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Two-particle spin-singlet excitations in coupled spin-1/2 antiferromagnetic alternating chains

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Abstract

The spectrum of the two-particle spin-singlet ($\Delta S^z = 0, \pm 1$) excitations of a weakly coupled antiferromagnetic spin-1/2 alternating ($J_1 \pm \delta$) chain is calculated using a mean-field approximation for the interchain exchange (J_2) by the quantum Monte Carlo method. The bandwidth change of these excitations, the mass gaps in the singlet–singlet excitation spectrum, the top boundaries of the bands and the velocities of these excitations are estimated as functions of the alternating bond δ. The temperatures at which the singlet gaps close are determined.

1. Introduction

One-dimensional spin systems with antiferromagnetic interactions have aroused great interest in researchers because exact solutions can be derived taking into account various kinds of interaction. In particular, the spin–phonon interaction can cause a spin–Peierls transition [1,2]. The magnetic properties of such systems are analysed in terms of Heisenberg [3] and sine–Gordon [4–6] models with alternating bonds. For a single alternating chain made up of two types of one-particle breather state with mass gaps of order M and $\sqrt{3}M$, the threshold of the breather–breather continuum $2M$ derived from the two-point correlators of cosines has been estimated [4–6]. The main difficulty as regards these exactly solvable models is the mapping to the continuum limit, which is uncontrolled; some information can be lost. So the spin operators have been expressed in terms of bosonic exponents and then the spin–spin correlation function for the transverse spin components of nearest neighbours has been calculated. Also, it is difficult to derive relations between parameters in the lattice and sine–Gordon models.

Most of the studies have been devoted to determining the triplet excitation spectrum using the Luttinger-model approximation to the Heisenberg chain [2] or spinon excitations by means of exact-diagonalization (ED) and density-matrix renormalization-group calculations [7]. The spinon energy is defined as the energy difference between the open chains of length $L = 2p+1$ and $L = 2p$, which can be considered as the excitation energy of a spin-1/2 topological

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defect. Within the framework of this model, the binding energy of the topological solitons with $S = 1/2$ is equal to zero because $\Delta_{s=1/2} = \Delta_{s=1}/2$. The singlet excitations play an important role in spin relaxation, muonium and neutron scattering diffusion, and magnetic heat conductivity. These excitations can be observed by means of Raman spectroscopy. For example, the Raman scattering energies are 1.73 meV and 3.7 meV [10,11] for the spin–Peierls compound $CuGeO_3$ [12]. The parameters for $CuGeO_3$ lead to a larger value of the energy of the two-particle continuum, which is at variance with experimental findings.

Earlier [9], on the basis of the estimated four-spin correlation function, the dimer order parameter, and a longitudinal component of the total spin squared as a function of temperature and magnetic field, we proposed the existence of massive singlet excitations with unchanging total spin. The interaction between two kinks can decrease the excitation energy and result in quasiparticle stability. Such quasiparticles are characterized by a fixed size l and can exist in the chain if the correlation radius of the antiferromagnetic ordering is $\xi \ll l$.

In the present paper we estimate the spin–spin correlation functions in space-time, $\langle S_i^\alpha(0) S_{i+l}^\alpha(0) S_j^\alpha(t) S_{j+l}^\alpha(t)\rangle$, where $\alpha = z, \pm$ correspond to longitudinal and transverse spin components in the antiferromagnetic chain with alternating exchange. The three-dimensional nature of the spin–Peierls transition and the interactions between chains are taken into account in the mean-field approximation [13]. We will show that the alternation of the bonds leads to bandwidth change of the longitudinal and transverse two-particle spin excitations. We successfully explain the results of Raman experiments on $CuGeO_3$. The quantum Monte Carlo (MC) method is used within schemes which rely on the Suzuki–Trotter discretization of imaginary time.

2. The model

We shall consider a Heisenberg model with negative interactions between nearest neighbours with $S = 1/2$ directed along an external-field OZ-direction. The Hamiltonian has the form

$$H = -J_1 \sum_{i,j} [1 + (-1)^i \delta] \boldsymbol{S}_{i,j} \cdot \boldsymbol{S}_{i+1,j} - J_2 \sum_{i,j,\gamma} \boldsymbol{S}_{i,j} \cdot \boldsymbol{S}_{i,j+\gamma} \tag{1}$$

where $J_1 < 0$, $J_2 < 0$ are the intrachain and interchain exchange, and γ is summed over the nearest neighbours in the transverse directions ($z = 4$). The Hamiltonian (1) transforms into an effective single-chain problem on applying a mean-field treatment of the interchain coupling [13]. The Hamiltonian then takes the form

$$H = -\sum_{i=1}^{L/2} J_{2i,2i-1} S_{2i} S_{2i-1} - \sum_{i=1}^{L/2} K_{2i,2i+1} S_{2i} S_{2i+1} - \sum_{i=1}^{L} h^z S_i^z - 2L J_2 m_0^2. \tag{2}$$

Here $J = J_1 + \delta$ and $K = J_1 - \delta$, and m_0 and h^z are the staggered magnetization and field determined as

$$m_0 = (2/L) \sum_{i=1}^{L/2} \sqrt{\mathrm{abs}(S_0^z S_i^z)} \qquad h_i^z = 4 J_2 \,\mathrm{sgn}(S_i^z) \sqrt{\mathrm{abs}(S_0^z S_i^z)}. \tag{3}$$

The algorithm and method used for determining the excitation spectrum have been considered in detail earlier [14]. In this report, we do not aim to obtain the dynamical correlation function for the whole real frequency domain, but simply wish to extract the low-lying eigenvalues as a function of q. Let us introduce an imaginary-time correlation function $S_{s1,2}(q, \tau)$ as follows:

$$S_{s1,2}(q, \tau) = \langle \exp(H\tau) S_{q,s1,2}^\alpha \exp(-H\tau) S_{-q,s1,2}^\alpha \rangle_{MC} \qquad (\alpha = z, +, -)$$

where

$$S^z_{q,s1} = (1/(L-l)) \sum_{j=1}^{(L-l)/2} [S^z_{2j} S^z_{2j+l} \exp(\mathrm{i}q(2j+l)) + S^z_{2j-1} S^z_{2j+l-1} \exp(\mathrm{i}q(2j+l-1))]$$
$$S^{+,-}_{q,s2} = (1/2(L-l)) \sum_{j=1}^{(L-l)/2} [(S^+_{2j} S^-_{2j+l} + S^-_{2j} S^+_{2j+l}) \exp(\mathrm{i}q(2j+l)) + (S^+_{2j-1} S^-_{2j+l-1} + S^-_{2j-1} S^+_{2j+l-1}) \exp(\mathrm{i}q(2j+l-1))]. \quad (4)$$

Here $\langle \cdots \rangle_{MC}$ denotes a Monte Carlo average of the quantity at a given temperature T; the momentum changes in the range $0 < q < \pi$ and is equal to $q = \pi n/(L-l)$, where $n = 0, 1, \ldots, L-l$, and $l = 0, 1, \ldots, 12$. In quantum MC simulations, the imaginary time τ takes a set of discrete numerical values $\tau = \beta n/m$, where $n = 1, 2, \ldots, m$ and $\beta = [1/(k_B T)]$. $S_{s1,2}(q, \tau)$ is estimated from the spin–spin correlation function for two Trotter layers separated by $m\tau/\beta$, with $0 < \tau < \beta/2$, in the transformed two-dimensional Ising system. The imaginary-time correlation functions $S_{s1,2}(q, \tau)$ are satisfactorily fitted by an exponential dependence on time τ in the range $\tau_0 < \tau < \tau_{max}$, on the basis of which the dispersion relation is calculated as follows:

$$E_{s1,2}(q) = -\ln([S_{s1,2}(q, \tau)/S_{s1,2}(q, \tau_0)])/(\tau - \tau_0) \quad (5)$$

where $\tau_0 = 1/E_{s1,2}$, and τ_{max} is close to the time of the onset of statistical fluctuations in $S(q, \tau)$; also, E_{s1}, E_{s2} are associated with the low-lying longitudinal $\Delta S^z = 0$ and transverse $\Delta S^z = \pm 1$ singlet excitations. This classification is based on the action of two operators $S^z_j S^z_{j+l}$ and $S^+_j S^-_{j+l}$ on any states—for example, the singlet states

$$S^z_j S^z_{j+l} |s_j, s_{j+l}\rangle = (1/4)|t_{0,j} t_{0,j+l}\rangle$$

and

$$(1/2)(S^+_j S^-_{j+l} + S^-_j S^+_{j+l})|s_j, s_{j+l}\rangle = (-1/4)(|t_{-1,j} t_{1,j+l} + t_{1,j} t_{-1,j+l}\rangle)$$

where

$$|s_j\rangle = (1/\sqrt{2})(|\uparrow_j \downarrow_{j+1}\rangle - |\downarrow_j \uparrow_{j+1}\rangle) \qquad |t_{0,j}\rangle = (1/\sqrt{2})(|\uparrow_j \downarrow_{j+1}\rangle + |\downarrow_j \uparrow_{j+1}\rangle)$$

and where $t_{\pm 1,j}$ are respectively the singlet and triplet states with $S^z = 0, \pm 1$. The total-system spin does not change upon the action of these operators, $\sum_i S^z_i = 0$, and these excitations are of the singlet type.

3. Results and discussion

The singlet excitation spectra are shown in figure 1 for an alternating AF chain at the low temperatures $T/J = 0.05, 0.07$. The bandwidth of the two-particle spin excitations is decreased for the spin pair $S^+_j S^-_{j+1}$ and increased for $S^z_j S^z_{j+1}$ as a result of the alternating bond. The singlet excitation spectrum with $\Delta S^z = \pm 1$ exhibits a minimum at the momenta $q \neq 0, \pi$. This energy minimum is associated with a mass gap of Δ_{s2}. The dispersion relation $E(q)$ can be described using three parameters: the energy maximum E_{max} or top boundary of the spin excitation band, the velocity v, and a gap energy Δ at the centre and at the edges of a band. These parameters were determined from a fit to the MC result with the following functions:

$$E(q) = \sqrt{\Delta^2 + v^2 q^2} \qquad q < \pi/8$$
$$E(q) = \sqrt{\Delta^2 + v^2(\pi - q)^2}.$$

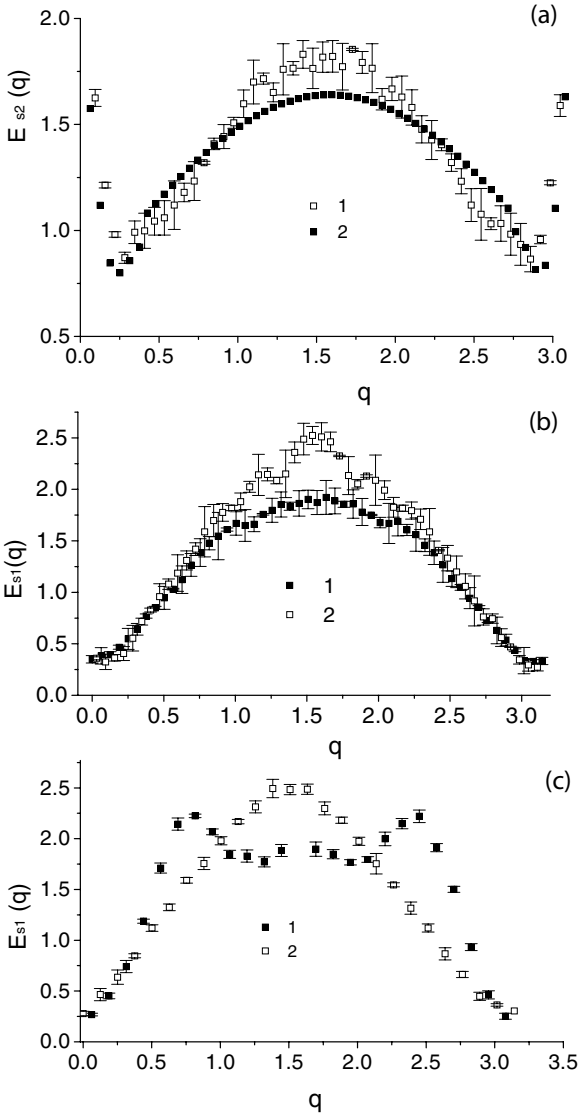

Figure 1. The singlet excitation spectra of the AF chain ($J_2 = 0$) with $\Delta S^z = \pm 1$ (a) and $\Delta S^z = 0$ ((b), (c)) for $l = 1$ ((a), (b)), 3 (c) as functions of momentum q.

The dispersion relations of the singlet excitations on the J- and K-bonds are split in the middle of the Brillouin zone (see figure 1). This is easily understood if we consider four spins in the limit $K \to 0$ ($1 \overset{J}{\Longleftrightarrow} 2 \overset{K}{\cdots} 3 \overset{J}{\Longleftrightarrow} 4$). The action of the two operators $S_2^z S_3^z$ and $S_1^z S_2^z$ on the singlet state $\psi \sim (|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle)(|\uparrow_3\downarrow_4\rangle - |\downarrow_3\uparrow_4\rangle)$ gives respectively the excitation energies $E_{s,K\to 0} \simeq 2(J_1 + \delta)$ and $E_{s,J} \simeq (J_1 + \delta)$. The excitation energy maxima versus the exchange alternation are presented in figure 2(a). They are well parametrized by polynomials of the second degree:

$$E_{max,s1,K} = 1.63(3) + 4.2(4)\delta - 2.8(6)\delta^2 \qquad E_{max,s1,J} = 1.63(1) + 1.28(6)\delta - 1.08(7)\delta^2$$

for a singlet with $\Delta S^z = 0$ and

$$E_{max,s2,K} = 1.55(3) + 1.1(2)\delta + 1.45(9)\delta^2 \qquad E_{max,s2,J} = 1.50(3) + 1.1(2)\delta - 0.5(2)\delta^2$$

for one with $\Delta S^z = \pm 1$ excitations. These dependencies characterize a change of the top boundary of the singlet excitation band versus the bond alternation.

The velocities of the singlet excitations on the J- and K-bonds are also varied as shown in figure 2(b). In order to estimate the velocity variation $v_{s1}(\delta)$ in the limit case where $\delta \to 1$, $v_{sJ} \to 0$, $v_{sK} \to 2v_s(\delta = 0)$, the polynomial approximation is used. The values of the singlet excitation velocity $v_{s1}(\delta)$ simulated by the MC method are well described by the following functions:

$$v_{sK} = 1.75(3) + 0.02(2)\delta + 1.1(2)\delta^2 \qquad v_{sJ} = 1.71(2) + 0.3(1)\delta - 1.6(2)\delta^2.$$

Now we estimate the excitation spectrum for various spin pairs versus its size l, which may be illustrated as

$$\cdots \uparrow \underbrace{s \cdots s}_{l} \downarrow \cdots \uparrow \underbrace{s \cdots s}_{l} \downarrow$$

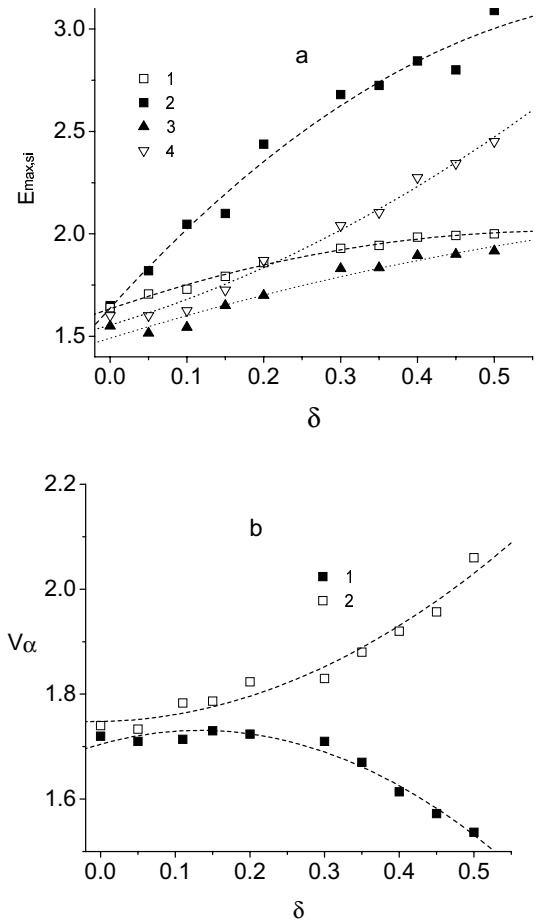

Figure 2. The top boundaries (a) and velocities (b) of the singlet excitations for the K ((2), (4)) and J ((1), (3)) exchanges with $\Delta S^z = 0$ ((1), (2)), $\Delta S^z = \pm 1$ ((3), (4)) versus the alternating bond δ for $J_2 = 0$.

where 's' stands for a singlet state. In figure 1(c) the dispersion relations for $l = 3$ are presented.

The gap in the spectrum of singlet–singlet excitations decreases with the increase of the distance l and has a minimum at $l \sim \xi/2 = 1/\delta$ for $\Delta S^z = 0$ and at $l \sim \xi = 2/\delta$ for $\Delta S^z = \pm 1$ as shown in figure 3. Consequently, the antiferromagnetic correlation or the short-range order causes spin repulsion in the excited state.

The gaps in the spectrum of the singlet excitations at $l = 1,\ 1/\delta,\ 2/\delta$ are presented in figure 4. The δ-dependence of Δ may be parametrized by a power-law behaviour: $\Delta_i = A_i \delta^{\beta_i}$ where $A_1 = 2.8(2)$, $\beta_1 = 1.4(1)$ ($\Delta S^z = 0, l = 1/\delta$), $A_2 = 2.2(2)$, $\beta_2 = 1.07(7)$ ($\Delta S^z = 0, l = 1$), $A_3 = 2.1(1)$, $\beta_3 = 0.69(6)$ ($\Delta S^z = \pm 1, l = 2/\delta$), $A_4 = 2.1(1)$, $\beta_4 = 0.54(3)$ ($\Delta S^z = \pm 1, l = 1$). This differs from the δ-dependence of the gap in the spectrum of the singlet–triplet excitations which varies as $\Delta \sim \delta^{(2/3)}$ [2, 7] and agrees qualitatively with theoretical predictions derived in terms of the sine–Gordon model: $\Delta_{s2}/\Delta_t = \sqrt{3}$ [4–6], and in the limit case $\delta \to 0$: $\Delta_{s2} \simeq \Delta_t \to 0$.

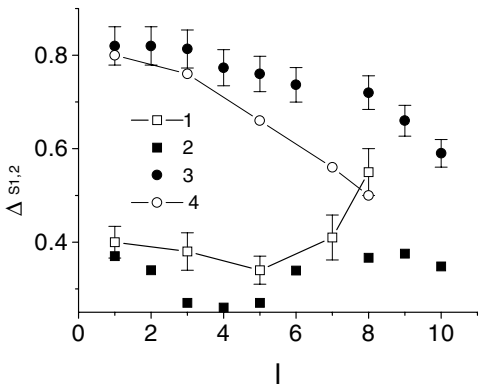

Figure 3. The gaps Δ_{s1} ((1), (2)) and Δ_{s2} ((3), (4)) in the singlet–singlet excitation spectra versus size of particle for $J_2/J_1 = 0.0$ ((2), (3)), 0.06 ((1), (4)).

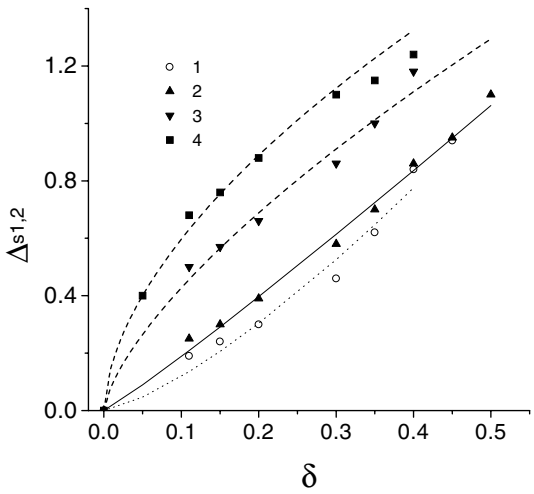

Figure 4. The gap energy in the singlet excitation spectrum with $\Delta S^z = 0$ ((1), (2)) and $\Delta S^z = \pm 1$ ((3), (4)) at $l = 1$ ((2), (4)), $1/\delta$ (1), $2/\delta$ (3), $J_2 = 0$, as functions of δ. The fitting functions $\Delta_{si} = A_i \delta^{\beta_i}$ are presented as lines.

Now we shall estimate the effect of the interchain coupling on the singlet excitation spectrum. The top band boundaries of the singlet excitations for the weak-K and strong-J exchanges respectively decrease and increase as a result of the interchain interaction, and they coincide at $J_2/J_1 \simeq 0.2\delta$. The biggest changes in the mass gap occur for the size of particle equal to the correlation radius of the short-range antiferromagnetic order, $l \simeq \xi$, as shown in figure 3. The correlation radius does not depend on temperature in the singlet state for $T \ll T_c$, where T_c is the spin–Peierls temperature, and it is related to the alternating bond as $\xi \simeq 2/\delta$. A transition from a singlet state into a state with long-range antiferromagnetic order takes place at $J_2/J_1 = 0.52(3)\delta$ [13]. The density of states of the singlet excitations has a massive peak at the bottom of the band for quasi-one-dimensional antiferromagnets and in the vicinity of the gap energy $\Delta_{s1,2}$ for the alternating chain. As a result, the Raman scattering should show a dependence on the polarization of light. If the polarization vector $\boldsymbol{e}$ is directed along OZ the scattering occurs at the energy Δ_{s1} and for the other case, $\boldsymbol{e}$ perpendicular to OZ, the effect is observed at the energy Δ_{s2}. These energies vary in the range $\Delta_{s2}/\Delta_{s1} \sim 2.4$–$1.3$ for $\delta = 0.124$–0.5 and coincide in the limit cases $\delta \to 0, 1$. Similar effects should appear in the spin–spin scattering when allowance is made for the spin correlation $(S_j^{\pm,z}, S_i^z S_{i+l}^z)$ or $(S_j^{\pm,z}, S_i^{\pm} S_{i+l}^{\mp})$. Muonium and neutron scattering diffusion will reveal two maxima at these energies, $\Delta_{s1,2}$.

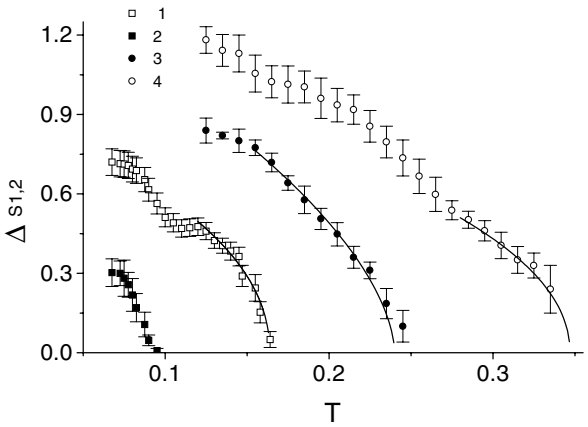

Figure 5. The gap energies Δ_{s1} ((2), (3)) and Δ_{s2} ((1), (4)) in the singlet excitation spectrum for $\delta = 0.12$ ((1), (2)), 0.4 ((3), (4)), $J_2 = 0$, $l = 1$, against temperature. The fitting functions $\Delta_{s1,2} = A(T - T_{s1,2})^{\beta}$ are plotted as solid lines.

The gaps in the singlet excitation spectra disappear at the critical temperatures T_{s1} and T_{s2}, which are plotted in figure 5. The magnitude of $T_{s1,2}$ is determined from a fit of the MC results to the function $\Delta_{s1,2} = A(T - T_{1,2})^{\beta}$ with three parameters $A, \beta, T_{s1,2}$, where β is varied in the range 0.31–0.38. The power approximations $T_{s1} = 0.43(2)\delta^{0.63(4)}$ and $T_{s2} = 0.57(3)\delta^{0.55(4)}$ agree well with the MC results, presented in figure 6. The magnetic heat conductivity should show two anomalies at these temperatures in the quasi-one-dimensional antiferromagnet with alternating bonds. According to our calculations, the gaps in the spin–Peierls compound $CuGeO_3$ observed by means of Raman spectroscopy at 1.73 meV and 3.7 meV [10, 11] result from the singlet two-particle excitations with size of particle $l \sim 8c$ for the parameters $J_c \simeq 94$ K, $\delta = 0.12$, $J_2/J_1 \simeq 0.04$. We predict a polarization dependence of the Raman scattering for $\boldsymbol{e} \parallel \boldsymbol{c}$, $\Delta_{s1} \simeq 1.6$ meV, and for $\boldsymbol{e} \perp \boldsymbol{c}$, $\Delta_{s2} \simeq 3.9$ meV.

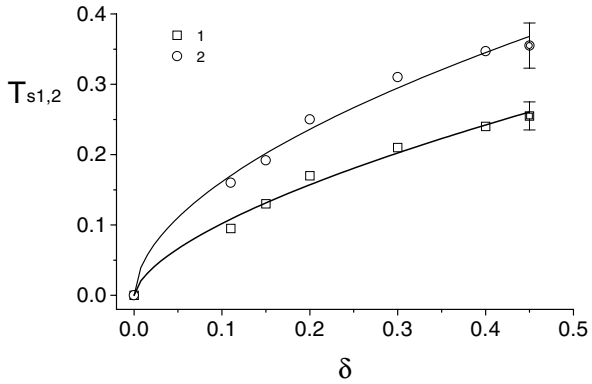

Figure 6. Critical temperatures T_{s1} (1) and T_{s2} (2) at which the gaps close as functions of the alternating bond for $l = 1$, $J_2 = 0$.

4. Summary of our results

We determined the gap values and the top boundaries in the spectra of the two-particle singlet excitations with $\Delta S^z = 0, \pm 1$ for weakly coupled AF alternating chains. Alternation of the bonds leads to changes of the bandwidth of the singlet–singlet excitations which do not affect the longitudinal component of the total spin. Antiferromagnetic interaction causes a repulsion between two spins in the exciting particle at a distance equal to the correlation radius, which decreases the gap value. However, the interchain coupling leads to coincidence of the gap values, $\Delta_{s1}(l \sim \xi) \simeq \Delta_{s2}(l \sim \xi)$, at the critical exchange J_2 in the singlet state. Polarization dependence of the Raman scattering is predicted for the spin–Peierls compound $CuGeO_3$.

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