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Magnetic transition and polaron crossover in a two-site single polaron model including double exchange interaction

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Abstract. A two-site double exchange model with a single polaron is studied using a perturbation expansion based on the modified Lang-Firsov transformation. The antiferromagnetic to ferromagnetic transition and the crossover from small to large polaron are investigated for different values of the antiferromagnetic interaction (J) between the core spins and the hopping (t) of the itinerant electron. Effect of the external magnetic field on the small to large polaron crossover and on the polaronic kinetic energy are studied. When the magnetic transition and the small to large polaron crossover coincide for some suitable range of J/t, the magnetic field has very pronounced effect on the dynamics of polarons.

PACS. 71.38.+i Polarons and electron-phonon interactions – 63.20.Kr Phonon-electron and phonon-phonon interactions – 75.30.Vn Colossal magnetoresistance

1 Introduction

The origin of ferromagnetism in manganese perovskites $La_{1-x}X_xMnO_3$ (X = Ba, Sr, Ca etc.) is the double exchange mechanism [1,2]. The discovery of anomalous magnetotransport phenomena [3] in these compounds has stimulated intensive studies in its magnetic as well as electrical properties. However, the simple double-exchange interaction alone is not sufficient to explain the experimental results [3]. Depending on the doping, temperature and the radius of the dopant ion these oxides show various phases [4]. A complete understanding of the properties of Mn perovskites is still far from clear.

One of the keys to understand unusual physical properties is to find the role of coupling between the carriers and the underlying lattice. Several theoretical models have been proposed based on lattice-carrier coupling [5–10]. Many recent experiments [11] indicate that the electron-phonon (e-ph) interaction shapes its properties very crucially. Moreover, small to large polaron crossover is reported by many experimental groups [12–15]. There are models [6,7,10] which incorporate double exchange interaction in a polaronic model. Min and co-workers [6] have studied the role of e-ph interaction in systems where double exchange interaction is present. The combined model of spin double exchange and lattice polaron [6] is used to investigate the effect of small to large polaron crossover on the magnetic and transport properties under the mean-field approximation scheme. It concludes that the effect of polaron narrowing on the colossal magnetoresistance (CMR) is more pronounced in CMR manganites with low magnetic transition temperature (T_c) than in the high T_c manganites. Within the mean field theory [6] the magnetic transition, metal-insulator transition and a large drop in magnetoresistance occur at the same temperature.

In an attempt to have a clearer view from a nearly exact calculation, we include double exchange interaction in a two site one polaron model and follow a perturbation expansion [16] based on a modified Lang-Firsov (MLF) phonon basis where the lattice distortions produced by the electron are treated as variational parameters [17–19]. This method shows good convergence as well as nearly exact results [16] for almost the entire range of e-ph coupling for $t/\omega_0 \leq 1$, where ω_0 is the phonon frequency. We investigate the ferromagnetic (FM) to antiferromagnetic (AFM) transition, the crossover from large to small polaron, the behavior of the effective hopping of the itinerant electron and the kinetic energy as a function of the e-ph coupling strength for the ground state of the system. Effect of the magnetic field on the large to small polaron crossover and on the polaronic kinetic energy are also studied.

The paper is organized as follows. In Section 2 we define the model Hamiltonian describing different interactions and calculate different physical quantities which indicate the behaviour of small-large polaron crossover and AFM-FM transition as a function of e-ph coupling. In Section 3 we present the results obtained in our calculation and discussions. Section 4 contains the concluding remarks.

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2 Formalism

The Holstein model [20] including double exchange interaction for the two-site single-polaron system is described by the Hamiltonian

$$H = \sum_{i,\sigma} \epsilon n_{i\sigma} - \sum_{\sigma} t\cos(\frac{\theta}{2})(c^{\dagger}_{1\sigma}c_{2\sigma} + c^{\dagger}_{2\sigma}c_{1\sigma}) + g\omega_0 \sum_{i,\sigma} n_{i\sigma}(b_i + b^{\dagger}_i) + \omega_0 \sum_i b^{\dagger}_i b_i + J\sum_{\langle ij\rangle} \mathbf{S}_i\mathbf{S}_j \quad (1)$$

where i =1 or 2, denotes the site. $c_{i\sigma}$ ($c^{\dagger}_{i\sigma}$) is the annihilation (creation) operator for the itinerant electron with spin σ at site i, $n_{i\sigma}$ ($= c^{\dagger}_{i\sigma}c_{i\sigma}$) is the corresponding number operator and g denotes the on-site e-ph coupling strength. b_i and $b^{\dagger}_i$ are the annihilation and creation operators respectively for the phonons corresponding to interatomic vibrations at site i, ω_0 is the corresponding phonon frequency and ϵ is the bare-site energy. $\mathbf{S}_i$ and $\mathbf{S}_j$ are the core-spins at the site i and j respectively, θ is the angle between the core-spins $\mathbf{S}_i$ and $\mathbf{S}_j$. The transfer hopping integral (t) is modified through the relative angle θ as $t\cos(\frac{\theta}{2})$ because of the strong Hund's coupling between the spins of the core electrons and itinerant electron [2]. J is the superexchange antiferromagnetic interaction between the neighbouring core-spins. Since we will restrict ourselves to the single-electron case we will not consider the electron spin indices.

If one compares our simple model with the manganite (*e.g.* $La_{1-x}Ca_xMnO_3$) system the core spin and the itinerant electron may be identified with the t^3_{2g} (localized) electrons and the e^1_g (mobile) electron of Mn^{3+} of the manganite system respectively. It may be mentioned that for manganites the Jahn-Teller(JT) coupling is important whereas the Holstein phonon mode, in general, should correspond to the breathing mode in manganites [8]. To include JT coupling one should consider two e_g orbitals (say α and β) and their coupling to the JT phonon mode. For the single electron case, as considered here, if one neglects the interorbital hopping ($t_{\alpha\beta} = 0$) then the Hamiltonian can be separated into two parts each corresponds to 1-orbital case and as a consequence the 2-orbital problem is reduced to an effective single orbital problem with an effective Holstein type interaction as in Hamiltonian (1). For the two electron case the above simplification cannot be done and a study of two-site two-electron problem considering two orbitals in the context of manganites is in progress. We have considered here one itinerant electron in a two-site 1-orbital system. So, the density of itinerant electron $\langle n\rangle$ is equal to 0.5 which, in principle,corresponds to $x = 0.5$ for manganite system. It may be mentioned that the studies of 1-orbital [9] and 2-orbital Kondo model [7] in the context of manganites using Monte Carlo techniques showed that the results for 2-orbital model at $n = 1$ ($x = 0$) are similar to those for single orbital model at $x = 0.5$.

By introducing new phonon operators $a = (b_1 + b_2)/\sqrt{2}$ and $d = (b_1 - b_2)/\sqrt{2}$, the Hamiltonian (H) is separated into two parts : one corresponding to the in-phase mode which does not couple with the electronic degrees of freedom and the other involving out-of-phase mode (H_d) which represents an effective e-ph system and cannot be solved analytically [21].

The MLF transformation with variable phonon basis is used so that a convergent perturbation expansion can be obtained. The transformed Hamiltonian is

$$\tilde{H}_d = \mathrm{e}^{R}H_d\mathrm{e}^{-R} = \omega_0 d^{\dagger}d + \sum_i \epsilon_p n_i - t\cos\left(\frac{\theta}{2}\right) \times \left[c^{\dagger}_1 c_2 \exp(2\lambda(d^{\dagger} - d)) + c^{\dagger}_2 c_1 \exp(-2\lambda(d^{\dagger} - d))\right] + \omega_0(g_+ - \lambda)(n_1 - n_2)(d + d^{\dagger}) + JS^2\cos\theta \quad (2)$$

where $R = \lambda(n_1 - n_2)(d^{\dagger} - d)$, λ is a variational parameter related to the displacement of the d oscillator, $g_+ = g/\sqrt{2}$ and $\epsilon_p = \epsilon - \omega_0(2g_+ - \lambda)\lambda$.

For the perturbative expansion following reference [16] the basis set is chosen as $|\pm, N\rangle = \frac{1}{\sqrt{2}}(c^{\dagger}_1 \pm c^{\dagger}_2)\ |0\rangle_e|N\rangle$, where $|+\rangle$ and $|-\rangle$ are the bonding and antibonding electronic states and $|N\rangle$ denotes the Nth excited oscillator state within the MLF phonon basis. The diagonal part of the Hamiltonian $\tilde{H}_d$ in the chosen basis is treated as the unperturbed Hamiltonian (H_0) and the remaining part of the Hamiltonian $H_1 = \tilde{H}_d - H_0$, as a perturbation.

The unperturbed energy of the state $|\pm, N\rangle$ is given by

$$E^{(0)}_{\pm,N} = \langle N, \pm|H_0|\pm, N\rangle = N\omega_0 + \epsilon_p \mp t_{\mathrm{eff}}\left[\sum_{i=0}^{N} \frac{(2\lambda)^{2i}}{i!}(-1)^i N_{C_i}\right] + JS^2\cos\theta \quad (3)$$

where $t_{\mathrm{eff}} = t\ \cos\frac{\theta}{2}\ \exp(-2\lambda^2)$ and the general off-diagonal matrix elements of H_1 between the two states $|\pm, N\rangle$ and $|\pm, M\rangle$ may be calculated for $(N - M) > 0$ as in reference [16].

Within the chosen basis, the unperturbed ground state is the $|+\rangle|0\rangle$ state with the unperturbed energy, $E^{(0)}_0 = \epsilon_p - t_{\mathrm{eff}} + JS^2\cos\theta$.

The first order correction to the ground state wave function is obtained as,

$$|\psi^{(1)}_0\rangle = \frac{[\omega_0(g_+ - \lambda) - 2\lambda t_{\mathrm{eff}}]}{(E^{(0)}_0 - E^{(0)}_{+,1})}\ |-, 1\rangle - \sum_{N=2,3,4,..} \frac{t_{\mathrm{eff}}(2\lambda)^N}{\sqrt{N!}(E^{(0)}_0 - E^{(0)}_{e,N})}\ |e, N\rangle \quad (4)$$

where $e = +$ or $-$ for even and odd N respectively.

The first order correction to the energy ($E^{(1)}_0$) is zero since H_1 has no diagonal matrix element in the chosen basis. The second order correction to the ground state

energy is given by

$$E_0^{(2)} = \sum_{N=1,3..} \frac{|\frac{-t_{\rm eff}(2\lambda)^N}{\sqrt{N!}} + \omega_0(g_+ - \lambda)\ \delta_{N,1}|^2}{(E_0^{(0)} - E_{-,N}^{(0)})} + \sum_{N=2,4..} \frac{|\frac{-t_{\rm eff}(2\lambda)^N}{\sqrt{N!}}|^2}{(E_0^{(0)} - E_{+,N}^{(0)})}. \quad (5)$$

Higher order corrections to the ground state wavefunction and energy are obtained following reference [16]. For the study of the effect of the magnetic field (h) we include a term $-\tilde{g}\sum_i \mu_{\rm B} h S_i \cos\frac{\theta}{2}$ to the Hamiltonian in equation (1) (where $\tilde{g}$ is the Lande g factor) and as a result a term $-2\mu_{\rm eff} h \cos\frac{\theta}{2}$ is added to $E_0^{(0)}$ where $\mu_{\rm eff}$ $(= \tilde{g} S \mu_{\rm B})$ is the local moment of the core spins. In this paper we express the magnetic field (h) in a unit of $\mu_{\rm eff} = 1$.

Now a proper choice of λ is to be made so that the perturbative expansion becomes convergent. Our previous work [16] has shown that the λ, obtained by minimizing the unperturbed ground state energy, gives satisfactory convergence to the perturbation series for $t/\omega_0 \leq 1$ and the convergence becomes very rapid and excellent with decreasing value of t/ω_0. Here also we will follow the similar procedure. Minimizing the unperturbed ground state energy $E_0^{(0)}$ with respect to λ we obtain

$$\lambda = \frac{\omega_0 g_+}{\omega_0 + 2t_{\rm eff}}\,. \quad (6)$$

Minimization of the unperturbed ground state energy with respect to θ gives an approximate value ($\theta_{\rm MLF}$) of θ

$$\cos\frac{\theta_{\rm MLF}}{2} = \left[\frac{t\exp(-2\lambda^2)}{4JS^2} + \frac{\mu_{\rm eff} h}{2JS^2}\right] \quad \text{(for nonzero solution of } \theta_{\rm MLF}).$$

However, exact θ should be evaluated from the minimization of the exact ground state energy. For each value of g_+ we calculate the energy up to the sixth order in perturbation [16] and find out for which value of θ the energy (including perturbation corrections) is minimum.

We have also calculated $t_{\rm eff}^{\rm KE} = -E_{\rm Kin} = \langle\psi_{\rm G}|\ t\cos(\frac{\theta}{2})\ [c_1^\dagger c_2\ \exp(2\lambda(d^\dagger - d)) + c_2^\dagger c_1\ \exp(-2\lambda(d^\dagger - d))]|\psi_{\rm G}\rangle$ where $\psi_{\rm G}$ is the ground state wave-function which we have calculated up to the fifth order corrections in perturbation. $t_{\rm eff}^{\rm KE}$ describes the kinetic energy of the system and it reduces to the effective hopping ($t_{\rm eff}$) for both the weak and strong coupling limits [22].

The static correlation functions $\langle n_1 u_1\rangle_0$ and $\langle n_1 u_2\rangle_0$, where u_1 and u_2 are the lattice deformations at sites 1 and 2 respectively, produced by an electron at site 1, are the standard measure of polaronic character. The correlation functions for this two-site system may be written as [16]

$$\langle n_1 u_1\rangle_0 = \frac{1}{2}\left[-(g_+ + \lambda) + \frac{A_0}{N_{\rm G}}\right] \quad (7)$$

$$\langle n_1 u_2\rangle_0 = \frac{1}{2}\left[-(g_+ - \lambda) - \frac{A_0}{N_{\rm G}}\right]$$

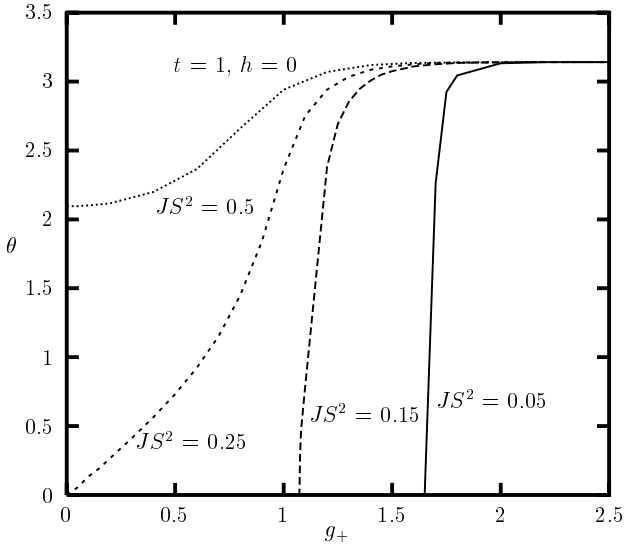

Fig. 1. Variation of the relative angle (θ) between the core-spins, with g_+ for $t = 1.0$ and $JS^2 = 0.05$, 0.15, 0.25 and 0.5 (in units of ω_0=1).

where

$$A_0 \equiv \langle\psi_{\rm G}|n_1(d + d^\dagger)|\psi_{\rm G}\rangle$$

where $N_{\rm G}$ is the normalization factor to the wavefunction. The variation of the physical quantity $\lambda_{\rm corr}/g_+ = -\langle n_1(u_1 - u_2)\rangle_0/g_+$ with the e-ph coupling strength manifests the nature of large to small polaron crossover. Its value becomes 1 in the extreme small polaronic limit where the lattice distortion is very local and decreases significantly in the small to large polaron crossover region as g_+ decreases.

3 Results and discussions

In the combined model of polaron and double exchange interaction, the nature of variation of θ, $t_{\rm eff}^{\rm KE}$ and $\lambda_{\rm corr}/g_+$ with g_+ reveal different transitions and crossover regions. We focus our attention to the parameter space where $t > J$ as it corresponds to real systems [7,23]. We present the results for $t = 1.0$ and $JS^2 = 0.05, 0.15, 0.25$ and 0.5 (in a scale of $\omega_0 = 1.0$). The bare-site energy ϵ is taken as zero.

Figure 1 shows the change of relative orientation (θ) of two core spins as a function of e-ph coupling strength (g_+) for the ground state. For small values of g_+ two core spins are either aligned parallel or canted depending on the strength of t/JS^2. With increasing g_+ the angle (θ) increases and finally a transition from the FM or canted AFM state to the AFM state occurs. Nature of this transition depends on the ratio of t to JS^2. For $t = 1.0$, $JS^2 = 0.05$ the FM to AFM transition is very sharp. With increasing value of JS^2 the transition occurs at a lower value of g_+ and it becomes broader. For higher values of JS^2 $(= 0.25, 0.5)$, the crossover from the canted

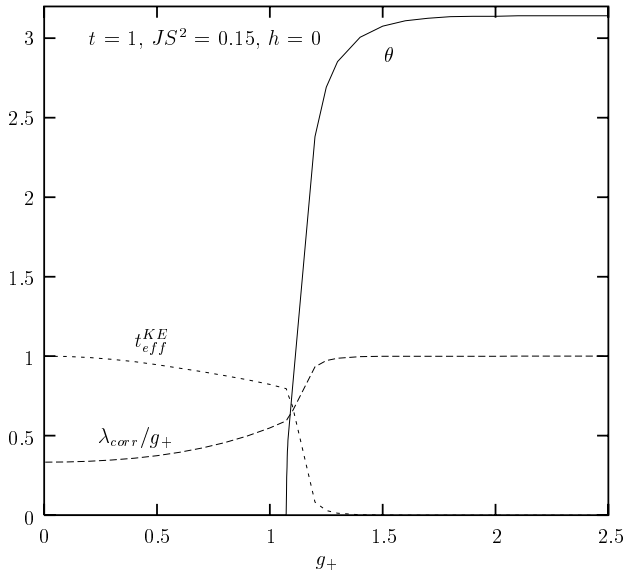

Fig. 2. Variations of θ, $t_{\rm eff}^{\rm KE}$ and $\lambda_{\rm corr}/g_+$ with g_+ for $t = 1.0$, $JS^2 = 0.15$ and $h = 0$.

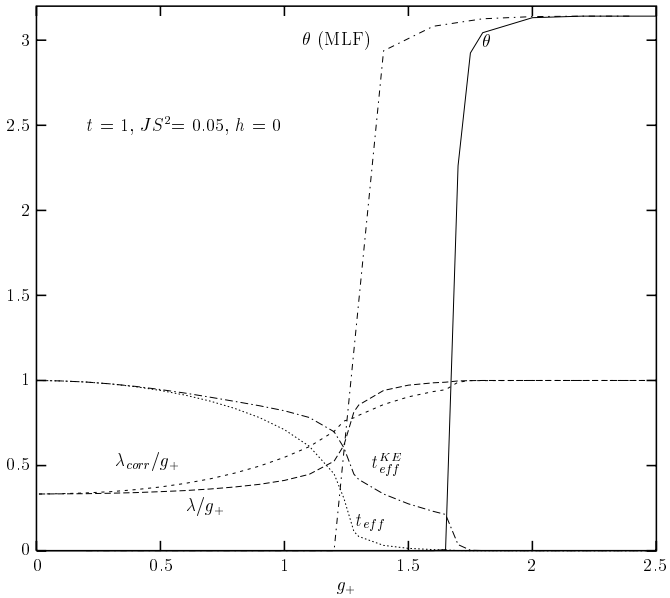

Fig. 3. Variations of θ (with and without perturbation), $t_{\rm eff}^{\rm KE}$, $t_{\rm eff}$, $\lambda_{\rm corr}/g_+$ and λ/g_+ with g_+ for $t = 1.0$, $JS^2 = 0.05$ and $h = 0$. Note that θ(MLF), $t_{\rm eff}$ and λ/g_+ are obtained within the MLF method without considering perturbation corrections.

state to the AFM state is very smooth. It may be mentioned that Yunoki and Moreo [9] studied the 1-orbital Kondo model with AF interaction (J) between the core spins (in absence of e-ph interaction) in the context of manganites and obtained FM state at $n = 0.5$ for low values of J. With increasing J the ground state at quarter filling ($n = 0.5$) appears to be a spin spiral state and ultimately an AF state. For the two orbital model in presence of e-ph coupling a similar FM state is observed for low values of e-ph coupling for $n = 1$ and an AF state for large values of e-ph coupling.

With increasing g_+ a large to small polaron crossover occurs in the system in addition to the magnetic transition. This is clearly seen from Figures 2 and 3 where the variations of $t_{\rm eff}^{\rm KE}$, $\lambda_{\rm corr}/g_+$ and θ with g_+ are shown. The large to small polaron crossover is identified by the sharp fall in $t_{\rm eff}^{\rm KE}$ and rise in $\lambda_{\rm corr}$. For $t = 1.0$ and $JS^2 = 0.15$ the FM to AFM transition and the large to small polaron crossover occur simultaneously (Fig. 2) around $g_+ = 1.2$. However, for JS^2=0.05 the magnetic transition occurs at a higher values of g_+ than that corresponding to large to small polaron crossover (Fig. 3). In this situation the FM state has two regions: one with appreciable value of $t_{\rm eff}^{\rm KE}$ and the other with much reduced value of $t_{\rm eff}^{\rm KE}$ (Fig. 3). In the former case, charge carrier is the delocalized large polaron with appreciable hopping and this regime is expected to be metallic in the thermodynamic limit. In the latter region the hopping would be heavily suppressed by the small polaron formation and it may result in an insulating FM state in some cases. Thus we find that there is a possibility of FM metal-FM insulator-AFM insulator transition for low values of JS^2/t. It is to be noted that if one follows the usual MLF method (zeroth order of perturbation within our approach) the FM-AFM transition coincides with large to small polaron crossover for any value of JS^2/t. This is demonstrated in Figure 3 for $JS^2/t = 0.05$, where we find the coincidence of the FM-AFM transition with the large-small polaron crossover within the usual MLF approach whereas the nearly exact calculations yield different result. The nearly exact results, obtained from the MLF perturbation method, makes the large-small polaron crossover smooth in favour of the conclusion of Löwen [24].

In Figure 4 we have shown the effect of the external magnetic field on θ, $\lambda_{\rm corr}/g_+$ and $t_{\rm eff}^{\rm KE}$ for $JS^2 = 0.15$. The magnetic field favours the FM state, hence the FM-AFM transition and associated large-small polaron crossover (for $JS^2/t = 0.15$) take place at a higher value of g_+ with increasing field. From Figure 4 it is clear that the magnetic field reduces the value of the local distortion ($\lambda_{\rm corr}/g_+$) and enhances $t_{\rm eff}^{\rm KE}$ significantly in the large-small polaron crossover region provided the crossover is associated with a FM-AFM transition.

For $JS^2/t = 0.05$ the system remains in the FM state in the large-small polaron crossover region, consequently the magnetic field has no effect on $t_{\rm eff}^{\rm KE}$ in this crossover region. However, the field has an effect on θ, hence on $t_{\rm eff}^{\rm KE}$ at the FM-AFM transition.

In Figure 5 we plot the change in $t_{\rm eff}^{\rm KE}$ due to the magnetic field as a function of g_+. This quantity may be related to the magnetoresistance for a system in the thermodynamic limit. For polarons the conductivity at low temperatures is dominated by the tunneling mobility which is proportional to $t_{\rm eff}^2$ within the zeroth order of perturbation [10]. In general the $t_{\rm eff}^{\rm KE}$ is a measure of delocalization of the electron. A reduction of $t_{\rm eff}^{\rm KE}$ will cause a reduction in mobility. In Figure 5 the change in $t_{\rm eff}^{\rm KE}$ due to the field is very sharp and prominent at the FM-AFM transition when it is associated with the large-small polaron

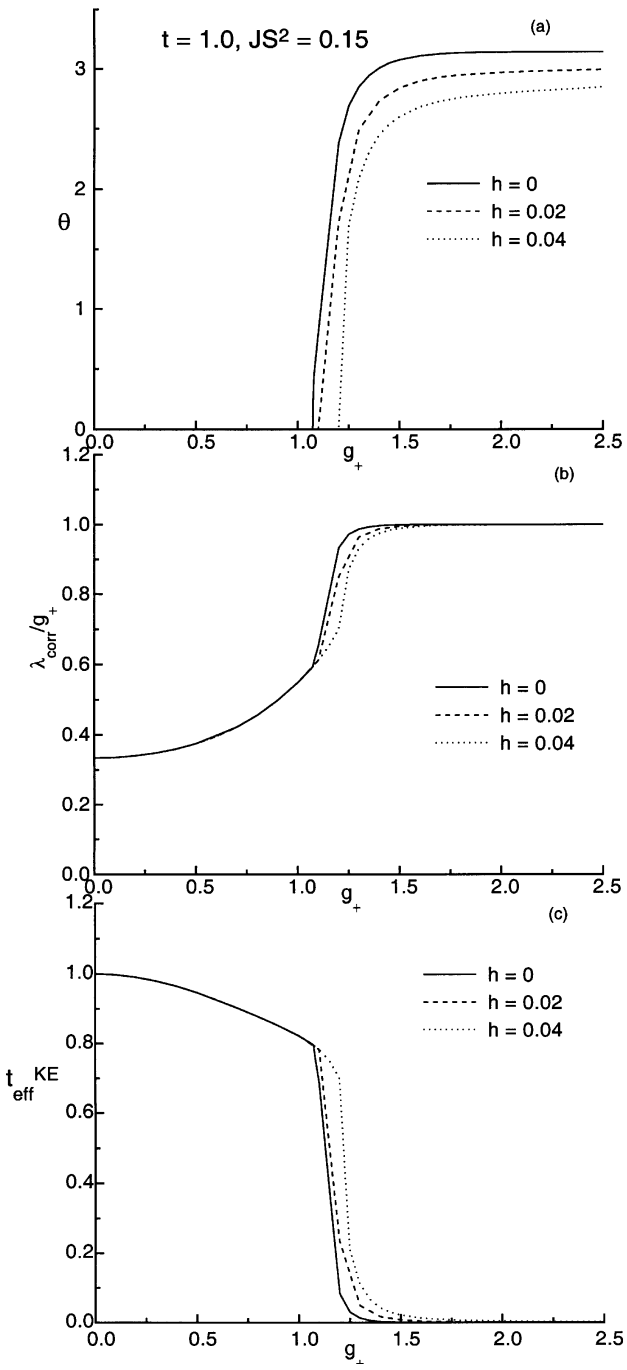

Fig. 4. Variation of (a) θ, (b) $\lambda_{\rm corr}/g_+$ and (c) $t_{\rm eff}^{\rm KE}$ with g_+ for different values of the magnetic field $h = 0$, 0.02 and 0.04 (in units of $\mu_{\rm eff}$=1) for $t = 1.0$ and $JS^2 = 0.15$.

crossover for $JS^2/t = 0.15$, while it shows a broad smaller peak for $JS^2/t = 0.05$ where the FM-AFM transition is not associated with the polaron crossover. For the former case the magnetic field would have pronounced effect on the transport properties.

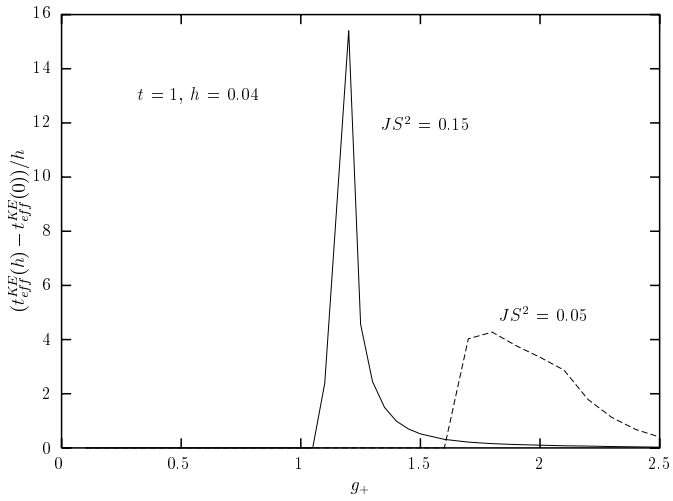

Fig. 5. Variation of the change in $t_{\rm eff}^{\rm KE}$ due to the magnetic field $\left((t_{\rm eff}^{\rm KE}(h) - t_{\rm eff}^{\rm KE}(0))/h\right)$ as a function of g_+ for $t = 1.0$, $JS^2 = 0.05$ and 0.15 and $h = 0.04$.

4 Conclusions

From our studies on the two-site double exchange model with a single polaron as a function of e-ph coupling strength we conclude that the nature of the FM-AFM transition depends on the relative values of J and t. The transition is sharper for smaller values of J/t. For high values of J/t a canted state is stable instead of a FM state for weak e-ph coupling (g_+) and the crossover from the canted state to the AFM state is very smooth with increasing g_+. For suitable values of J/t (= 0.15) the FM-AFM transition coincides with the large-small polaron crossover. For this case the external magnetic field has very prominent effect on the polaronic local distortion and kinetic energy in the transition region (as in Figs. 4b and 4c respectively). Within the MLF method (zeroth order of perturbation) the polaron crossover is always associated with FM-AFM transition for any value of J/t whereas our results based on convergent perturbation expansion yield that the coincidence of magnetic transition and polaron crossover will depend on the value of t and J. For low values of J/t a crossover from the FM large polaronic state to the FM small polaronic state with reduced hopping occurs and then a transition to the AFM state takes place at a higher value of g_+.

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