Quantum spin liquid in antiferromagnetic chain $S=1/2$ with Acoustic Phonons

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A spin and phonon excitations spectrum are studied using quantum Monte Carlo method in antiferromagnetic chain with spins $S = 1/2$ coupled nonadiabaticity with acoustic phonons. It is found the critical coupling exists to open gap in the triplet excitation spectrum for any phonon velocity. The phase boundaries of delocalized phonons and propagated the bound states of magnon and a phonon are calculated. It is shown that the spherical symmetry of the spin-spin correlation functions is broken. The magnetic and optical properties $CuGeO_3$ are explained without using spin-Peierls transition.

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The existence and stability of multiquanta bound states are now established for a wide variety models with prescribed nonlinearity and exhibited some distinctive observable signatures in terms of spatiotemporal correlations. The typical origin of effective nonlinearity in quantum system is the coupling of two or more fields. Adiabatic slaving of fields usually results in nonlinear Schrödinger models. Realistically nonadiabatic effects must be considered and the influences of nonlinearity and nonadiabaticity are inevitably interrelated. Here we consider spin-phonon coupled model frequently used to describe the opening of a singlet-triplet spin gap and an isotropic drop of the magnetic susceptibility below critical temperature. As a rule a optical phonon interacts with spin system and lead to lattice instability. The interaction between acoustic phonons and spins has been studied in two limit cases. A weak coupling lead to a renomalization of the phonon mode and give rise to four spin interaction. As a result of a gap is opened but the other well-defined magnetic and phonon bound states has been loosed. A strong coupling dos’t correspond to real compounds and the results obtained in the intermediate coupling are incorrect.

A magnetic, elastic and optical properties in the inorganic quasi-one-dimensional $CuGeO_3$ compound fail to describe in terms of one model. The thorough structure measurements [1] and Cu-nuclear quadrupole resonance experiments [2] have not been found a lattice dimerization and a softening of optical phonons. The static distortions below critical temperature $T_c = 14K$ cannot be ascribed to any pseudomode in the high-symmetry phase [3]. The magneto induced structural effects depend on the used frequency, so strong lattice fluctuations have been observed by electron diffractions up to $T \sim 100K$ [4], by optical methods up to $T \sim 200K$ [5]. A frustration interaction due to a asymmetric or a next nearest- neighbors antiferromagnetic exchange can cause a gap in the triplet excitation spectrum though no experimental proves have existence of these exchanges. The physical nature of the critical temperature existence and opening gap in triplet excitation spectrum, the Raman spectrum, thermal conductivity in magnetic field remains unclear.

Numerical methods, such as exact diagonalization and density-matrix renormalization group, face the potential difficulty in dealing with a very large Hilbert space of the phonons. Here,
we use Monte Carlo approaches restricted to finite chains $L = 100, 200$ but without any adiabatic approximation and the truncation of the infinite phonon Hilbert space. The method is based on a path-integral representation for discrete system in which we work directly in the Euclidean time continuum. All the configuration update procedures contain no small parameters. Being based on local updates only, it allows to work with the grand canonical ensemble and to calculate any dynamical correlation function, expectation values.

We consider a model Hamiltonian of a spin-phonon system:

$$H = \sum_{i=1}^{L} [\mathcal{J} + \alpha(u_i - u_{i+1})][S_z^i S_{z+1}^i + (S_{z+1}^i S_{z+1}^i + S_{z+1}^i S_z^i)]/2 + M\dot{u}_i^2/2 + K(u_i - u_{i+1})^2/2. \quad (1)$$

Here $S_z^{\pm}$ are a spin operator components associated with the site $i$, $J > 0$ is the usual antiferromagnetic exchange integral, $\alpha$ is the spin-phonon coupling constant, $u_i$ is the displacement in the $z$-direction, $M$ is the mass of the ion and $K$ the spring constant. Using the quantum representation for phonon operators $b, b^+$, the Hamiltonian becomes

$$H = \sum_q \sum_{i=1}^{L} [\mathcal{J} + \alpha \sqrt{2\hbar/M}\omega_0 \sin (q(i - 0.5))(b_q + b_q^+)] [S_z^i S_{z+1}^i + (S_{z+1}^i S_{z+1}^i + S_{z+1}^i S_z^i)]/2 + \sum_q \hbar\Omega(q)b_q^+ b_q, \quad \Omega(q) = 2\sqrt{K/M} \sin (q/2), \quad \omega_0 = 2\sqrt{K/M}, \quad q = 2\pi n/L, n = 1, 2, \ldots L. \quad (2)$$

The phonon frequency, spin-phonon coupling, energy, temperature are normalized on the exchange $J$. The temperature used in calculation is $\beta = J/T = 25$. Our system consists of the two subsystem interacted. The elastic subsystem is described by phonons with the number of occupation $n_{ph} = 0, 1, 2, \ldots$ and magnetic subsystem is characterized by the number occupation $n_m = 0, 1$ and Pauli operators $a, a^+$ which coincide with $S^\pm$ spin operators. We start with the standard Green function of the phonon in the momentum $q$-imaginary-time $\tau$ representation:

$$G(q, \tau) = \sum_\nu | < \nu|b_q^+|\text{vac}>|^2 \exp [- (E_\nu(q) - E_0)\tau], \quad (3)$$

where $|\nu>$ is a complete set of the Hamiltonian $H$ in the sector of given $q$, $H|\nu(q) > = E_\nu(q)|\nu(q) >$, $H|\text{vac}> = E_0|\text{vac}>$, $E_0 = 0$. Rewriting Eq.(3) as

$$G(q, \tau) = \int_0^\infty d\omega A(q, \omega) \exp - (\omega\tau), A(q, \omega) = \sum_\nu \delta (\omega - E_\nu(q))| < \nu|b_q^+|\text{vac}>|^2 \quad (4)$$

one defines the spectral function $A(q, \omega)$. We calculate a one-particle spin Green function

$$< a(\tau)a^+(0) >= \int_0^\infty d\omega \rho_1(\omega) \exp - (\omega\tau), \quad (5)$$

where $\rho_1(\omega)$ -spectral density function, and two-particle spin Green function associated with singlet excitation $\Delta S^z = 0$

$$< a_0(\tau)a_1^+(\tau)a_1(0)a_0^+(0) >= \int_0^\infty d\omega \rho_s(\omega) \exp - (\omega\tau). \quad (6)$$

Correlation functions are determined on the basis of a complete set of eigenstates of the Hamiltonian:

$$<O> = \frac{\sum_\nu <\nu|O|\nu>}{\sum_\nu <\nu|\nu>}, \quad (7)$$
where \( O = (a_0 a_1^+ + a_0^+ a_1), (b_q + b_q^+) S_0^z S_i^z, (a_0 a_1^+ + a_0^+ a_1)(b_q + b_q^+) \). The longitudinal spin-phonon correlation function \( < S_i^z S_{i+h}^z > \), the phonon density- density \( < n_{ph}(q)n_{ph}(q+p) > \), the distribution of phonons number \( n_{ph}(q) \) and magnons number \( n_m(k) \) as a function of momentum are simulated.

Energies excitation of the two-particles \( S_k^+ S_{k-p}^-, p = \pi \) and one- particle excitation differ by less than \( \sim 1\% \) in the isotropic antiferromagnetic chains. The interaction with the elastic system lead to qualitatively various dispersion relation. The two-particle excitation spectrum became gapped and one-particle excitation gapless for \( \alpha < \alpha_{c1} \). The gap energy determined from spectral density well fit on the linear dependence \( \Delta_s(\alpha) \sim \frac{1}{2} k_0 \), \( \Delta < J \).

For small spin-phonon coupling \( \alpha < \alpha_{c1} \) the phonons are created near the upper bound of triplet excitation band \( W_t \) with momentum \( q = 2 \arcsin \frac{W_t}{\omega_0} \). It follow from the distribution of the phonon number versus momentum and the spectral density. The spins number in the structure of the spin-phonon quasiparticle \( b_{q-k_1-k_2-...k_n}^+ S_{k_1}^z S_{k_2}^z ... S_{k_n}^z, \gamma = z, \pm \) increase and the correlation function between spin and phonon

\[
R_{bs} = \sqrt{\frac{2\hbar}{M}} < b_q^+ b_q \sqrt{\frac{\sin \frac{q}{2} \cos (q(i-0.5))}{\sin \frac{q}{2}}} S_i^z S_{i+1}^z >,
\]

qualitatively confirms it as illustrated in Fig.1a . For \( \alpha < \alpha_{c2} \) the correlation function is parameterized as \( R_{bs} \sim \frac{\alpha}{\omega_0}, \omega_0 > 3 \). \( R_{bs} \sim \frac{\alpha}{2W_t}, \omega_0 \leq 3 \). Correlation function between the longitudinal acoustic phonon and the transverse spin components \( < (b_i^+ + b) a_i^+ a_i > \) is less by a factor 3-5 than \( R_{bs} \). Number of two-particle singlet excitation falls as to an exponent \( N_s \sim \frac{1}{\alpha} \exp [\frac{3a}{2\alpha_2}] \). The formation of the bound states and nonlinear excitations causes to break the spherical symmetry of the spin-spin correlation function. The ratio of the correlation functions of transverse spin components to longitudinal is parameterized as the linear dependence on the spin-phonon coupling

\[
\frac{[< S_0^+ S_1^- > - 2]}{< S_0^z S_1^z >} \sim \frac{5, 2\alpha}{\omega_0}, \alpha < \alpha_{c2},
\]

as presented in Fig.1b. The magnetic susceptibility is represented by spin-spin correlation function when the long-range order is absent \( \chi_{xy}/\chi_{zz} = \sum_r S_0^+ S_r^- / \sum_r S_0^z S_r^z \). Conventionally the physical nature of anisotropy result from spin-orbital interaction for spin \( S = 1/2 \).

We suggest a new mechanism the dynamical interaction between the longitudinal acoustic phonon and spin. It is important for S- ions.

For \( \alpha > \alpha_{c1} \) the gap in the triplet excitation spectrum simulated from the spectral density function is open . The functions \( \rho \) are plotted in Fig.2. The gaps energies fit satisfactory on the straight line \( \Delta \sim 0.8(9)(\alpha - \alpha_{c1}) \) in the range of \( \alpha_{c1} < \alpha < \alpha_{c2} \). Here exists the bound states of spin-phonon quasiparticles similar to bipolaron with the symbolic kind \( < b_q^+ \sum_{i_1 k_i^1} b_{q+p}^+ \sum_{i_2 k_i^2} \Pi^{n_1+n_2}_i S_{k_i}^z >, \) the correlation function of the phonon number \( < N_{ph}(q)N_{ph}(q+p) > \), presented in Fig.3b is nonzero at the certain wave number \( p \) for \( \alpha \approx \alpha_{c1} \) and for all \( p \) at the condition \( \alpha > \alpha_{c2} \). The magnon distribution function demonstrates a small oscillations approximately within a five percents as compared to \( n_m(q = 0, \pi) \approx 0.22 \) for \( \alpha > \alpha_{c1} \) and the number phonon distribution function reveals four local maxima for \( \alpha > \alpha_{c2} \) and one sharp maximum for \( \alpha > \alpha_{c3} \). Analysis of the spin- spin correlation function plotted in Fig.3 lead to two important length scales:

\[
| < S_0^z S_r^z > | \sim \frac{A}{r} \exp (r/\xi), r < r_c, < S_0^z S_r^z > \sim (1)^r \frac{\cos (Qr) \cos (qr)}{r^q}, r > r_c.
\]
where \( \xi \) correlation radius, \( Q,q,\theta \) fitted parameters. \( R \sim \pi/Q \) may be interpreted as the typical size of a cloud of spins and phonons; \( \xi \) may be considered as the width of the transition region separating the nearest spin-phonon quasiparticles. These coherent nonlinear excitations cause the local extremums of the spectral density phonons and spin excitations at the same energies including the spin gap range as shown in Fig.2. It is possibly a standing vibration with the spin kink in sites exists here. The disjoint vibrations are performed at the condition that the wave length is changed in two times \( l/a = 2;4;8;16; \ldots \) \( c \ k = \pi/l \), \( E = w_0 sin(k/l/2) \). These estimates of energy are in good agreement with Monte Carlo results up to \( n = 4 \) because the temperature fluctuations (\( T = 0.04 \)), \( \xi < l/a = 64 \) cut the kinks interaction radius. The average number of phonons increase vs. \( \alpha \) according to the power law
\[
N_{av} = 0, 0012\left(\frac{\alpha}{\alpha_{c2}}\right)^{1.75(6)}, v_{ph} \leq v_m; \quad N_{av} = 0, 0006\left(\frac{\alpha}{\alpha_{c2}}\right)^{2.4(6)}, v_{ph} > v_m, \tag{11}
\]
here \( v_{ph} \) and \( v_m \) is the velocity of phonon and magnon. The spectral density of phonons and the distribution of phonon number became continuum at \( \alpha = \alpha_{c2} \). The value \( \alpha_{c2} \) is similar to critical concentration when dressed phonons effectively ”percolate” along the three low-lying bands. The correlation function \( \langle N_{ph}(q)N_{ph}(q+p) \rangle \) is not equal to zero for all momentums as shown in Fig.3. The transition from one state with localized phonons to delocalized is like to Anderson’s transition in disordered systems. The width of band increases versus \( \alpha \) as \( W_{ph}(\alpha_{c2})/W_{ph}(\alpha_{c1}) \sim 1.5 \). When the spin-phonon coupling is compared to the energy of the boundary of top band of phonon and triplet spin excitation \( \alpha > \alpha_{c3} \sim 0.8w_0 \) the soliton lattice is formed with the wave vector of structure being in the range of \( \pi < Q < 2\pi \). A nonlinear excitation, for example, the spin breather is gapless and the spectral density of phonons and spin excitations are comparable values at the low energies. The gap in one-particle spectrum excitation pass into quasigap. The area of parameters in the plane the energy of top band of acoustic phonon \( \omega_0 \) - the value of the spin-phonon coupling \( \alpha \) , where a gap in the one-particle excitation spectrum is arisen, is limited by two fitting lines
\[
\alpha_{c1} \sim \frac{\omega_0}{4w}, \quad \text{and} \quad \alpha_{c3} \sim 0.8w_0. \quad \text{The complete filling of the three acoustic bands occurs at the}
\alpha_{c2} \sim \frac{\omega_0}{2w}. \tag{11}
\]
Now we will apply our results for interpretation and prediction of new effects for the one-dimensional spin system \( CuGeO_3 \) with intrachain exchange \( J_{Ge} = 120K, J_1/J_{Ge} = 0.1 \). The crystal structure \( CuSiO_3 \) is isostructural to \( CuGeO_3 \). However \( CuSiO_3 \) is antiferromagnet with \( J_{Si} = 21K, J_1/J_{Si} = 0.14 \) (\( J_1 \) - interchain exchange) \( \text{[8]} \). The interaction between spins and acoustic phonon mode in chain leads to gap in the triplet excitation spectrum \( CuGeO_3 \). For the same value of the spin - phonon coupling is \( \left[ \omega_0/J \right]_{Si} \gg \left[ \omega_0/J \right]_{Ge} \) and \( \alpha_{c1,Si} > \alpha_{c1,Ge} \). The magnon velocity \( CuGeO_3 \) is \( v_m \sim 1300m/s \), the phonon velocity depends on the ultrasound frequency \( \text{[7]} \) and we take the average value \( v_{ph} \sim 5000m/s \) \( v_{ph} = \frac{\omega_0}{w} \). The anisotropic elastic constants are \( c_{11} = 64GPa, c_{22} = 37.6GPa, c_{33} = 317.3GPa \) \( \text{[7]} \) and confirm validity using one dimensional model to study the dynamic of the interacting magnetic and elastic systems. According to our results the spin-phonon coupling in \( CuGeO_3 \) has been established \( \alpha \approx 2.2 \) for \( \Delta/J = 0.2, \omega_0 = 12 \). The estimated energies of the bound spin-phonon excitation are \( E_{sph} = 95cm^{-1}, 48cm^{-1}, 24.4cm^{-1}, 12.5cm^{-1}, 6.2cm^{-1}, 3cm^{-1} \) and are in general agreement with Raman spectrum \( E_{sph}^{ex} = 98cm^{-1}, 48cm^{-1}, 30cm^{-1}, 13.2cm^{-1} \) \( \text{[1]} \). The localized phonon excitation in the second \( E_{2}^{MC} = 0.22eV \) and in the third \( E_{3}^{MC} = 0.34eV \) bands agree satisfactorily with data of the optical absorption \( E_{2}^{ex} = 0.21eV \) \( \text{[1]} \), \( E_{3}^{ex} = 0.36eV \) \( \text{[13]} \) and make clear the physical origin of these peculiarities. Up to present time the existence of isolated energy level \( E_{ex}^{ex} \sim 6cm^{-1} \) below the triplet gap
has been remained unclear. This gap is found by the inelastic neutron scattering [14] and the submillimiter resonance [15]. We predict a new resonances at the frequencies $\omega \sim 130\text{GHz}; \sim 65\text{GHz}$ at low temperatures $T < 1K$.

The bound spin-phonon excitations will result in additional phonon scattering which depends on magnetic field. The energy of spin-phonon quasiparticle normalized on gap value $E^{MC}/\Delta = 0.37; 1.5$ is well agreement with temperatures normalized on critical temperature $T_c = 14K$, $T^{ex}/T_c \simeq 0.39; 1.57$ related to local maxima of thermal conductivity [16]. This effect result from interaction between elastic and magnetic system and magnetostriction parameter exhibits also two maximum at the same temperatures [17]. The relationship of transverse spin-spin correlation function to longitudinal $[<S^+_0S^-_1> - 2] \simeq 0,9$ is associated with the anisotropy exchange $(J_b/J_c)^{MC} = 1, 45$ which agrees with experimental data $J_b/J_c = \Theta_b/\Theta_c = 1,38$ [18]. The sharp rise of $g-$ factor near the transition $T > T_s$ may be explained in term of formation of the coherent state of spin-phonon quasiparticles which may be broken by thermal phonons. The average number of phonons resulted in spin-phonon interaction $< N_0 > = 1,7 \cdot 10^{-4}$ in $CuGeO_3$ is equal to number of thermal phonons define in terms of Debye approximation with Debye temperature $\Theta = 330K$ [19] at the temperature $T = 18K$.

In conclusion, we summarize the main results. The interaction between magnetic and elastic system lead to gap in one-particle and two-particle excitation spectrum. The gapped triplet spectrum arises at the certain critical value of the spin-phonon coupling. Bound spin-phonon excitation localize for $\alpha < \alpha_{c3}$ and causes a peculiarities set of spectral density of phonon and spin excitations. Spherical symmetry between transverse and longitudinal spin-spin correlation function is broken. The triplet gap in $CuGeO_3$ arises from the interaction between spin and acoustic phonon and this compound has not possessed spin-Peierls ordering. From our results the magnetic, elastic and optic properties $CuGeO_3$ is explained.

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Fig.1 The correlation function of the nearest-neighbor spin-spin and displacement $< A_q (b_i^+ + b_q) S_i^z S_{i+1}^z >$, where $A_q = \sqrt{\frac{2\pi}{M}} \sqrt{\sin \frac{q}{2}} \cos (q(i - 0.5))$ (a) and ratio transverse spin-spin correlation function to longitudinal (b) with $\omega_0 = 10(1), 8(2), 6(3), 1(4)$ versus spin-phonon coupling.

Fig.2 The spectral density one-particle excitations with $\omega_0 = 8, \alpha = 0.4(1), 0.2(2), 3.2(3)(a), \alpha = 1.6(1), 2(2)(b)$; and two-particle singlet excitations with $\omega_0 = 8, \alpha = 2.6(2), 3.2(1)(c)$ and phonon excitations with $\omega_0 = 8, \alpha = 2(1), 3.2(2)(d)$.

Fig.3 Longitudinal spin-spin correlation function $v_r r$ with $\omega_0 = 8, \alpha = 0.8(1), 2(2), 3.2(3)(a)$. Correlation function of the phonon number normalized on its maximum value versus momentum with $\omega_0 = 8, \alpha = 1.4(1), 2(2), 2.6(3)(b)$. 
