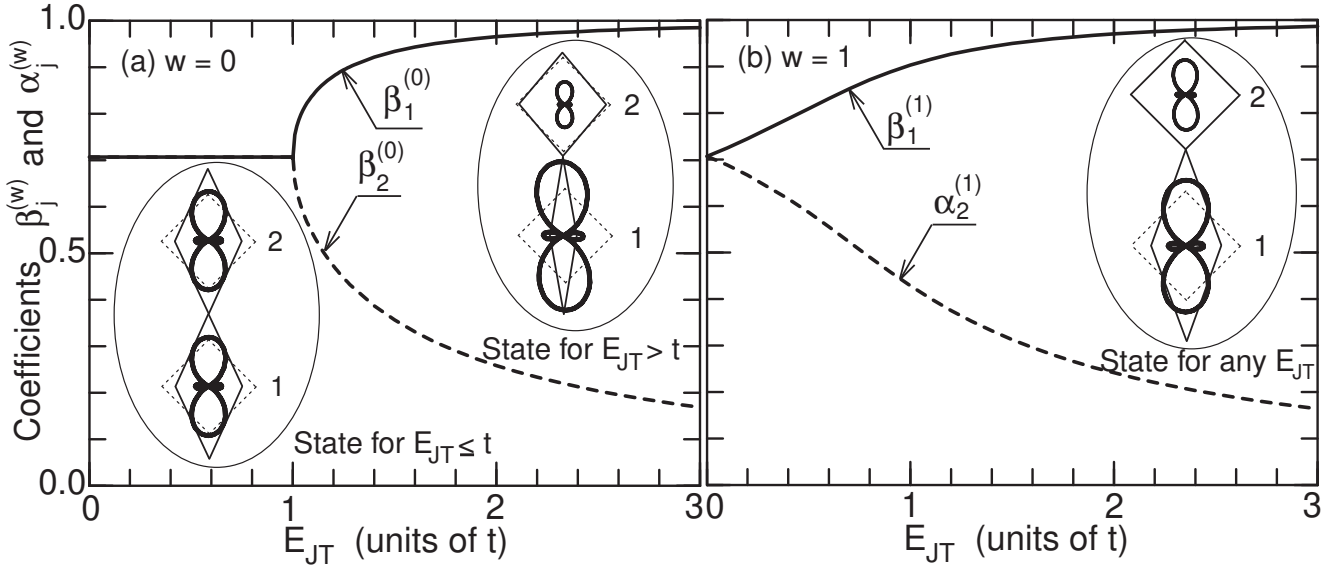

FIG. 1. Coefficients to specify the lowest single-electron wavefunctions plotted as a function of $E_{\rm JT}$ for (a) $w = 0$ and (b) $w = 1$.

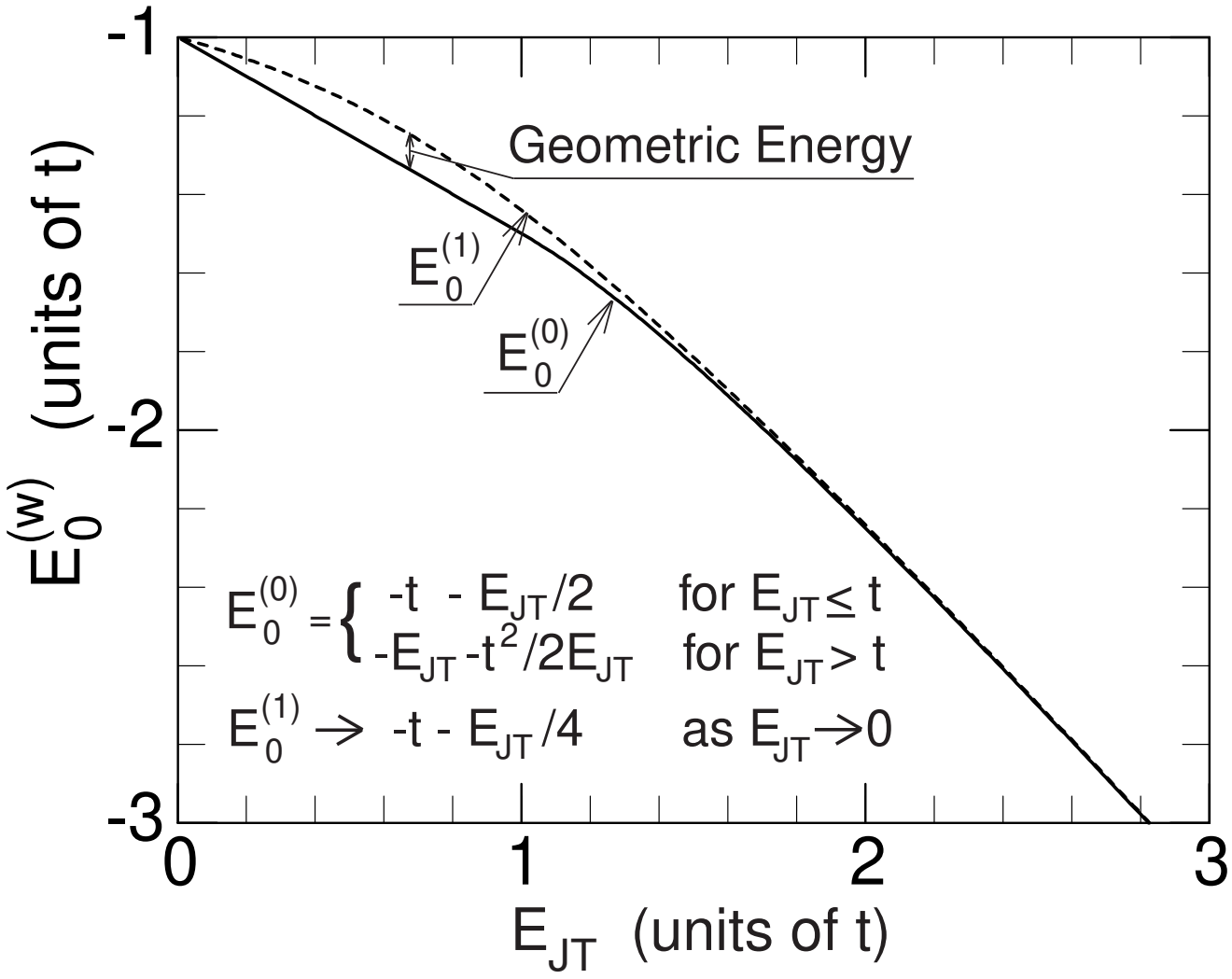

FIG. 2. Lowest single-electron energies as a function of $E_{\rm JT}$ for $w = 0$ and 1.