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Density of states and phase diagram of the antiferromagnetic spin chain with Dzyaloshinsky–Moriya interaction and spin-phonon coupling

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Abstract

The effects of DM interaction on the density-of-states, the dimerization and the phase diagram in the antiferromagnetic Heisenberg chain coupled with quantum phonons have been studied by a nonadiabatic analytical approach. The results show that the effect of the DM interaction is to increase the staggered antisymmetric spin exchange interaction order but to decrease the spin dimerization and their competitions result in the lattice dimerization ordering parameter to increase for large staggered DM interaction parameter β and decrease for small β. A crossover of β exists in which the dimerization ordering parameter changes non-monotonously. As the DM interaction parameter D increases, depending on the appropriate values of spin-phonon coupling, phonon frequency and β, the system undergoes phase transition from spin gapless state to gapped state or reversely and can even reenter between the two states. The relation between the phonon-staggered ordering parameter, the spin-dimer order parameter and the staggered DM interaction order parameter gives clearly their contributing weights to the lattice dimerization.

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In recent years, the Dzyaloshinsky–Moriya (DM) interaction [1] has gained renewed interest because of the evidence of its existences in a variety of quasi-one-dimensional magnets [2–4] and its applications to describe the unusual anisotropy of the magnetic susceptibility observed in cuprate superconductors [5] and to interpret an anomalous magnetic behavior in $BaCu_2M_2O_7$ ($M = Si, Ge$) [6,7], La_2CuO_4 [8], Yb_4As_3 [14] and YVO_3–$SrVO_3$ systems [9]. The experiments such as high-field neutron-scattering measurements on Cu benzoate [10] and electron paramagnetic resonance investigations in $CuGeO_3$ [11–13] manifest that in these materials the DM interaction plays an important role [14–16] and a study of one-dimensional DM Hamiltonian seems to be of great importance [17]. The antisymmetric character of DM interaction is expressed by two principal cases, the uniform [18,19] and the staggered DM interactions [17,19,20], and has been studied by many previous authors. A numerical calculations of the ground-state energy of the spin-Peierls XX model and Heisenberg chain with DM interaction by Derzhko et al. [21] found that uniform DM interaction may act against the dimerization but staggered DM interaction may act in favor of the dimerization. However, whether the staggered DM interaction always enhances the dimerization or the uniform DM interaction always acts against the dimerization has not been clearly answered. Up to now, all of the theoretical studies on DM interaction in spin-chain systems have used the static model and treated the problem in adiabatic limit. By considering the static model, several attempts have been performed to treat the spin-phonon coupling [21,22]. The validity of this static model is based on the assumption that the frequencies of the phonons associated with the dimerization are much smaller than the dimerization gap and the exchange integral. However, this is questionable, and the amplitude of the dimerization is

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substantially underestimated when compared with estimates from structural data in the spin-Peierls phase [23]. The effect of quantum fluctuations in quasi-one-dimensional systems is significant, resulting in many interesting phenomena and must be taken into account to satisfactorily describe some physical properties of quasi-one-dimensional spin-Peierls system [24,25]. An interesting and still controversial problem is how the effect of DM interaction on the dimerized ground state is modified when quantum lattice fluctuations are taken into account. However, with the coupling of spin systems to quantum phonons, this problem is rather difficult to handle analytically, which has brought much uncertainty in the interpretation of experimental data and has limited our understanding of many interesting quantum phenomena of low-dimensional magnetic materials. An analytical and nonadiabatic study of the DM interaction in spin-Peierls system will make it possible to have an insight into the intrinsical properties of the spin chain materials. In a recent work [26], the effect of DM interaction on the spin-Peierls dimerization was studied by considering the XY spin chain and some interesting results were obtained. In this paper, we focus on the properties of the density-of-states (DOS), the phase diagram and the dimerization order parameter of the antiferromagnetic Heisenberg model with spin-phonon coupling by developing a nonadiabatic analytical approach with the view of understanding the effects of the DM interaction in the system.

We start from the antiferromagnetic Heisenberg model with DM interaction and spin-phonon coupling [17,21,27]

$$H=\sum_l J_l\mathbf{S}_l\cdot\mathbf{S}_{l+1}+\sum_l \mathbf{D}_l\cdot(\mathbf{S}_l\times\mathbf{S}_{l+1})+H_{\rm ph}, \tag{1}$$

where the spin exchange energy $J_l=J[1+\lambda(u_l-u_{l+1})]$, the module of the DM vector $D_l=D[1+\lambda\beta(u_l-u_{l+1})]$, $\mathbf{S}_l\cdot\mathbf{S}_{l+1}=\frac{1}{2}(S_l^+S_{l+1}^-+S_l^-S_{l+1}^+)+S_l^zS_{l+1}^z$, and the phonon energy $H_{\rm ph}=\sum_l((1/2M)p_l^2+\frac{1}{2}Ku_l^2)$. The notations in the model are the same as usual [17]. Though the directions of $\mathbf{D}_l$ are not changed by the dimerization [13], the dependence of the isotropic exchange interaction and the DM interaction on the intersite distance may be different [1], therefore the staggered DM interaction parameter β is introduced to describe the effect of different dependence of DM interaction on the intersite distance. If $\beta=0$ the DM interaction does not depend on the lattice distortion, i.e. DM interaction is uniform, whereas for $\beta=1$ the dependence of DM interaction on the lattice distortion (the staggered DM interaction) is the same as that for the isotropic exchange interaction. Although it was estimated that the DM vector had two components [17] from the specific heat, neutron scattering, and ESR measurement data of copper benzoate, the numerical study by density matrix renormalization group found that a DM vector with only one component gave the best fit to the experimental observations [28]. In view of this as well as for simplification, we choose the vector $\mathbf{D}_l$ to be directed along the z-axis, $\mathbf{D}_l=D_l\mathbf{k}$, After the expansion of the lattice modes u_l by phonon operators b_l, and the Jordan–Wigner transformation with the definition of a bond-dependent mean field $\gamma=\langle c_lc_{l+1}^\dagger\rangle$ [29] (which implies a nearest-neighbor "covalent bonding" of the Jordan–Wigner fermions, analogous to the chemical bonding of the electrons), the Hamiltonian (1) is mapped into spinless Wigner fermions and in momentum space it becomes

$$H=JN\gamma^2+\sum_q\omega_\pi\left(b_q^\dagger b_q+\frac{1}{2}\right)+\sum_k\varepsilon_kc_k^\dagger c_k+\frac{1}{\sqrt{N}}\sum_{k,q}g(k,k+q)(b_{-q}^\dagger+b_q)c_{k+q}^\dagger c_k, \tag{2}$$

where N is the total number of lattice sites, the bare band function of the fermions $\varepsilon_k=J(1+2\gamma)\cos k-D\sin k$, the phonon frequency $\omega_\pi=\sqrt{K/M}$, and the coupling function

$$g(k,k+q)=\mathrm{i}\lambda(2M\omega_\pi)^{-1/2}\{J(1+2\gamma)[\sin k-\sin(k+q)]+D\beta[\cos k-\cos(k+q)]\}. \tag{3}$$

Since ε_k can be rewritten as

$$\varepsilon_k=J\eta\cos(k+\theta), \tag{4}$$

with

$$\theta=\arctan[D/J(1+2\gamma)], \tag{5}$$

$$\eta=\sqrt{(1+2\gamma)^2+(D/J)^2}, \tag{6}$$

the Fermi level is given by the condition $\varepsilon_{k_{\rm F}}=0$, therefore the Fermi wave vector $k_{\rm F}=\pm\pi/2-\theta$. In momentum space, compared with the spin-Peierls system without the DM interaction, the Fermi surface $k_{\rm F}$ shifts by θ as D varies, but the size of the Fermi sea is unchanging. Accordingly, the filling situation of fermions is also unchanging.

To take into account the spin-phonon coupling and the static phonon-staggered ordering due to the dimerization, two unitary transformations are applied to H [30], $\tilde{H}=\exp(R)\exp(S)H\exp(-S)\exp(-R)$, with the generators

$$S=\frac{1}{\sqrt{N}}\sum_{q,k}\frac{g(k,k+q)}{\omega_\pi}(b_{-q}^\dagger-b_q)\delta(k+q,k)c_{k+q}^\dagger c_k, \tag{7}$$

$$R=\sum_l(-1)^lu_0\sqrt{\frac{M\omega_\pi}{2}}(b_l-b_l^\dagger). \tag{8}$$

Here the introduced $\delta(k',k)$ is a function of the energies of the incoming and outgoing fermions in the fermion-phonon scattering process and u_0 is the dimerized lattice displacement ordering parameter. By making the matrix element of the first-order terms in the transformed Hamiltonian between ground state and excited states vanishing, $\delta(k',k)$ can be determined as

$$\delta(k+q,k)=\left(1+\frac{|\varepsilon_{k+q}-\varepsilon_k|}{\omega_\pi}\right)^{-1}. \tag{9}$$

This choice of $\delta(k',k)$ leads the first-order terms of the transformed Hamiltonian to be related only to the higher-lying excited states and should be irrelevant under renormalization. u_0 will be determined below by variational principle.

After averaging the transformed Hamiltonian over the phonon vacuum state we get an effective Hamiltonian for the fermions

$$H_{\rm eff} = JN\gamma^2 + \frac{1}{2}KNu_0^2 + \sum_k E_0(k)c_k^\dagger c_k + \sum_k \mathrm{i}\Delta_0(k)c_{k-\pi}^\dagger c_k - \frac{1}{N}\sum_{q,k,k'} \frac{g(k,k+q)g(k',k'-q)}{\omega_\pi}\delta(k+q,k) \times[2-\delta(k'-q,k')]c_{k+q}^\dagger c_k c_{k'-q}^\dagger c_{k'}, \quad (10)$$

where

$$E_0(k) = \varepsilon_k - \frac{1}{N}\sum_{k'} \frac{g(k',k)g(k,k')}{\omega_\pi^2}\delta(k',k)\delta(k,k')(\varepsilon_k - \varepsilon_{k'}), \quad (11)$$

$$\Delta_0(k) = 2\lambda u_0[J(1+2\gamma)\sin k + D\beta\cos k][1-\delta(k-\pi,k)]. \quad (12)$$

In the adiabatic limit, where $\omega_\pi = 0$, $H_{\rm eff}$ goes back to the adiabatic mean-field Hamiltonian,

$$H_{\rm eff}(\omega_\pi = 0) = JN\gamma^2 + \frac{1}{2}KNu_0^2 + \sum_k \varepsilon_k c_k^\dagger c_k + \sum_k \mathrm{i}2\lambda u_0[J(1+2\gamma)\sin k + D\beta\cos k]c_{k-\pi}^\dagger c_k. \quad (13)$$

It can be diagonalized exactly. Note that in Eq. (10), since $\delta\sim\omega_\pi$, $g\sim\sqrt{\omega_\pi/K}$, the four-fermion term induced by the nonadiabatic effect due to finite phonon frequency goes to zero when $\omega_\pi \to 0$ [see Eq. (13)], therefore, in this case, this term can be treated as a perturbation and the others, being able to be diagonalized exactly, as unperturbed Hamiltonian. By means of the Green's function method to implement the perturbation treatment on the four-fermion term in Eq. (10), we get the renormalized band function and gap function [30]

$$E(k) = E_0(k) - \frac{1}{N}\sum_{k'} \frac{g(k,k')g(k',k)}{\omega_\pi}\delta(k',k)[2-\delta(k,k')] \times \frac{E_0(k')}{\sqrt{E_0^2(k') + \Delta_0^2(k')}}, \quad (14)$$

$$\Delta(k) = 2\lambda u_0[(1+2\gamma)J\sin k + D\beta\cos k][c - d\delta(k-\pi,k)], \quad (15)$$

where

$$c = 1 + \frac{\alpha}{NJ}\sum_{k>0}[J(1+2\gamma)\sin k + D\beta\cos k]^2 \times[1-\delta(k-\pi,k)]\delta(k-\pi,k)[E_0^2(k) + \Delta_0^2(k)]^{-1/2}, \quad (16)$$

$$d = 1 - \frac{\alpha}{NJ}\sum_{k>0}[J(1+2\gamma)\sin k + D\beta\cos k]^2 \times[1-\delta(k-\pi,k)]^2\left[E_0^2(k) + \Delta_0^2(k)\right]^{-1/2}, \quad (17)$$

and $\alpha = 2\lambda^2 J/K$ is the spin-phonon coupling constant. The fermionic spectrum in the gapped state is given by $W(k) = \sqrt{E^2(k) + \Delta^2(k)}$.

By variational principle to minimize the ground state energy $E_g = \langle g|H_{\rm eff}|g\rangle$, the dimerized lattice displacement ordering parameter u_0 is determined by

$$1 = \frac{2\alpha}{NJ}\sum_{k>0}[J(1+2\gamma)\sin k + D\beta\cos k]^2 \times[1-\delta(k-\pi,k)]\frac{c - d\delta(k-\pi,k)}{\sqrt{E^2(k) + \Delta^2(k)}}. \quad (18)$$

The nearest-neighbor covalent-bonding $\gamma = (1/N)\sum_{k>0}\cos k(E(k)/W(k))$. When $\lambda = 0$, $\gamma = (\pi\eta - 2)^{-1}$, and in this case, if $D = 0$ also, the Hamiltonian (1) reduces to Heisenberg model and γ becomes the same as Wang's $\gamma = 1/\pi$ [29].

For investigation on the nonadiabatic effect of DM interaction on the dimerization, we calculate the phonon-staggered ordering parameter

$$m_p = \frac{1}{N}\sum_l (-1)^l\langle u_l\rangle = \frac{1}{N}\sum_{k>0}\frac{2\lambda}{K}[J(1+2\gamma)\sin k + D\beta\cos k]\frac{\Delta(k)}{W(k)}, \quad (19)$$

and the spin-dimer order parameter

$$m_s = \frac{1}{N}\sum_l (-1)^l\langle \mathbf{S}_l\cdot\mathbf{S}_{l+1}\rangle = \frac{1}{N}\sum_{k>0}(1+2\gamma)\sin k\frac{\Delta(k)}{W(k)}. \quad (20)$$

The effects of the staggered and the uniform DM interactions on the dimerization can be clearly identified by defining the staggered and the uniform DM interaction order parameters,

$$D_{\rm stag} = \frac{1}{N}\sum_l (-1)^l\langle \mathbf{k}\cdot(\mathbf{S}_l\times\mathbf{S}_{l+1})\rangle = \frac{1}{N}\sum_{k>0}\cos k\frac{\Delta(k)}{W(k)}, \quad (21)$$

$$D_{\rm unif} = \frac{1}{N}\sum_l \langle \mathbf{k}\cdot(\mathbf{S}_l\times\mathbf{S}_{l+1})\rangle = \frac{1}{N}\sum_{k>0}\sin k\frac{E(k)}{W(k)}. \quad (22)$$

Thus, we get the relation between the phonon-staggered ordering parameter, the spin-dimer order parameter and the staggered DM interaction order parameter,

$$\lambda m_p = \alpha\left(m_s + \frac{\beta D}{J}D_{\rm stag}\right), \quad (23)$$

which gives clearly the contributing weights of m_s and $D_{\rm stag}$ to the lattice dimerization. If there is no spin-phonon coupling $\lambda = 0, m_p = m_s = D_{\rm stag} = 0$ and

$$D_{\rm unif} = -\frac{\sin\theta}{\pi} = -\frac{D}{\pi\sqrt{[J(+2\gamma)]^2 + D^2}}. \quad (24)$$

When $0<\beta\leqslant 1$, the DM interaction includes both the uniform and the staggered components. Fig. 1(A)–(C) show $D_{\rm stag}$, m_s and λm_p, respectively, as functions of D/J in the cases of $\alpha=1.0$ and $\omega_\pi/J=0.01$ with different staggered DM interaction parameters β from zero to 1. When $D=0$, i.e., there exists no DM interaction, the spin-dimer order parameter takes its maximum value and the staggered DM interaction order parameter the minimum value. As β or D increases, $D_{\rm stag}$ increases but m_s decreases, which indicates the effect of the DM interaction is to decrease the spin dimerization. The competition result between $D_{\rm stag}$ and m_s determines the lattice dimerization ordering parameters m_p to be increasing for large β and decreasing for small β. Obviously, there exists a finite threshold value of β and when β increases to cross this value, the effect of the DM interaction on the lattice dimerization changes from suppression to promotion. The threshold value β_c can be obtained by letting the variation $\partial m_p/\partial D=0$ ($D\neq 0$), and in the nonadiabatic case, β_c is determined by a set of values of D, λ and ω_π. If β_c was simply a constant as previous works predicted [21], λm_p would not change with D when $\beta=\beta_c$. However, in view of the determination of m_p by D, β, λ and ω_π, one might wonder why the DM interaction should have no effect on dimerization when $\beta=\beta_c$. Our calculation indicates that β_c is not simply a constant but a crossover. The change of λm_p with D/J in the region of this crossover is presented in Fig. 1(D) for $\beta=0.54$, 0.56, 0.58 and 0.60.

The DOS of the Wigner fermions is

$$\rho(\omega)=\frac{1}{N}\sum_k \delta[\omega-W(k)]=\frac{1}{2\pi}\left(\left.\frac{{\rm d}W(k)}{{\rm d}k}\right|_{k=f(\omega)}\right)^{-1}, \quad (25)$$

where, $k=f(\omega)$ is the inverse function of the excitation energy $\omega=\sqrt{E^2(k)+\Delta^2(k)}$. Fig. 2 shows the calculated DOS for $\alpha=0.4$ and $\omega_\pi/J=0.01$ with different β and D. One can see that a nonzero DOS starts from the spin dimerization gap edge and there exists a peak above the gap edge with a significant tail between it and the true spin gap edge. As D/J increases, the peak height of DOS decreases and the peak position $\omega_{\rm peak}$ shifts to lower excitation energy for smaller β but contrary to that for

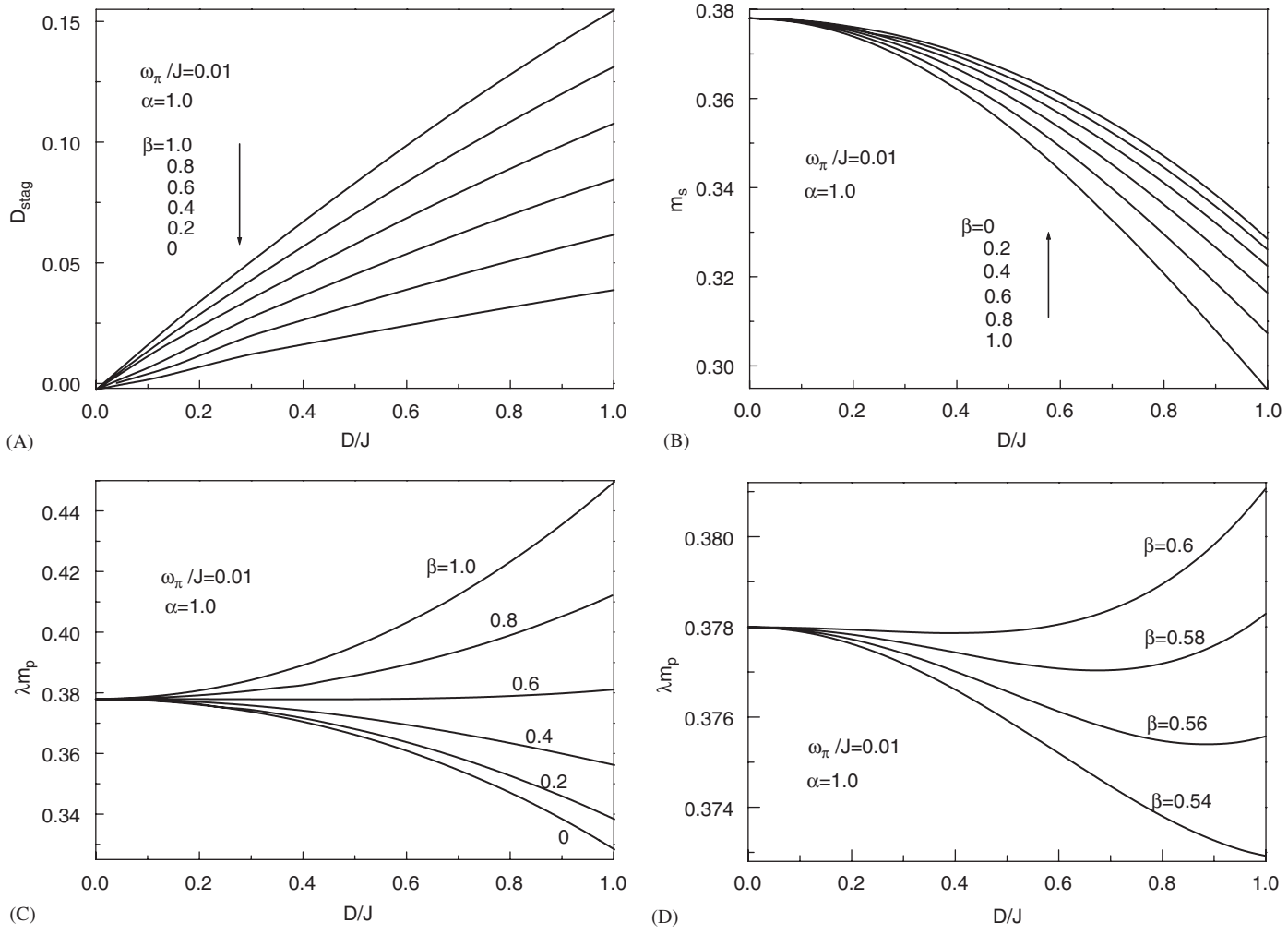

Fig. 1. (A) The $D_{\rm stag}$, (B) m_s and (C) λm_p, respectively, as functions of D/J in the cases of $\alpha=1.0$ and $\omega_\pi/J=0.01$ with different staggered DM interaction parameters β from zero to 1. (D) The change of λm_p with D/J for $\beta=0.54$, 0.56, 0.58 and 0.60.

larger β. A full view of ω_{peak} as functions of D/J for wider range of β is plotted in Fig. 3(A). Fig. 3(B) illustrates the ω_{peak} as functions of D/J for β in the crossover. As shown in the figure, for certain α and ω_π/J, the crossover has its bottom boundary β_{bot} and top one β_{top}. When β is smaller (larger) than β_{bot} (β_{top}), ω_{peak}/J decreases (increases) monotonously as D/J increases, while when $\beta_{bot}<\beta<\beta_{top}$, as D/J increases ω_{peak}/J decreases at first until it reaches its minimum at a definite value of D/J, and thereafter increases. For appropriate fixed value of α and small β, ω_{peak}/J decreases as D/J increases and goes to zero at a critical value $(D/J)_c$ which indicates the spin gapped state is destroyed. If β is large, as D/J increases ω_{peak}/J keeps zero at first and at a critical value of D/J changes from zero to finite and then increases, in other words, the system undergoes a phase transition from spin gapless state to gapped state. These processes are illustrated in Fig. 3(C).

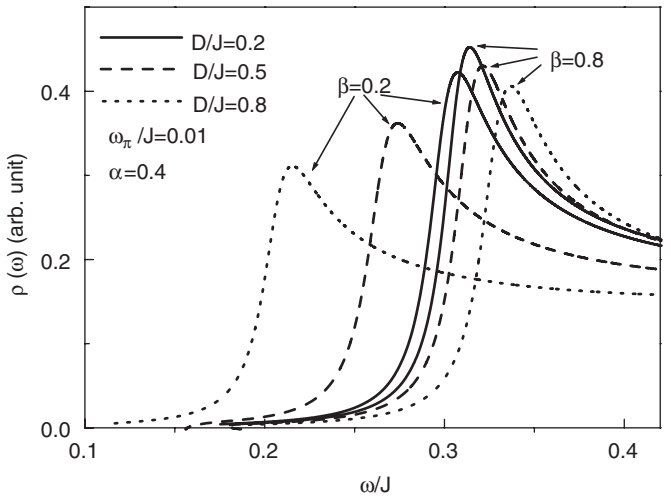

Fig. 2. The DOS for $\alpha = 0.4$ and $\omega_\pi/J = 0.01$ with different β and D.

Substituting $u_0 = 0$ into Eq. (18), we get the self-consistent equation of phase-transition points

$$1 = \frac{2\alpha}{NJ}\sum_{k>0}[J(1+2\gamma)\sin k + D\beta\cos k]^2[1-\delta(k-\pi,k)] \times \frac{c - d\delta(k-\pi,k)}{|E(k)|}. \quad (26)$$

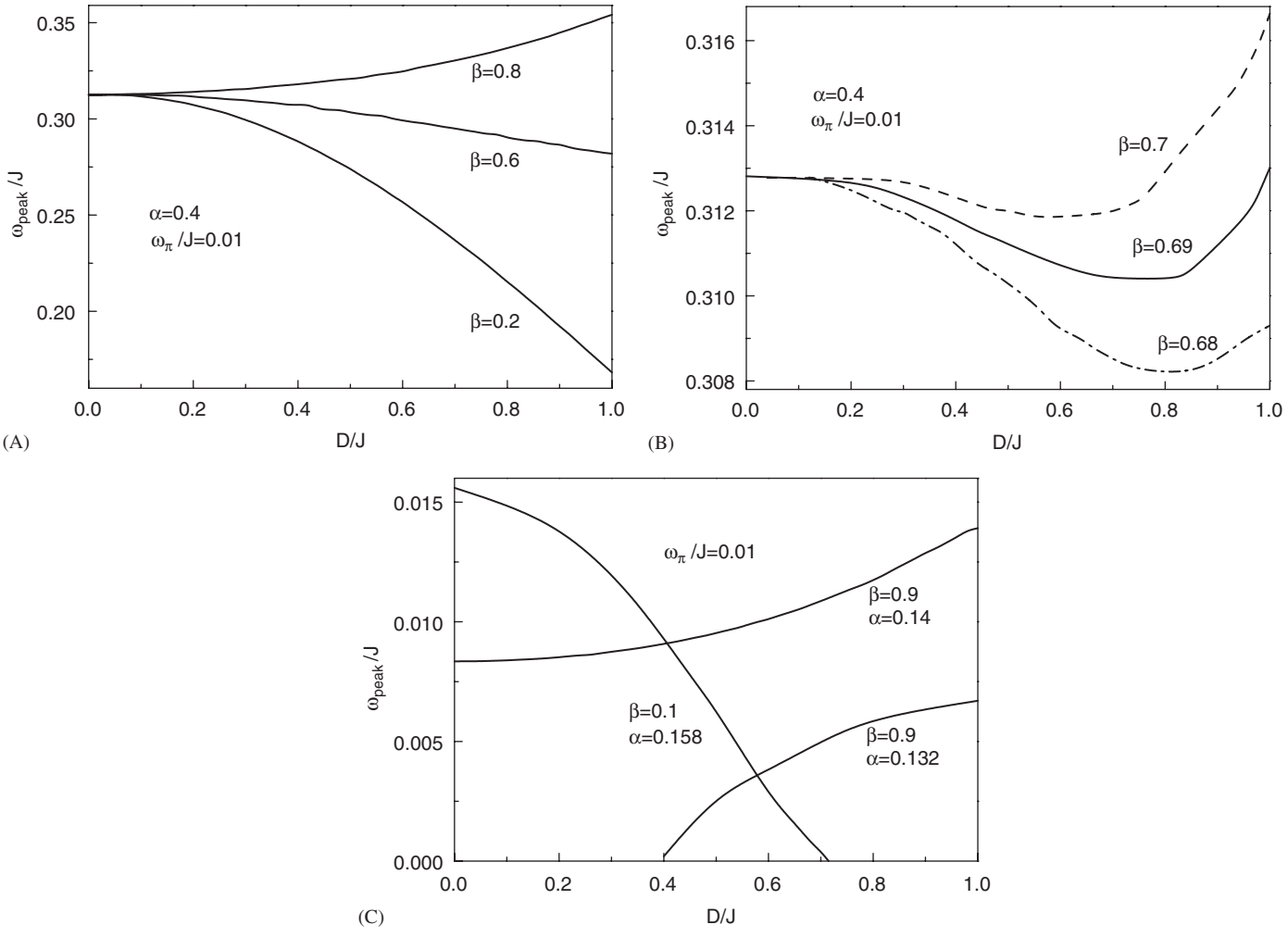

Fig. 3. (A) The ω_{peak} as functions of D/J in the cases of $\alpha = 0.4$ and $\omega_\pi/J = 0.01$ for wider range of β. (B) ω_{peak} as functions of D/J for β in the crossover $\beta = 0.7$, 0.69 and 0.68. (C) ω_{peak} as functions of D/J with $\omega_\pi/J = 0.01$ for different appropriate fixed values of α and β.

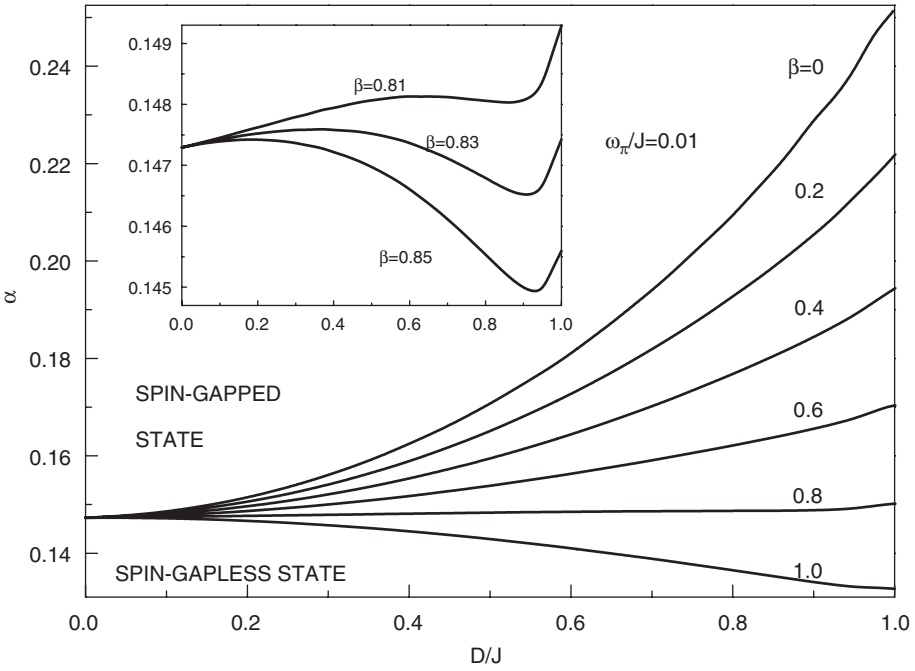

Fig. 4. The phase diagram in the $\alpha\sim D/J$ plane for $\omega_\pi/J = 0.01$ with β varying in whole range. Inset: a zoom in view of the phase diagram in the crossover region.

Fig. 4 shows the phase diagram in the $\alpha\sim D/J$ plane for $\omega_\pi/J = 0.01$ with β varying in whole range. The increase in D/J leads the phase boundary to move to larger spin-phonon coupling when β is small, but to smaller one when β is large. The change of β strongly influences the phase boundary. The inset is a zoom in view of the phase diagram in the crossover region. It indicates that for appropriate fixed values of α and β the system can reenter between the spin gapless and the spin gapped states as D increases.

Within the adiabatic approximation, the calculated DOS of the dimerized spin-phonon systems has an inverse-square-root edge singularity and there is no state inside the gap edge, which is discrepant to experiments. In observations, the singularity is absent, and there is a significant tail below the maximum of DOS. In our theory, by considering the nonadiabatic effect due to the finite phonon frequency, the Fermi surface is smeared and the singularity related to the infrared divergence of the Fermi edge effect is eliminated, which is consistent with the measurement of optical absorption spectrum in quasi-one-dimensional spin-Peierls systems. By using the Green's function method to implement the perturbation treatment, our results are more suitable in the small ω_π regime, which is theoretically and experimentally significant since, from the view point of experiment, for quite a lot of realistic cases the energy of quantum phonon ω_π is small. In the perturbation study, we use the spin-phonon coupling λ as the perturbation order parameter and the coefficient of the four-fermion term in the effective Hamiltonian (10) is proportional to $2\lambda^2 J\omega_\pi/K$, therefore, $2\lambda^2 J\omega_\pi/K = \alpha\omega_\pi$ should be small enough to ensure that the perturbation treatment is appropriate at nonadiabatic case.

In conclusion, the effects of DM interaction in the antiferromagnetic Heisenberg model with spin-phonon coupling have been studied through a nonadiabatic analytical approach and the phonon-staggered ordering parameter, the DOS, and the phase diagram of the system are derived. The results show that the effect of the DM interaction is to increase the staggered antisymmetric spin exchange interaction but to decrease the spin dimerization and their competitions result in the lattice dimerization ordering parameter m_p to be increase for large β and decrease for small β. A crossover of β exists and the system can reenter between the spin gapless and the spin gapped states.

Acknowledgments

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