# Effect of Jahn-Teller coupling on Curie temperature in the double-exchange model

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(Received 22 April 2009; revised manuscript received 22 June 2009; published 15 July 2009)

We consider the two-band double-exchange model for manganites with Jahn-Teller (JT) coupling and explore the suppression of the ferromagnetism because of the JT distortion. The localized spins of the $t_{2g}$ electrons are represented in terms of the Schwinger bosons and two spin-singlet Fermion operators are introduced instead of the $e_g$ electrons' operators. In terms of the new Fermi fields the on-site Hund's interaction is in a diagonal form and one accounts for it exactly. Integrating out the spin-singlet fermions, we derive an effective Heisenberg model for a vector which describes the local orientations of the total magnetization. The exchange constants are different for different space directions and depend on the density $n$ of $e_g$ electrons and JT energy. At zero temperature, with increasing the density of the $e_g$ electrons the system undergoes phase transition from ferromagnetic phase ($0<n<n_c$) to $A$-type antiferromagnetic phase ($n_c<n$). The critical value $n_c$ decreases as JT energy is increased. At finite temperature we calculate the Curie temperature as a function of electron density for different JT energy. The results show that JT coupling strongly suppresses the spin fluctuations and decreases the Curie temperature.

DOI: 10.1103/PhysRevB.80.012403 PACS number(s): 75.47.Lx, 63.20.kd, 71.27.+a, 75.30.Ds

Jahn-Teller (JT) effect is related to systems with degenerated electronic states.[1] The importance of the JT coupling for manganites was first discussed in Ref. 2 and 3 with regard to the colossal magnetoresistance. The most widely studied representatives have chemical formula $Re_{1-x}A_xMnO_3$, where Re is rare earth such as La or Nd and $A$ is a divalent alkali such as Ca or Sr. The important electrons in these compounds are Mn $d$ electrons. They have five degenerate levels.[4] The crystal environment results in a particular splitting of the five $d$ orbitals (*crystal-field spliting*) into two groups: the $e_g$ and $t_{2g}$ states. The electrons from the $e_g$ sector form a doublet while the $t_{2g}$ electrons form a triplet. The population of the $t_{2g}$ electrons remains constant and the Hund rule enforces alignment of the three $t_{2g}$ spins into a $S=3/2$ state. Then, the $t_{2g}$ sector can be replaced by a *localized spin* at each manganese ion, reducing the complexity of the original five orbital model. The only important interaction between the two sectors is the Hund coupling between localized $t_{2g}$ spins and mobile $e_g$ electrons. The oxygens surrounding the manganese ion readjust their locations creating an asymmetry between the different directions. This effectively removes the degeneracy of the $e_g$ electrons' states. The lifting of the degeneracy due to the orbital-lattice interaction is called Jahn-Teller effect.

The interaction between the electrons and phonons is unusually strong and leads to a wide range of striking physical phenomena. Changing the $e_g$ electrons' concentration produces a variety of phases, which may be characterized by their magnetic, transport, and charge-ordering properties.[5] The manganites $La_{1-x}Ca_xMnO_3$ have attracted interest due to their colossal magnetoresistance. The phase boundary between ferromagnetism and paramagnetism, in these materials, also separates a low-temperature metallic phase from a high-temperature insulating phase. At temperatures below Curie temperature $T<T_C$ the resistivity is relatively low and increases as $T$ is increased, whereas at $T>T_C$ the resistivity is very high and (for most $x$) decreases as $T$ is increased. The magnetoresistance for $T\approx T_C$ can be very large.[5]

The double-exchange model with JT coupling is a widely used model for manganites. The procedures followed to obtain the essential features of the model are different: numerical studies,[6,7] dynamical mean-field theory,[8–10] *ab initio* density-functional calculations,[11] and analytical calculations.[8,9,12,13] In spite of the common conclusion that JT coupling suppresses the ferromagnetic state, the results are quite different and do not match the experimental results. For example, the calculated Curie temperatures are two and even three times larger than the experimentally measured. Because of that it is important to formulate theoretical criteria for adequacy of the method of calculation. In our opinion the calculations should be in accordance with the Mermin-Wagner theorem.[14] It claims that at nonzero temperature, a one-dimensional or two-dimensional (2D) isotropic-spin-$S$ Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. We employ a technique of calculation,[15] which captures the essentials of the magnon fluctuations in the theory and for 2D systems one obtains zero Curie temperature, in accordance with Mermin-Wagner theorem. The physics of the ferromagnetic manganites near the Curie temperature is dominated by the magnon fluctuations and it is important to account for them in the best way.

The present paper is focused on the influence of the JT distortion on the ferromagnetism of manganites. To model the manganites we employ the Hamiltonian $H=H_{DE}+H_{el-ph}$. The first term describes the hopping of $e_g$ electrons and the Hund interaction between the spin $\mathbf{s}_i$ of the $e_g$ electron and the localized $t_{2g}$ spin $\mathbf{S}_i$

$$H_{DE}=-\sum_{i\mathbf{a}ll'\alpha} t^{\mathbf{a}}_{ll'} c^{+}_{il\alpha} c_{i+\mathbf{a}l'\alpha} - 2J_H \sum_i \mathbf{s}_i \cdot \mathbf{S}_i, \qquad (1)$$

where $c^{+}_{il\alpha}$ and $c_{il\alpha}$ are creation and annihilation operators for $e_g$ electron with spin $\alpha$ on orbitala $d_{x^2-y^2}(l=a)$ and $d_{3z-r^2}(l=b)$ at site $i$, and $\mathbf{a}$ is the vector connecting nearest-neighbor sites. For the cubic lattice, the hopping amplitudes between the orbitals along the $x,y,z$ directions are

 

$$t^x_{aa} = -\sqrt{3}t^x_{ab} = -\sqrt{3}t^x_{ba} = 3t^x_{bb} = t,$$

$$t^y_{aa} = \sqrt{3}t^y_{ab} = \sqrt{3}t^y_{ba} = 3t^y_{bb} = t,$$

$$t^z_{aa} = t^z_{ab} = t^z_{ba} = 0, \quad t^z_{bb} = 4t/3. \tag{2}$$

The second term in Eq. (1) is the Hund interaction between the spin $\mathbf{s}_i$ of the $e_g$ electron and the localized $t_{2g}$ spin $\mathbf{S}_i$ with $s^\nu_i = 1/2\Sigma_{l\alpha\beta} c^+_{il\alpha}\sigma^\nu_{\alpha\beta}c_{il\beta}$, where $\sigma^x, \sigma^y, \sigma^z$ are Pauli matrices and the Hund's constant $J_H$ is positive.

The $H_{el-ph}$ Hamiltonian models the coupling of $e_g$ electrons to the lattice distortion

$$H_{el-ph} = g\sum_i (Q_{2i}\tau_{xi} + Q_{3i}\tau_{zi}) + \frac{k}{2}\sum_i (Q^2_{2i} + Q^2_{3i}), \tag{3}$$

where $\tau_{xi} = \Sigma_\alpha (c^+_{ia\alpha}c_{ib\alpha} + c^+_{ib\alpha}c_{ia\alpha})$ and $\tau_{zi} = \Sigma_\alpha (c^+_{ia\alpha}c_{ia\alpha} - c^+_{ib\alpha}c_{ib\alpha})$. In Eq. (3) $g$ is the electron-phonon coupling constant while $Q_{2i}$ and $Q_{3i}$ are JT phonon modes. The second term in $H_{el-ph}$ is the usual quadratic potential for distortions with constant $k$. The important energy scale of the phonon-electron interaction is the static JT energy $E_{\mathrm{JT}} = g^2/(2k)$.

One can represent the spin operators $\mathbf{S}_i$ of the localized $t_{2g}$ electrons in terms of Schwinger bosons $(\varphi_{i\alpha}, \varphi^+_{i\alpha})$ $S^\nu_i = \frac{1}{2}\Sigma_{\alpha\beta}\varphi^+_{i\alpha}\sigma^\nu_{\alpha\beta}\varphi_{i\beta}$ and $\Sigma_\alpha \varphi^+_{i\alpha}\varphi_{i\alpha} = 2s$. By means of the Schwinger bosons we introduce spin-singlet Fermi fields

$$\Psi^A_{il}(\tau) = \frac{1}{\sqrt{2s}}[\varphi^+_{i1}(\tau)c_{il1}(\tau) + \varphi^+_{i2}(\tau)c_{il2}(\tau)], \tag{4}$$

$$\Psi^B_{il}(\tau) = \frac{1}{\sqrt{2s}}[\varphi_{i1}(\tau)c_{il2}(\tau) - \varphi_{i2}(\tau)c_{il1}(\tau)] \tag{5}$$

and write the spin of the $e_g$ electron and the total spin of the system $\mathbf{S}^{\mathrm{tot}}_i = \mathbf{S}_i + \mathbf{s}_i$ in terms of the singlet fermions.[15] Further, we average the total spin of the system in the subspace of the singlet fermions $A$ and $B$. The vector $\mathbf{M}_i = \langle \mathbf{S}^{\mathrm{tot}}_i \rangle_f$ identifies the local orientation of the total magnetization. Because of the fact that $t_{2g}$-electron spin is parallel with $e_g$-electron spin we obtain $\mathbf{M}_i = \frac{M}{S}\mathbf{S}_i$ with $M = S + \frac{1}{2}\Sigma_l \langle (\Psi^{A+}_{il}\Psi^A_{il} - \Psi^{B+}_{il}\Psi^B_{il}) \rangle_f$. Now, if we use Holstein-Primakoff representation for the vectors $\mathbf{M}_i(a^+, a)$ with $M$ as an "effective spin" of the system $(\mathbf{M}^2_i = M^2)$, the bose fields $a_i$ and $a^+_i$ are the true magnons in the system.

An important advantage of working with singlet fermions is the fact that in terms of these spin-singlet fields the spin-fermion interaction is in a diagonal form, the spin variables (magnons) are removed, and one accounts for it exactly. The theory is quadratic with respect to the spin-singlet fermions and one can integrate them out to obtain the free energy of fermions as a function of the magnons' fields $a^+_i, a_i$. We expand the free energy in powers of magnons' fields and keep only the first two terms. The first term $F_{f0}$, which does not depend on the magnons' fields, is a free energy of Fermions with spins of localized $t_{2g}$ electrons treated classically. We fix the model parameters and consider this term as a function of the JT distortion modes independent on the lattice sites. If we represent $Q_2 = \hat{Q}\cos\gamma$ and $Q_3 = \hat{Q}\sin\gamma$, the numerical calculations show that the function does not depend on the angle

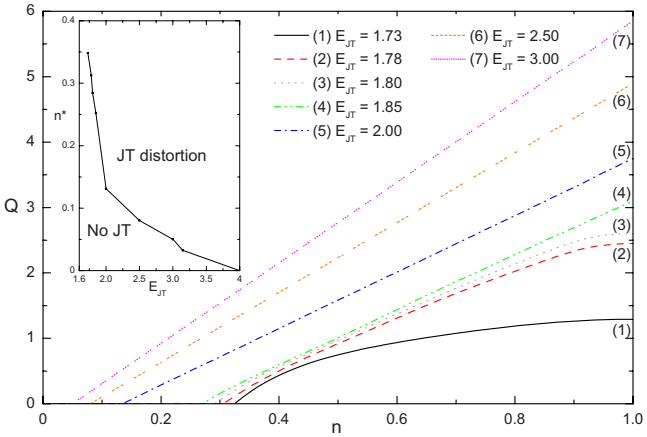


FIG. 1. (Color online) The renormalized distortion $Q = gQ_2$ as a function of density $n$ for $J_H = 15$ and different values of the JT energy $E_{\mathrm{JT}}$. Inset: the density $n^*$ at which the JT distortion appears as a function of JT energy $E_{\mathrm{JT}}$.

$\gamma$. This allows us to fix $\gamma = 0$ $(Q_3 = 0)$. The physical value of the JT distortion is the value at which $F_{f0}$ has a minimum. In this way we obtain the distortion as a function of the density of $e_g$ electrons for different values of JT energy and fixed Hund's coupling. We fix the hopping parameter $t = 1$ to set the energy unit. The results for the renormalized distortion $Q = gQ_2$ as a function of charge-carrier density $n$ are plotted in Fig. 1 for different values of the JT energy $E_{\mathrm{JT}}$ and $J_H = 15$. Figure 1 shows that JT distortion appears at critical value of the charge-carrier density $n^*$ and increases as density $n$ is increased. The inset demonstrates that $n^*$ decreases and approaches zero as JT energy $E_{\mathrm{JT}}$ is increased.

The second term in the Fermion free energy is quadratic with respect to the magnons' fields $a^+_i, a_i$ and defines the effective magnon Hamiltonian in Gaussian approximation,

$$H_{\mathrm{eff}} = \sum_{i\mathbf{a}} \rho^{\mathbf{a}}(a^+_i a_i + a^+_{i+\mathbf{a}}a_{i+\mathbf{a}} - a^+_i a_{i+\mathbf{a}} - a^+_{i+\mathbf{a}}a_i). \tag{6}$$

In Eq. (6) $\rho^{\mathbf{a}}$ are spin-stiffness constants which depend on the space directions $\mathbf{a}$. They are calculated at zero temperature, for fixed Hund's coupling, JT energy, charge density, and JT distortion determined above. The calculations follow the technique developed in Ref. 15. Based on the rotational symmetry, one can supplement Hamiltonian (6) up to an effective Heisenberg-type Hamiltonian, written in terms of the vectors $\mathbf{M}_i$

$$H_{\mathrm{eff}} = -\sum_{i\mathbf{a}} J^{\mathbf{a}}\mathbf{M}_i \cdot \mathbf{M}_{i+\mathbf{a}}, \tag{7}$$

where $J^{\mathbf{a}} = \rho^{\mathbf{a}}/M$. The ferromagnetic phase is stable if all effective exchange coupling constants are positive $J^{\mathbf{a}} > 0$ $(\rho^{\mathbf{a}} > 0)$. If one of them is negative, for example, $J^y < 0$ $(\rho^y < 0)$ and the others are positive $J^x > 0$ and $J^z > 0$ $(\rho^x > 0, \rho^z > 0)$ the stable state is $A$-type antiferromagnetic phase which has planes $(x, z)$ that are ferromagnetic (parallel moments) with antiferromagnetic (antiparallel) moments between them. The spin-stiffness constant, as a function of charge-carrier density, is depicted in Fig. 2 for $J_H = 15$ and three different values of JT energy, $E_{\mathrm{JT}} = 1.73$, $E_{\mathrm{JT}} = 2$, and

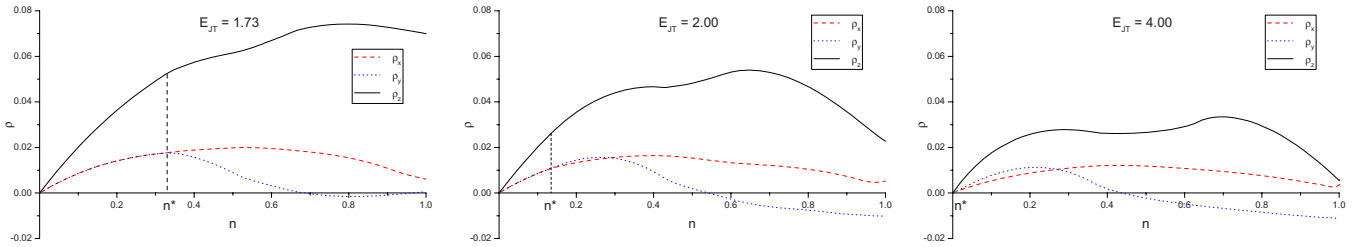


FIG. 2. (Color online) Spin-stiffness constants as a functions of density $n$ for $J_H=15$, $E_{\mathrm{JT}}=1.73$ (left), $E_{\mathrm{JT}}=2$ (middle), and $E_{\mathrm{JT}}=4$ (right). The vertical dash lines correspond to the density $n^*$ at which the JT distortion appears.

$E_{\mathrm{JT}}=4$. The vertical dashed lines correspond to the density $n^*$ at which the JT distortion appears. The figure on the left illustrates in the best way the impact of the JT distortion on the spin-stiffness constants. The appearance of the distortion at $n^*$ is accompanied with a change in the slopes of the curves. The distortion splits the $\rho^y$ (dotted) and $\rho^x$ (dashed) lines, and $\rho^y$ starts to decrease. At critical density $n_c$ and $\rho^y$ become equal to zero and the system undergoes a transition from ferromagnetic phase to $A$-type antiferromagnetic phase. The two other figures shows that spin-stiffness constants decrease when JT energy increases and the critical density $n_c$ decreases too. As the spin-stiffness constants are a measure for the magnon fluctuations in the ferromagnetic phase we conclude that JT distortion suppresses the magnon fluctuations.

The most evident consequence of this suppression is the Curie temperature ($T_{\mathrm{C}}$) decreasing. To calculate $T_{\mathrm{C}}$ we utilize the Schwinger bosons mean-field theory.[16] We represent the vector $\mathbf{M}_i$ Eq. (7) by means of Schwinger bosons $(\phi_{i\alpha}, \phi^+_{i\alpha})$

$$M^{\nu}_i = \frac{1}{2}\sum_{\alpha\beta} \phi^+_{i\alpha}\sigma^{\nu}_{\alpha\beta}\phi_{i\beta} \quad \phi^+_{i\alpha}\phi_{i\alpha} = 2M. \tag{8}$$

Next we use the identity

$$\mathbf{M}_i \cdot \mathbf{M}_j = \frac{1}{2}(\phi^+_{i\alpha}\phi_{j\alpha})(\phi^+_{j\beta}\phi_{i\beta}) - \frac{1}{4}(\phi^+_{i\alpha}\phi_{i\alpha})(\phi^+_{j\beta}\phi_{j\beta}) \tag{9}$$

and rewrite the effective Hamiltonian in the form

$$H_{\mathrm{eff}} = -\frac{1}{2}\sum_{i\mathbf{a}} J^{\mathbf{a}}(\phi^+_{i\alpha}\phi_{i+\mathbf{a}\alpha})(\phi^+_{i+\mathbf{a}\beta}\phi_{i\beta}), \tag{10}$$

where the constant term is dropped. To ensure the constraint we introduce a parameter ($\lambda$) and add a new term to the effective Hamiltonian (10),

$$\hat{H}_{\mathrm{eff}} = H_{\mathrm{eff}} + \lambda\sum_i (\phi^+_{i\sigma}\phi_{i\sigma} - 2M). \tag{11}$$

We treat the four-boson interaction within Hartree-Fock approximation. The Hartree-Fock hamiltonian which corresponds to the effective Hamiltonian reads

$$H_{\mathrm{H\text{-}F}} = \frac{1}{2}\sum_{i\mathbf{a}} J^{\mathbf{a}}\bar{u}_{i,i+\mathbf{a}}u_{i,i+\mathbf{a}} + \lambda\sum_i (\phi^+_{i\sigma}\phi_{i\sigma} - 2M)$$
$$- \frac{1}{2}\sum_{i\mathbf{a}} J^{\mathbf{a}}[\bar{u}_{i,i+\mathbf{a}}\phi^+_{i\alpha}\phi_{i+\mathbf{a}\alpha} + u_{i,i+\mathbf{a}}\phi^+_{i+\mathbf{a}\alpha}\phi_{i\alpha}], \tag{12}$$

where $\bar{u}_{i,i+\mathbf{a}}$ ($u_{i,i+\mathbf{a}}$) are Hartree-Fock parameters to be determined self-consistently. We are interested in real parameters which do not depend on the lattice sites but depend on the space directions $u_{i,i+\mathbf{a}}=\bar{u}_{i,i+\mathbf{a}}=u_{\mathbf{a}}$. Then in momentum space representation, Hamilonian (12) has the form

$$H_{\mathrm{H\text{-}F}} = \frac{N}{2}\sum_{\mathbf{a}} u^2_{\mathbf{a}}J^{\mathbf{a}} - 2\lambda MN + \sum_k \varepsilon_k\phi^+_k\phi_k, \tag{13}$$

where $N$ is the number of lattice sites and $\varepsilon_k$ is the dispersion of the $\phi_k$ boson (spinon). The free energy of the theory with Hamiltonian $H_{\mathrm{H\text{-}F}}$ is

$$F = \frac{1}{2}\sum_{\mathbf{a}} u^2_{\mathbf{a}}J^{\mathbf{a}} - 2\lambda M + \frac{2T}{N}\sum_k \ln(1 - e^{-\varepsilon_k/T}), \tag{14}$$

where $T$ is the temperature. The equations for the parameters $u_{\mathbf{a}}$ and $\lambda$ are $\partial F/\partial u_{\mathbf{a}}=0$ and $\partial F/\partial\lambda=0$.

To solve the system of four equations it is more convenient to introduce a new parameter ($\mu$) instead of ($\lambda$): $\lambda = \Sigma_{\mathbf{a}}(u_{\mathbf{a}}J^{\mathbf{a}} + \mu u_{\mathbf{a}})$. In terms of the new parameter the $\phi_k$-boson dispersion is $\varepsilon_k = \Sigma_{\mathbf{a}}[u_{\mathbf{a}}J^{\mathbf{a}}(1-\cos k_{\mathbf{a}}) + \mu u_{\mathbf{a}}]$ and the theory is well defined for positive constants $u_{\mathbf{a}} \geq 0$ and $\mu \geq 0$. For high enough temperatures $\mu(T)$ and $u_{\mathbf{a}}(T)$ are positive and the

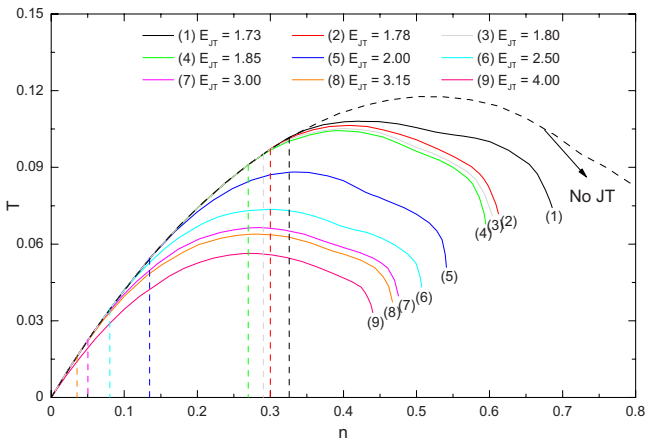


FIG. 3. (Color online) $T_{\mathrm{C}}$ as a function of $e_g$ electron density $n$ for $J_H=15$ and different values of the JT energy.

TABLE I. Maximal Curie temperatures ($T_C^{max}$[K]), for $J_H/t=15$ ($J_H=12$ eV), different JT energies ($E_{JT}/t$), and the corresponding $e_g$-electron densities ($n$).

| $E_{JT}/t$ | 1.73 | 1.78 | 1.80 | 1.85 | 2.00 | 2.50 | 3.00 | 3.15 | 4.00 |
|---|---|---|---|---|---|---|---|---|---|
| $n$ | 0.42 | 0.41 | 0.40 | 0.39 | 0.33 | 0.30 | 0.283 | 0.282 | 0.27 |
| $T_C^{max}$ (K) | 1004 | 988 | 976 | 970 | 818 | 684 | 618 | 594 | 522 |

excitation is gapped. Decreasing the temperature leads to decrease in $\mu(T)$. At temperature $T_C$ it becomes equal to zero $\mu(T_C)=0$ and long-range excitation emerges in the spectrum. Therefore this is the Curie temperature. We set $\mu=0$ and obtain a system of equations for the Curie temperature $T_C$ and $u_{\mathbf{a}}$

$$u_{\mathbf{a}'} = \frac{2}{N}\sum_k \frac{\cos k_{\mathbf{a}'}}{\exp\left[\frac{1}{MT_C}\sum_{\mathbf{a}} u_{\mathbf{a}}\rho^{\mathbf{a}}(1-\cos k_{\mathbf{a}})\right]-1},$$

$$M = \frac{1}{N}\sum_k \frac{1}{\exp\left[\frac{1}{MT_C}\sum_{\mathbf{a}} u_{\mathbf{a}}\rho^{\mathbf{a}}(1-\cos k_{\mathbf{a}})\right]-1}. \quad (15)$$

The results for the Curie temperature $T_C$ as a function of $e_g$ electrons density $n$ are plotted in Fig. 3 for $J_H=15$ and different values of the JT energy. The upper (black) dashed line is a reference line which corresponds to the case without JT distortion. The vertical dashed lines correspond to the density $n^*$ while the ends of the $T_C-n$ curves correspond to the critical density $n_c$ at which the system undergoes transition to $A$-type antiferromagnetic state. The appearance of the JT distortion at $n^*$ leads to a splitting of the reference curve and the curve for a system with JT distortion. The density $n^*$ decreases when JT energy increases and the ferromagnetic phase is strongly suppressed, which in turn leads to the decreasing of the Curie temperature.

So far we have fixed $t=1$ to set the energy unit. Now, we want to report the numerical results from Fig. 3 in Kelvin and in order to do this we utilize $t=0.8$ eV.[10] (Table I).

We have used a large value for Hund's constant to better demonstrate the impact of the JT distortion on the ferromagnetism. Decreasing of $J_H$ suppresses the ferromagnetic phase, decreases the Curie temperature, and reduces the impact of the JT distortion on the ferromagnetism. For example, for $J_H/t=15$ and absence of JT distortion we have $T_C=1092$ K while for $J_H/t=5$ we obtain $T_C=738$ K. For nonzero distortion, $E_{JT}/t=2$, the Curie temperatures are $T_C=818$ and 620 K, respectively.

In summary, we have shown that JT distortion strongly suppresses the ferromagnetic order because of the suppression of the magnon fluctuations, which in turn, results from the decreasing of the spin-stiffness constants. Our results demonstrate that the reduction in the Curie temperature due to JT distortion depends on the value of Hund's constant $J_H$.

The authors acknowledge the financial support of the Sofia University under Grant No. 095/2009. This work was partly supported by a Grant-in-Aid DO02-264/18.12.08 from NSF, Bulgaria.

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