Néel temperature for undoped spin-Peierls quasi-one-dimensional model

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Abstract
It is usually believed that Néel and spin-Peierls orderings cannot co-exist. Very recently a transition to an ordered magnetic Néel state was observed in a pure organic spin-Peierls system p-CyDOV, and in one doped with p-CyDTV. While known theories can describe the onset of a Néel ordering in spin-Peierls compounds by the influence of impurities (e.g., in doped $CuGeO_3$), they exclude the possibility of onset of a Néel state in spin-Peierls pure systems. We propose a simple theoretical model, which manifests the onset of a Néel ordering in a spin-Peierls ordered state in the ground state and for nonzero temperatures, and calculate critical temperatures for phase transitions in those states.

The spin-Peierls transition, as one of the most interesting phenomena in physics of low-dimensional quantum spin systems, has attracted much interest of theorists and experimentalists during recent years. The main feature of such a phase transition is the interaction between a one-dimensional (1D) quantum antiferromagnetic spin chain and a 3D crystal lattice. Because of such an interaction it is possible to speak about a nonzero temperature of such a phase transition, even for a 1D spin system with short-range spin–spin interactions (in which, according to the Mermin–Wagner theorem [1], no magnetic ordering can exist for $T \neq 0$). The order parameter for a spin-Peierls transition is, in fact, the alternating displacements of magnetic ions along the chain, which belong to the 3D lattice. For a magnetic (1D) subsystem such displacements produce an alternating exchange between neighbouring spins along the chain direction, which, in turn, causes the onset of a spin gap for low-lying excitations of an antiferromagnetic quantum spin chain (whose ground state is a nonmagnetic singlet with or without alternations).

It is usually believed (see, for example, [2]) that the consequence of the singlet ground state and the presence of the gap is that no long-range magnetic ordering should exist in the ideal system, even at $T = 0$. However, for doped (non-ideal) systems an antiferromagnetic order was observed in the spin-Peierls compound $CuGeO_3$ doped with Si, or metallic ions,

like Zn, Mg, or Ni impurities [3]. Several theories were proposed to describe such a transition to a magnetically ordered state in a doped spin-Peierls system [4–6]. It is stated in [4, 6] for $T = 0$ and in [5] for both the ground state and nonzero temperatures, that a spin-Peierls ordering can co-exist with an antiferromagnetic Néel ordering in the presence of disordered impurities, while for a pure system, in the absence of disorder, the spin-Peierls phase and antiferromagnetic Néel state cannot co-exist. Earlier theoretical studies of undoped spin-$\frac{1}{2}$ antiferromagnetic systems [7] also stated that in the ground state the spin-Peierls state and Néel state exist and are stable for different ranges of parameters.

Recently it has been observed that the magnetic susceptibility of an organic compound 3-(4-cyanophenyl)-1,5-dimethyl-6-oxoverdazyl (abbreviated as p-CyDOV) with spin $S = \frac{1}{2}$ reveals the characteristic for spin-Peierls system behaviour [8]. The temperature dependence exhibits a broad maximum at 54 K. Such a behaviour can be well described within a one-dimensional antiferromagnetic spin-$\frac{1}{2}$ chain [9, 10] with the exchange constant $J = 42.1$ K. On the other hand, the susceptibility abruptly decreases at approximately 15 K, characteristic for spin-Peierls system [11–13]. Finally, below 5.6 K a small increase in susceptibility was observed. First, that upturn was attributed to isolated monoradicals (impurities), or broken chain effects (free edges of a spin chain, cf [14]). However, very recently the specific heat study of that compound (the pure one, and one doped with similar paramagnetic radicals 3-(4-cyanophenyl)-1,5-dimethyl-6-thioxoverdazyl, p-CyDTV with $S = \frac{1}{2}$) was performed [15]. That investigation found a sharp peak in the low-temperature behaviour of the magnetic specific heat at low temperatures for both doped and pure p-CyDOV (for the latter at 0.135 K), and a broad hump (for the pure compound at 5.6 K). The authors of [15] concluded that the system undergoes a phase transition to the Néel (magnetically ordered) state at the temperature of the sharp peak. Naturally, the presence of a sharp peak in the temperature dependence of the (magnetic) specific heat cannot be explained by minute impurities, or free edges of spin chains for undoped crystal. Such a conclusion implies that for $T < 0.135$ K the Néel state co-exists with the spin-Peierls state for a *pure* p-CyDOV system. While the co-existence of a magnetically ordered Néel state and spin-Peierls state for doped systems can be understood in the framework of the above mentioned theories [4–6], for a pure system the question remains, whether it is possible, by lowering the temperature, to transfer from a spin-Peierls state with a singlet ground state and spin-gapped excitations to a Néel state.

Motivated by these experimental findings, we propose a simple model (in which a spin subsystem possesses an exact solution) that can manifest a transition at some temperature T_{sP} to the spin-Peierls state, and then, at $T_N < T_{sP}$, a transition to the Néel state. It must be emphasized that our simple model cannot describe the behaviour of p-CyDOV, but rather it gives an insight with the help of a simple example to the possible co-existence of the antiferromagnetic and spin-Peierls orderings in a quasi-1D quantum spin system.

Let us consider, first, a $S = \frac{1}{2}$ antiferromagnetic chain with the Hamiltonian

$$\mathcal{H}_0 = \sum_n \left[J_1 (S_{n,1}^x S_{n,2}^x + S_{n,1}^y S_{n,2}^y) + J_2 (S_{n,2}^x S_{n+1,1}^x + S_{n,2}^y S_{n+1,1}^y) \right], \tag{1}$$

where $S_{n,1,2}^{x,y,x}$ are operators of the projections of a spin $\frac{1}{2}$ in site n, which belong to the first or to the second sublattice, and $J_{1,2}$ are exchange coupling constants for spins in the chain with the right and left neighbours, respectively. In [11] the exact solution of the model was obtained. After a Jordan–Wigner transformation [16] and following unitary transformations, equation (1) can be exactly diagonalized.

We can suppose that $J_{1,2}$ are different due to spin-Peierls alternating distortions of the lattice. To describe the spin-Peierls transition we can follow, for example, the strategy of [12], i.e., consider the lattice in the adiabatic approximation with $J_{1,2} = J(1 \pm u)$, where J is the
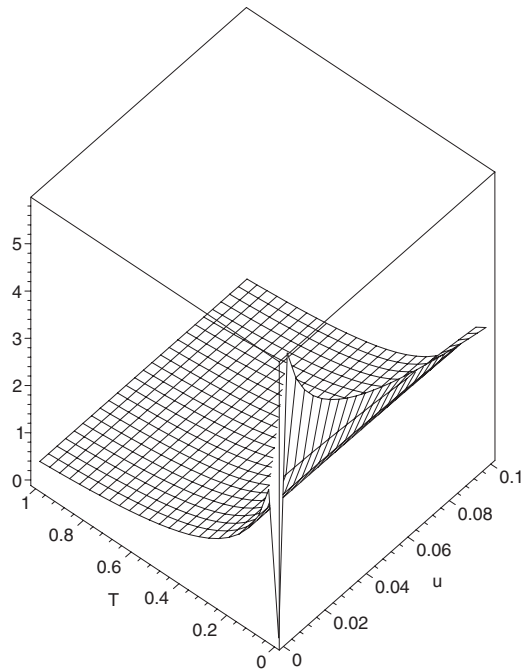
Figure 1. The integral from the left-hand side of equation (3) for $J = 1$.

exchange constant of the non-distorted spin chain (at temperatures higher than the temperature of the transition to a spin-Peierls phase), and u are weak distortions (they are supposed to be nonzero for temperatures lower than the temperature of the spin-Peierls transition). Including the elastic energy of distortions in the lowest order in u as $\mathcal{H}_1 = NCu^2$ (where the kinetic energy of ions in the lattice is completely neglected), with even N being the total number of sites in the chain, and C being an elastic constant, associated with the distortion u, we can minimize the free energy of the coupled system (for $H = 0$, because a nonzero magnetic field H can yield additional phase transitions to multimerized states), which is described by $\mathcal{H}_0 + \mathcal{H}_1$, with respect to u. In the ground state such a minimization yields two solutions for u: $u = 0$, and the solution of the following equation

$$\frac{\pi C}{J} = \int_0^{\pi/2} dk \frac{\sin^2 k}{\sqrt{u^2 \sin^2 k + \cos^2 k}}. \tag{2}$$

For small u the asymptotic solution of that equation is $u \approx \exp(-\pi C/J)$. It is easy to show that the latter possesses lower energy than the energy of the non-dimerized chain (with $u = 0$). For nonzero temperatures the minimization of the free energy of the coupled system yields

$$\int_0^{\pi/2} dk \frac{\sin^2 k}{\sqrt{u^2 \sin^2 k + \cos^2 k}} \tanh\left(\frac{J\sqrt{u^2 \sin^2 k + \cos^2 k}}{2T}\right) = \frac{\pi C}{J}. \tag{3}$$

The critical temperature of the transition to the spin-Peierls state, T_{sP}, is obtained when we put $T = T_{sP}$ with $u = 0$. Obviously, T_{sP} decreases with the increase of u and becomes zero at the value of u which satisfies equation (2). The left-hand side of equation (3) for $J = 1$ is presented in figure 1. One can see that a solution to equation (3) exists. Also, it turns out that for reasonable values of C, the steady-state distortion is a smooth function of T, and u weakly

changes with T at low temperatures, cf [17]. A different approach, but with similar results, is based on the Frölich-like spin–phonon Hamiltonian of a quantum spin chain, cf [18]; see the appendix.

The dimerized spin chain with the Hamiltonian equation (1) for $u \neq 0$ has the singlet ground state (for N even) and spin-gapped excitations. The value of the gap for $H = 0$ is $|J_1 - J_2| = Ju$, i.e. it is related to the value of the distortion u. Then, the correlation length, which is proportional to the ratio of the characteristic velocity of spin excitations divided by the gap, is u.

Now, let us consider a set of 1D spin chains with the Hamiltonian $\mathcal{H}$, coupled via the inter-chain exchange interaction with the exchange constant J'. Then the Néel temperature of the total system of weakly coupled chains is determined from the equation

$$zJ'\chi_{\mathrm{st}}(T_{\mathrm{N}}) = 1, \tag{4}$$

where z is the number of neighbouring chains, and $\chi_{\mathrm{st}}(T)$ is the staggered magnetic susceptibility of a single chain with the Hamiltonian equation (1). Equation (4) is determined by the pole of the three-dimensional staggered magnetic susceptibility in the RPA approximation (Dyson's equation). A similar description of quantum spin chains with gapped low-lying excitations was used, for example, in [19]. The dispersion laws of a spin chain in the presence of a staggered magnetic field $H_{\mathrm{st}}(T)$ can be easily written

$$\varepsilon_{1,2}(k) = \pm\tfrac{1}{2}\sqrt{4H_{\mathrm{st}}^2 + J_1^2 + J_2^2 + 2J_1 J_2 \cos k}. \tag{5}$$

The staggered magnetic susceptibility of a single dimerized chain is equal to

$$
\chi_{\mathrm{st}} = \frac{1}{\pi} \int_0^{\pi/2} \frac{\mathrm{d}k}{H_{\mathrm{st}}^2(T) + J^2[\cos^2 k + u^2(T)\sin^2 k]} \left[\frac{J^2[\cos^2 k + u^2(T)\sin^2 k]}{\sqrt{H_{\mathrm{st}}^2(T) + J^2[\cos^2 k + u^2(T)\sin^2 k]}} \right.
$$
$$
\times \tanh\left(\frac{\sqrt{H_{\mathrm{st}}^2(T) + J^2[\cos^2 k + u^2(T)\sin^2 k]}}{2T} \right)
$$
$$
\left. + \frac{H_{\mathrm{st}}^2(T)}{T \cosh^2\left(\frac{\sqrt{H_{\mathrm{st}}^2(T)+J^2[\cos^2 k+u^2(T)\sin^2 k]}}{2T} \right)} \right], \tag{6}
$$

which for $H_{\mathrm{st}} = 0$ (notice that $H_{\mathrm{st}}(T) = 0$ for $T \geqslant T_{\mathrm{N}}$, and to determine T_{N} it is necessary to consider $H_{\mathrm{st}} = 0$ in equation (4)) reduces to

$$\chi_{\mathrm{st}}(H_{\mathrm{st}} = 0) = \frac{1}{\pi J} \int_0^{\pi/2} \frac{\mathrm{d}k}{\sqrt{\cos^2 k + u^2(T)\sin^2 k}} \tanh\left(\frac{J\sqrt{\cos^2 k + u^2(T)\sin^2 k}}{2T} \right). \tag{7}$$

Then the equation which determines the Néel temperature is

$$\int_0^{\pi/2} \frac{\mathrm{d}k}{\sqrt{\cos^2 k + u^2(T_{\mathrm{N}})\sin^2 k}} \tanh\left(\frac{J\sqrt{\cos^2 k + u^2(T_{\mathrm{N}})\sin^2 k}}{2T_{\mathrm{N}}} \right) = \frac{\pi J}{zJ'}. \tag{8}$$

The left-hand side of equation (8) for $J = 1$ is presented in figure 2. One can see that a solution to equation (8) exists for $J' < J$ for small enough temperatures, so that $T_{\mathrm{N}} < T_{\mathrm{sP}}$, but T_{N} is not exponentially small, as it was believed. The confusion was because usually only homogeneous, not staggered, magnetic susceptibility was taken into account, when evaluating T_{N} with the help of Dyson's equation. The former, unlike the staggered susceptibility, is exponentially small at low temperatures for a dimerized spin chain [11], and, hence, such a
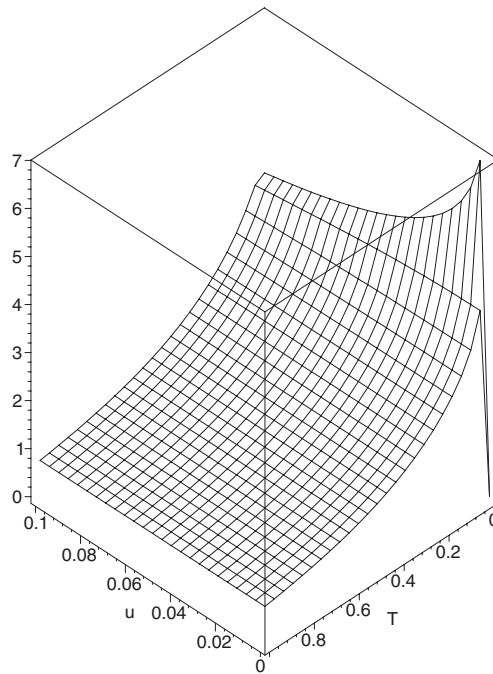
Figure 2. The integral from the left-hand side of equation (8), i.e. $\pi \chi_{\mathrm{st}}(H_{\mathrm{st}} = 0)$, for $J = 1$.

substitution implies the absence of magnetic ordering in the dimerized chain (the coupling constant between chains, J' has to be exponentially large in that case, which contradicts the condition of quasi-one-dimensionality $J' < J$). However, such a kind of magnetic ordering for a system of coupled spin chains corresponds to Curie–Weiss (ferromagnetic) ordering, while we are interested in Néel (antiferromagnetic) ordering. Notice that for $T_{\mathrm{N}} \leqslant T$ we have the solution $H_{\mathrm{st}} = 0$, while $H_{\mathrm{st}} \neq 0$ for $T \leqslant T_{\mathrm{N}}$.

In our analysis we considered the z-component of the staggered magnetic susceptibility of our 1D model. Obviously, the x or y components of the staggered susceptibility have to be larger than its z component for a gapped spin chain with an easy-plane (in the x–y plane) magnetic anisotropy (cf, for example, [20]), and therefore the consideration of the x-components of a staggered susceptibility will produce even larger values of T_{N}. Moreover, for the generic situation of the Heisenberg chain with an isotropic exchange interaction, such a difference in the direction of the staggered magnetic field does not play a role. Unfortunately, the Heisenberg model for a spin-$\frac{1}{2}$ antiferromagnetic chain (without magnetic anisotropy) cannot be solved exactly in the presence of dimerization. Such an analysis can be done, using, for example, the approximate bosonization scheme (see, for example, [21]). Consider instead of the Hamiltonian (1) the generic case

$$\mathcal{H}_{\mathrm{H}} = \mathcal{H}_0 + \sum_n \left[J_1^z S_{n,1}^z S_{n,2}^z + J_2^z S_{n,2}^z S_{n+1,1}^x \right], \tag{9}$$

where $J_{1,2}^z = J^z(1 \pm u)$ are the constants of the Ising part of the exchange interaction. Here $J = J^z$ corresponds to the isotropic Heisenberg spin chain. Let us use the Jordan–Wigner transformation [16]: the Ising part of the Hamiltonian (proportional to J^z) in the fermionic representation consists of the sum of the quadratic and quartic terms in Fermi operators $c^\dagger(n)$

and $c(n)$. One can introduce the slowly varying fermionic fields $F_R(n)$ and $F_L(n)$ as

$$c(n) \sim F_R(n)e^{ik_F n} + F_L(n)e^{-ik_F n}, \tag{10}$$

(and similarly for $c^\dagger(n)$), where k_F is the Fermi momentum,

$$
\begin{aligned}
F_R(x) &= \sum_{|k-k_F| \ll \alpha^{-1}} c_k e^{i(k-k_F)x}, \\
F_L(x) &= \sum_{|k+k_F| \ll \alpha^{-1}} c_k e^{i(k+k_F)x},
\end{aligned} \tag{11}
$$

and α is the cut-off (of order of the inter-site distance). Using the bosonization rules

$$
\begin{aligned}
F_R(x) &= \exp\left((i/2)\left[\phi - 4\pi \int_{-\infty}^{x} \Pi(y)\,dy \right] \right), \\
F_L(x) &= \exp\left((i/2)\left[\phi + 4\pi \int_{-\infty}^{x} \Pi(y)\,dy \right] \right), \\
[\phi(x), \Pi(y)] &= i\delta(x-y),
\end{aligned} \tag{12}
$$

we can approximately rewrite the Hamiltonian $\mathcal{H}_H$ in a staggered field H_{st} in terms of introduced Bose fields $\phi(x)$ and $\Pi(x)$ as

$$
\mathcal{H} = \int dx \left(v\pi\eta\Pi^2(x) + \frac{v}{4\pi\eta}\left[\frac{\partial\phi(x)}{\partial x} \right]^2 \right.
$$
$$
\left. - \frac{\sqrt{J^2 u^2 + H_{st}^2}}{\pi\alpha^2} \sin[\phi(x) + \gamma] + \frac{J^z}{2\pi^2\alpha^3} \cos[2\phi(x)] \right). \tag{13}
$$

Here the velocity of Bose excitations is equal to $v = (1/2)\sqrt{J^2 + [2JJ^z/\pi\alpha]}$, $\tan\gamma = Ju/H_{st}$ and the Luttinger liquid parameter $\eta = 2/\sqrt{1 + [2J^z/\pi J\alpha]}$. Notice that the Ising part of the Hamiltonian renormalizes the quadratic in the Bose fields part as well as introducing the cosine term. We point out here that the explicit bosonization procedure applied above can be formally justified only for the weak coupling limit $J^z \ll J$, but the form of equation (13) is known to be valid until the isotropic Heisenberg point, where the $SU(2)$ symmetry requires $\eta = 1$ [21] (i.e. for $0 \leqslant J^z \leqslant J$ we have to consider $2 \geqslant \eta \geqslant 1$). The renormalization group eigenvalue of the second term in equation (13) is equal to $2 - \eta/2$, while for the third term it is $2 - 2\eta$. Obviously for the antiferromagnetic situation with the easy-plane type of the anisotropy (including the isotropic point) $0 \leqslant J^z \leqslant J$ the exchange Ising interaction is always irrelevant, while the alternating part u and the staggered field H_{st} are equally relevant. Hence, the behaviour of the more generic isotropic Heisenberg antiferromagnetic spin chain has to be qualitatively similar to the results obtained above for the XY chain, because of the irrelevance of the easy-plane magnetic anisotropy.

A qualitative difference exists between the XY chain considered in our work and the isotropic Heisenberg chain. For small values of u, the ground state energy of a Heisenberg chain has behaviour $\propto u^{4/3}$, not $u^2 \ln^2 u$ as for the XY chain considered [13]. This implies higher values of the spin-Peierls transition temperature for the isotropic Heisenberg chain than for the XY chain, i.e. our consideration underestimates the possible transition temperature to the spin-Peierls state, which must be higher for the generic Heisenberg situation.

On the other hand, the staggered susceptibility of a dimerized spin-$\frac{1}{2}$ isotropic antiferromagnetic Heisenberg chain can be estimated using, for example, the dynamical correlated effective field approximation [22], or by an exact diagonalization of finite chains [23]. All these methods (including the bosonization) produce a temperature behaviour of a staggered magnetic susceptibility (which is finite for small T and decreases with the increase of T)

qualitatively similar to equation (7). This is why we can suppose that our analysis of the co-existence of an antiferromagnetic ordering and spin-Peierls ordering in the low-temperature phase will be valid (qualitatively, but not quantitatively) for Heisenberg quasi-1D quantum spin chains.

In summary, motivated by recent experiments, we have studied a simple theoretical model of co-existence of the Néel antiferromagnetic ordering and spin-Peierls ordering in an undoped (pure) quasi-one-dimensional quantum antiferromagnetic system, and calculated the spin-Peierls transition and the Néel transition temperatures in the framework of this model.

Appendix

If the exchange constant of the spin chain for spins at sites n and $n + 1$ can be considered as $J(n, n + 1) = J + [u(n) − u(n + 1)]\nabla_1 J(n, n + 1)$, the Hamiltonian of a spin chain and associated phonons can be written as

$$\mathcal{H} = \sum_k J \cos k\, a_k^\dagger a_k + \sum_q \hbar\omega_0(q) b_q^\dagger b_q + \frac{1}{\sqrt{N}} \sum_{k,q} g(q)(b_q + b_{-q}^\dagger) a_k^\dagger a_{k-q}, \tag{A.1}$$

where we used the Jordan–Wigner transformation for spins. The second term determines the energy of photons, related to distortions of magnetic ions in the chain (with their creation and destruction operators $b_q^\dagger$ and b_q, and energies $\hbar\omega_0(q)$), and the third term describes the spin–phonon coupling (with $g(q) = iM^{-1/2}\nabla_1 J(n, n + 1)[\sin k − \sin q − \sin(k − q)]$, where M is the mass of a magnetic ion). The 1D fermion system is unstable with respect to quasi-static density fluctuations of the wavevector $q_0 = \pm 2k_F$, where k_F is the Fermi momentum of fermions. Within the linear response the screened phonon frequencies are renormalized to

$$\Omega^2(q) = \omega_0^2(q) − \frac{\omega_0(q)|g(q)|^2}{J\hbar N} \sum_k \frac{f_k − f_{k-q}}{\cos(k − q) − \cos k}, \tag{A.2}$$

where $f_k = 1/[1 + \exp(J \cos k/T)]$. Taking into account that $\cos(k − q_0) = − \cos k$ we easily obtain

$$\Omega_{q_0}^2 = \frac{\omega_0(q_0)|g(q_0)|^2 N(0)}{\hbar} \ln\left(\frac{T}{T_{sP}}\right), \tag{A.3}$$

where $N(0) = 1/2\pi J$ is the density of states at the Fermi level, and

$$T_{sP} = 2.28 J \exp(−\hbar\omega_0(q_0)/|g(q_0)|^2 N(0)). \tag{A.4}$$

For the alternating spin chain with $J_{1,2} = J[1 \pm u(T)]$, one finds $u(T) = (g/J\sqrt{2\hbar\omega_0(q_0)})\langle b_{q_0} + b_{-q_0}^\dagger\rangle$, which implies $u(0) = 1.77T_{sP}/J$. Obviously, the two approaches produce similar results, i.e. exponentially small distortions due to the coupling between the quantum antiferromagnetic spin chain and a 3D lattice.

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