Dynamics of the $S = 1/2$ Alternating Chains at $T = \infty$

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The well-defined delocalization modes of J_{AF} and $2J_{\mathrm{AF}}$, where J_{AF} is the antiferromagnetic exchange integral in a spin pair, are obtained in the $S = 1/2$ alternating chains for $(\alpha/J_{\mathrm{AF}})^2 \ll 1$ at $T = \infty$ in terms of the continued fraction formalism with the recurrence relations method. These modes correspond to the single and the double singlet-to-triplet local excitations, respectively. Dynamically these very short-ranged and very weak correlations survive at $T = \infty$. We raise the possibility of observing these modes in neutron and Raman scattering at high temperatures, $k_{\mathrm{B}}T \gg J_{\mathrm{AF}}$.

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Since the Haldane conjectures [1,2] and the discovery of the inorganic spin-Peierls compound $CuGeO_3$ [3], the $S = 1/2$ alternating chains have attracted again the attention of theoretical and experimental workers. Experimental results on these spin chains have reported on magnetic susceptibility, specific heat, and neutron scattering data [3–8]. Although a number of studies have been made on, e.g., a gap formation and static quantities [4,9–14], little is known about the dynamics especially theoretically [15–17].

The purpose of this Letter is to show the emergence of well-defined delocalization modes in the $S = 1/2$ alternating chains at $T = \infty$ and to raise the possibility of observing the modes in neutron and Raman scattering at high temperatures. This study also opens the door to the dynamics with a new viewpoint. That is, high temperature dynamics can be distinguished in systems where, e.g., how dimers are geometrically configurated in the ground state. Although spins are uncorrelated at $T = \infty$, dynamics unlike thermodynamics may include the characteristic modes of a system.

The continued fraction formalism [18] has developed from the generalized Langevin equation [19] and applied to the linear response theory [20], as

$$\overline{a}_0(z) = \frac{1}{z+}\,\frac{\Delta_1}{z+}\,\frac{\Delta_2}{z+}\,\frac{\Delta_3}{z+}\cdots \tag{1}$$

for the Laplace transform $\overline{a}_0(z) = \int_0^\infty dt\, e^{-zt} a_0(t)$ of

$$a_0(t) = (A(t), A)\,(A, A)^{-1} \tag{2}$$

for a dynamical variable A, where the Kubo scalar product is $(A(t), A) \equiv 1/\beta \int_0^\beta \langle A(t - i\hbar\lambda)A^\dagger\rangle\, d\lambda - \langle A(t)\rangle\langle A^\dagger\rangle$ with $\langle O\rangle = \mathrm{Tr}[Oe^{-\beta H}]/\mathrm{Tr}[e^{-\beta H}]$ and $\beta = 1/k_{\mathrm{B}}T$. The anomalous ESR spectra [21] was explained using Eq. (1), but there were some difficulties to obtain the continued fraction coefficients $\{\Delta_n\}$. Later Lee [22,23] obtained a more simplified method of calculating a d-dimensional $\{\Delta_n\}$ as

$$f_{n+1} = iLf_n + \Delta_n f_{n-1}\,, \tag{3}$$

$$\Delta_n = (f_n, f_n^\dagger)\,(f_{n-1}, f_{n-1}^\dagger)^{-1} \tag{4}$$

with $iLO = (i/\hbar)[H, O]_-$, $f_0 = A$. The boundary conditions are $\Delta_0 = 1$ and $f_{-1} = 0$. We can reformulate $A(t) = \sum_{n=0}^{d-1} a_n(t) f_n$ with time-dependent c-number functions including Eq. (2). Equation (3) helps classify the excitation modes and Eq. (4) determines the excitation energy. Some issues such as electron gas [24,25], spin system [26–32], and strongly correlated system [33,34] have been discussed using Eqs. (3) and (4).

The Hamiltonian of the $S = 1/2$ alternating chains is

$$H = J_{\mathrm{AF}} \sum_i \boldsymbol{S}_{i,1} \cdot \boldsymbol{S}_{i,2} - \alpha \sum_i \boldsymbol{S}_{i-1,2} \cdot \boldsymbol{S}_{i,1}\,, \tag{5}$$

where a vector $\boldsymbol{S}_{i,1(2)}$ denotes a $S = 1/2$ operator at the left (right) side in a molecule on a site i [35]. The onsite antiferromagnetic (AF) exchange integral J_{AF} takes the energy unit and a coupling constant α controls the class and strength of alternation. Equation (5) describes the AF-ferromagnetic (AF-F) alternating chain for $\alpha > 0$ and the AF-AF alternating one for $\alpha < 0$. The system with $S = 1$ corresponds to that for $\alpha = +\infty$.

We investigate the dynamics of a sum of the spin z component at both sides on a certain site j,

$$A = S_{j,1}^z + S_{j,2}^z\,, \tag{6}$$

at $T = \infty$. That is, when the supply of energy obtained from a small external field is suddenly turned off at a time $t = 0$, how does the energy delocalize from a site j? Although each spin thermally fluctuates at $T = \infty$, dynamics different from thermodynamics includes the characteristic modes. Our interest is in the identification of these characteristic delocalization modes. Here we note that the Kubo scalar product at $T = \infty$ becomes the correlation function and that its evaluation can be done by taking the trace of products and neglecting fluctuations. We thus have $(f_0, f_0) = 2a$ with $a = S(S+1)\hbar^2/3$ because the trace of the cross terms vanishes. We classify Eq. (5) into three models as shown below.

We begin with the *Ising model* with $\boldsymbol{S}_{i,1(2)} = S_{i,1(2)}^x$. Using Eq. (3), we have $f_1 = f_1^0 - \alpha f_1^\alpha$ with $f_1^0 = S_{j,1}^x S_{j,2}^y + S_{j,1}^y S_{j,2}^x$ and $f_1^\alpha = S_{j-1,2}^x S_{j,1}^y + S_{j,2}^y S_{j+1,1}^x$, which leads to $(f_1, f_1) = 2a^2(1+\alpha^2)$. It is clear that $\Delta_1 = a(1+\alpha^2)$ from Eq. (4). By successive use of Eqs. (3) and (4), we obtain $f_2 = 2\alpha(S_{j-1,2}^x S_{j,1}^z S_{j,2}^x + S_{j,1}^x S_{j,2}^z S_{j+1,1}^x)$, $f_3 = 2a\alpha(1-\alpha^2)(\alpha f_1^0 + f_1^\alpha)/(1+\alpha^2)$

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and $f_4 = 0$, which lead to $\Delta_2 = 4a\alpha^2/(1+\alpha^2)$, $\Delta_3 = a(1-\alpha^2)^2/(1+\alpha^2)$ and $\Delta_4 = 0$, respectively. This is equivalent to the fact that the Hilbert space of A given by Eq. (6) is spanned by $\{f_{n\le 3}\}$ only and that the excitation localized on a site j does not spread but oscillates within a 3-spin alignment. This finite d-dimensional Hilbert space also persists at finite temperatures.

The oscillation mode is obtained by inserting the $\{\Delta_n\}$ into Eq. (1) and putting $z = -i\omega^+$ with $\omega^+ = \omega + i0^+$ and $a = 1/4$ in units of $\hbar = 1$:

$$\frac{4}{\pi}\,\mathrm{Re}\overline{a}_0(-i\omega^+) = \delta\left(\omega \pm \frac{\alpha+1}{2}\right) + \delta\left(\omega \pm \frac{\alpha-1}{2}\right). \qquad (7)$$

The first term on the right-hand side (rhs) in Eq. (7) corresponds to the excitation mode, e.g., between the configurations of (↑↓↓) and (↑↑↓) for a 3-spin alignment $(S_{j,1}S_{j,2}S_{j+1,1})$, the second between those of (↓↓↓) and (↑↓↑). The observation of the basis vector f_2 with the largest spin-component product is crucial for the identification of these modes. The case with $|\alpha| = 1$ satisfies the relation of $d = q+1$ with the dimension of the Hilbert space $d = 3$ and the coordination number $q = 2$ as shown in some Ising models by Sen [29]. These modes denote also the eigenvalues of the 4×4 matrix defined as $iL\{g_n\} = M\{g_n\}$ with $\{g_0 = f_0, g_1 = f_1^0, g_2 = f_1^\alpha, g_3 = f_2/2\alpha\}$. Here we note that these modes of order of β^0 are independent of temperatures and that the number of the modes, i.e., the structure of the Hilbert space, varies with α. The time autocorrelation function, Eq. (2), is then $\langle A(t)A\rangle/\langle A^2\rangle = \cos(\alpha t/2)\cos(t/2)$.

Next, the *XY model* with $\boldsymbol{S}_{i,1(2)} = (S^x_{i,1(2)}, S^y_{i,1(2)})$, which has been studied by the Jordan-Wigner transformation and other methods [15–17]. Since A given by Eq. (6) is a conserved quantity with respect to the first term on the rhs of Eq. (5), $f_1 = -\alpha f_1^\alpha$ is written with $f_1^\alpha = -S^y_{j-1,2}S^x_{j,1} + S^x_{j-1,2}S^y_{j,1} + S^y_{j,2}S^x_{j+1,1} - S^x_{j,2}S^y_{j+1,1}$ using Eq. (3). This shows the left- and right-directed flow of energy with a symmetric mode. Hereafter for brevity, we use a notation for the basis vectors $\{f_n\}$ such as $f_1^\alpha = [S^y_{j,2}S^x_{j+1,1}]$. What is meant by the square brackets is that a single term in it indicates four elements. The definition is twofold. First, we yield the second element by changing the spin $x(y)$ component into $y(x)$ and multiplying a sign of $(-)^n$. Second, we yield the rest of two elements by taking the mirror symmetry of the first two elements about a site j. See the expression of f_1^α.

We have $(f_1, f_1) = 4a^2\alpha^2$ and then $\Delta_1 = 2a\alpha^2$ using Eq. (4). Using Eq. (3) again, we have $f_2 = -\alpha([S^x_{j,2}S^z_{j+1,1}S^x_{j+1,2}] - [S^x_{j,1}S^z_{j,2}S^x_{j+1,1}]) + a\alpha^2[S^z_{j+1,1}]$. Here note that $[S^x_{j,1}S^z_{j,2}S^x_{j+1,1}] = S^y_{j-1,2}S^z_{j,1}S^y_{j,2} + S^x_{j-1,2}S^z_{j,1}S^x_{j,2} + S^x_{j,1}S^z_{j,2}S^x_{j+1,1} + S^y_{j,1}S^z_{j,2}S^y_{j+1,1}$, $[S^z_{j+1,1}] = 2(S^z_{j-1,2} + S^z_{j+1,1})$ due to $n = 2$. We have $\Delta_2 = 2a(1+\alpha^2)$. The $(iL)^n$ results in the terms up to α^n; however, f_n does not have those in general. The term $\propto \alpha^3$ already vanishes in f_3, i.e., $f_3 = -2\alpha[S^y_{j,1}S^z_{j,2}S^z_{j+1,1}S^x_{j+1,2}] + \alpha^2(3a[S^y_{j+1,1}S^x_{j+1,2}] - [S^y_{j,2}S^z_{j+1,1}S^z_{j+1,2}S^x_{j+2,1}])$ leading to $(f_3, f_3) = 8a^4\alpha^2 \times (2+5\alpha^2)$.

We now confine ourselves to the limiting cases of α. For $\alpha^2 \ll 1$, we have $\Delta_3 = a(2+3\alpha^2)$. Since the term $\propto \alpha$ vanishes, we have $f_4 = \alpha^2 f_4^{\alpha^2}$, $\Delta_4 = (17/2)a\alpha^2$, $f_5 = \alpha^2 f_5^{\alpha^2}$, and $\Delta_5 = (48/17)a$. One of the largest spin-component products in f_4 with the square brackets for 1 and 5 spin and f_5 with those for 2, 4, and 6 spin is, respectively,

$$[S^x_{j,2}S^z_{j+1,1}S^z_{j+1,2}S^z_{j+2,1}S^x_{j+2,2}] \in f_4^{\alpha^2}, \qquad (8)$$

$$[S^y_{j,1}S^z_{j,2}S^z_{j+1,1}S^z_{j+1,2}S^z_{j+2,1}S^x_{j+2,2}] \in f_5^{\alpha^2}. \qquad (9)$$

We observe that the higher f_n has the larger spin-component product. However, once the largest term in f_n is composed of the on-site spin pairs alone, f_{n+1} does not spread anymore as long as α is small enough. This is because the interactions, see Eq. (5), are dominated by the *on-site* interactions. In fact, not shown here, f_6 and f_7 spread not beyond $\boldsymbol{S}_{j+2,2}$ are $\propto \alpha^2$ like Eqs. (8) and (9), respectively, and we have $\Delta_6 = 4a$, $\Delta_7 = (4448/867)a$. We thus expect to have $\Delta_{n\ge 8} = O(a)$. Here we notice a single *dip* at $n = 4$ in the $\{\Delta_n\}$ such as $\{0.005, 0.505, 0.508, 0.021, 0.706, 1, \ldots\}$ for $\alpha^2 = 0.01$.

The dimensions of the $\{\Delta_n\}$ are infinite when the energy delocalization takes place. Now we make an approximation for summing up the continued fraction where $\Delta_{n\ge l+2} = \Delta_{l+1}$ with l being an order at a dip, namely,

$$\frac{\Delta_{l+1}}{z+}\,\frac{\Delta_{l+2}}{z+}\,\frac{\Delta_{l+3}}{z+}\cdots = \frac{-z + (z^2 + 4\Delta_{l+1})^{1/2}}{2}. \qquad (10)$$

This approximation is valid as long as a dip is very steep in the infinite $\{\Delta_n\}$. Inserting the $\{\Delta_n\}$ for $\alpha^2 = 0.01$ together with Eq. (10) with $l = 4$ in Eq. (1), we obtain $\mathrm{Re}\overline{a}_0(-i\omega^+)$ which has well-defined peaks around $\omega = \pm\alpha/2, \pm 1$ with finite widths. This is consistent with the $\Delta_4 = 0$-like features with $\delta(\omega \pm \alpha/2)$ and $\delta[\omega \pm (1 + 3\alpha^2/2)^{1/2}]$, and survive in the Heisenberg model.

For $\alpha^2 \gg 1$, we have $\Delta_3 = 5a$ and $f_4 = a\alpha^3 f_4^{\alpha^3} + 3\alpha^2[S^z_{j+1,1}]$ leading to $\Delta_4 = (16a/5)\alpha^2 + 68a/25$ and $f_5 = a\alpha^4 f_5^{\alpha^4} + \alpha^3 f_5^{\alpha^3}$, where $f_5^{\alpha^3}$ includes the square brackets for 2, 4, and 6 spin. One of the largest spin-component products is

$$[S^y_{j,2}S^z_{j+1,1}S^z_{j+1,2}S^z_{j+2,1}S^z_{j+2,2}S^x_{j+3,1}] \in f_5^{\alpha^3}. \qquad (11)$$

We have $\Delta_5 = (4/5)a\alpha^2 + (1647/400)a$ and thus associate Eq. (3) with $f_6 = O(\alpha^5)$; however, such a term vanishes and we obtain $f_6 = a\alpha^4 f_6^{\alpha^4}$ leading to $\Delta_6 = (345/16)a$, $f_7 = a\alpha^5 f_7^{\alpha^5}$, and $\Delta_7 = (86/23)a\alpha^2$. One of the largest spin-component products in f_6 and f_7 is, respectively,

$$[S^x_{j+1,1}S^z_{j+1,2}S^z_{j+2,1}S^z_{j+2,2}S^x_{j+3,1}] \in f_6^{\alpha^4}, \qquad (12)$$

$$[S^y_{j,2}S^z_{j+1,1}S^z_{j+1,2}S^z_{j+2,1}S^z_{j+2,2}S^x_{j+3,1}] \in f_7^{\alpha^5}. \qquad (13)$$

Since the interactions, see Eq. (5), are dominated by the *intersite* interactions, the operators including $\boldsymbol{S}_{j+3,1}$ commute with the second term of the rhs of Eq. (5). It follows as shown in Eqs. (11)–(13) that $f_{n\geq 8}$ does not spread beyond $\boldsymbol{S}_{j+3,1}$ and that we expect to have $f_{n\geq 8} \propto \alpha^{n-2}$ and then $\Delta_{n\geq 8} \propto \alpha^2$. Here we notice two *dips* at $n = 3, 6$ in the $\{\Delta_n\}$ such as $\{50, 50.5, 1.25, 80.7, 21.0, 5.39, 93.5, \ldots\}$ for $\alpha^2 = 100$. The first dip at $n = 3$ results in the peaks at $\omega = 0, \pm|\alpha|$ and the second dip at $n = 6$ divides those into two structures, respectively. This splitting is not observed in the Heisenberg model. We have $\mathrm{Re}\overline{a}_0(-i\omega^+) = (\pi/8)\{\delta[\omega \pm (|\alpha| \pm \omega_0)] + 2\delta(\omega \pm \omega_0)\}$ with $\omega_0 = (\Delta_3/10)^{1/2} = 0.354$ as $|\alpha| \to \infty$.

Finally, we study the *Heisenberg model* with $\boldsymbol{S}_{i,1(2)} = (S^x_{i,1(2)}, S^y_{i,1(2)}, S^z_{i,1(2)})$. We have the same f_1 leading to $\Delta_1 = 2a\alpha^2$ as in the *XY* model. We further have $f_2 = -\alpha([S^z_{j,1}S^x_{j,2}S^x_{j+1,1}] - [S^x_{j,1}S^z_{j,2}S^x_{j+1,1}] + [S^x_{j,2}S^z_{j+1,1}S^x_{j+1,2}] - [S^x_{j,2}S^x_{j+1,1}S^z_{j+1,2}]) + a\alpha^2[S^z_{j+1,1}]$ leading to $\Delta_2 = 4a + 2a\alpha^2$. We have no term $\propto \alpha^3$ in f_3 as in the *XY* model; however, the spin flip effect of Eq. (5) results in many terms as $f_3 = \alpha f_3^{\alpha} + \alpha^2 f_3^{\alpha^2}$, where f_3^{α} and $f_3^{\alpha^2}$ have the square brackets for 2 and 4 spin. We have $(f_3, f_3) = 24a^4\alpha^2(4 + 3\alpha^2)$.

For $\alpha^2 \ll 1$, we have $\Delta_3 = 6a + (3a/2)\alpha^2$ and $f_4 = -6a\alpha f_4^{\alpha} + \alpha^2 f_4^{\alpha^2}$, where $f_4^{\alpha} = [S^z_{j,1}S^x_{j,2}S^x_{j+1,2}] - [S^x_{j,1}S^z_{j,2}S^x_{j+1,2}] + [S^x_{j,1}S^z_{j+1,1}S^x_{j+1,2}] - [S^z_{j,1}S^x_{j+1,1}S^x_{j+1,2}]$ and $f_4^{\alpha^2}$ has the square brackets for 1, 3, and 5 spin. We then have $\Delta_4 = 6a + (91a/12)\alpha^2$ and $f_5 = -\alpha f_5^{\alpha} + \alpha^2 f_5^{\alpha^2}$, where $f_5^{\alpha} = 24a^2[S^y_{j,1}S^x_{j+1,2}]$ and $f_5^{\alpha^2}$ has the square brackets for 2, 4, and 6 spin. The 6-spin alignments are with three on-site spin pairs around the site j:

$$\begin{aligned}
&(S_{j-2,1}S_{j-2,2}S_{j-1,1}S_{j-1,2}S_{j,1}S_{j,2}), \\
&(S_{j-1,1}S_{j-1,2}S_{j,1}S_{j,2}S_{j+1,1}S_{j+1,2}), \qquad (14)\\
&(S_{j,1}S_{j,2}S_{j+1,1}S_{j+1,2}S_{j+2,1}S_{j+2,2}).
\end{aligned}$$

We further have $\Delta_5 = 4a + (299a/12)\alpha^2$ and $f_6 = -\alpha f_6^{\alpha} + \alpha^2 f_6^{\alpha^2}$; however, $f_6^{\alpha} = i[\sum_i \boldsymbol{S}_{i,1} \cdot \boldsymbol{S}_{i,2}, f_5^{\alpha}]_- + 4af_4^{\alpha}$ does vanish indeed. The leading order in f_6 is thus α^2 and $f_6^{\alpha^2}$ has the square brackets for 1, 3, and 5 spin; hence we have $\Delta_6 = (2393a/24)\alpha^2$ and $f_7 = \alpha^2 f_7^{\alpha^2}$ with $f_7^{\alpha^2} = i[\sum_i \boldsymbol{S}_{i,1} \cdot \boldsymbol{S}_{i,2}, f_6^{\alpha^2}]_-$, which has the square brackets for 2, 4, and 6 spin. This leads to $\Delta_7 = (147\,898/7179)a$. The 6-spin alignments are identical to those in (14).

Similarly as seen in the *XY* model for this case, we expect to have $f_{n\geq 8} = \alpha^2 f^{\alpha^2}_{n\geq 8}$ with $f^{\alpha^2}_{\mathrm{even}\geq 8}$ composed of the square brackets for 1, 3, and 5 spin and $f^{\alpha^2}_{\mathrm{odd}\geq 9}$ of those for 2, 4, and 6 spin, and $\Delta_{n\geq 8} \propto a$. We observe a *dip* at $n = 6$ in the $\{\Delta_n\}$ such as $\{0.005, 1.005, 1.504, 1.519, 1.062, 0.249, 5.150, \ldots\}$ for $\alpha^2 = 0.01$. We use Eq. (10) with $l = 6$ and calculate Eq. (1) to show the spectra in Fig. 1 for $\alpha^2 = 0.05^2$, 0.1^2, and 0.15^2. The result clearly shows the excitations around $\omega = |\alpha|/2$, 1, and 2.

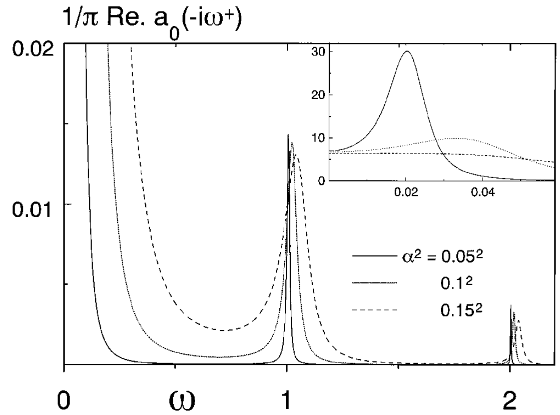

FIG. 1. The well-defined local excitations of $\omega = 1$ and 2 in $\mathrm{Re}\overline{a}_0(-i\omega^+)$ in the Heisenberg models with $\alpha^2 = 0.05^2$, 0.1^2, and 0.15^2. The energy unit is the on-site AF exchange integral J_{AF}. The inset shows the low energy part.

We explain the result shown in Fig. 1 mathematically and physically. We insert the $\{\Delta_n\}$ with $\Delta_6 = 0$, i.e., $\alpha = 0$ and $a = 1/4$ into Eq. (1) to yield

$$\overline{a}_0(z) = \frac{z(z^4 + 5z^2 + 4)}{z^2(z^4 + 5z^2 + 4) + g(z,\alpha)} \qquad (15)$$

with $g(z,\alpha) = 0$. This is simplified as $\overline{a}_0(z) = z^{-1}$ leading to $\mathrm{Re}\overline{a}_0(-i\omega^+) = \pi\delta(\omega)$, which means Eq. (6) is a constant of motion. In uniform chains we expect that Eq. (6) has a simple Lorentzian spectrum at $T = \infty$. On the contrary, when the alternation sets in, $g(z,\alpha) \propto \alpha^2$ for $\alpha^2 \ll 1$ splits $\delta(\omega)$ into two structures at $|\omega| = |\alpha|/2$ and produces well-defined peaks, no shoulders, at $|\omega| = 1, 2$ due to $z^4 + 5z^2 + 4 = (z - 2i)(z - i)(z + i)(z + 2i)$. This factor results from the dominant $\{\Delta_n\}$ up to $n = 5$: $\{\Delta_2, \Delta_3, \Delta_4, \Delta_5\} = \{4a, 6a, 6a, 4a\}$ with $a = 1/4$. The single *dip* in the present $\{\Delta_n\}$ reproduces these features, which are intrinsic for alternating chains not for uniform chains [27,30,31]. The spectra in Fig. 1 are compared to Eq. (7) in the Ising model and the results in the *XY* model for $\alpha^2 \ll 1$. The approximation of Eq. (10) gives finite widths for each spectrum superposed on the Lorentzian-like tail, the surviving $\delta(\omega)$, and almost α-independent value at $\omega = 0$.

We pay attention to the basis vectors $\{f_{n\leq 5}\}$ to classify these modes. The low energy mode around $\omega = |\alpha|/2$ is attributed to, e.g., the transition between the configurations (↑↓↓) and (↑↓↑) for a 3-spin alignment $(S_{j,1}S_{j,2}S_{j+1,1})$. The delocalization energy of unity corresponds to the single singlet-to-triplet local excitation like a soliton, $1 = 1/4 - (-3/4)$, in an on-site spin pair within a 6-spin alignment in (14), and twice to the double one in two on-site spin pairs within it. A similar consideration of (14) seems to make the triple one possible; however, it cannot be observed from the present $\{\Delta_n\}$, neither the quadruple one. While the ratio of the intensity at $\omega = 1, 2$ to that around $\omega = 0$ is $O(0.01)$. Therefore, it might follow that each spin in this system fluctuates not freely at all, but with very short-ranged and very weak correlations still existing at $T = \infty$.

The autocorrelation function is related to $S(\omega) = \sum_q S(q,\omega)$ of neutron scattering. The spectra shown in Fig. 1 are expected to be similar to the case at high temperatures, $k_B T \gg J_{AF}$. As α^2 increases for this regime, the Lorentzian-like tail masks the intensity around $\omega = |\alpha|/2$, while those around $\omega = 1, 2$ almost unchange and do survive against the growth of the tail. See Fig. 1. When the excitations are dispersionless at high temperatures, $I(\omega)$ in Raman scattering with $q = 0$ and $S(q,\omega)$ are then likely to show similar spectra. We further obtain $T\sum_q \mathrm{Im}\chi(q,\omega)/\omega = \mathrm{Re}\overline{a}_0(-i\omega^+)$. However, discussion on NMR relaxation rate $1/T_1$ is inappropriate because the almost α-independent value at $\omega = 0$ is an artifact of the approximation. Noting that the sign of α does not affect the $\{\Delta_n\}$, the dynamics of the AF-F and the AF-AF alternating chains are not distinguished. The energy unit of reported compounds is at most $O(10K)$. A candidate for observation of such spectra is, e.g., $(CH_3)_2CHNH_3CuBr_3$ with $\alpha = -0.54$ [7].

For $\alpha^2 \gg 1$, we have $\Delta_3 = 9a$ and $f_4 = -a\alpha^3 f_4^{\alpha^3}$, where $f_4^{\alpha^3} = i[\sum_i \boldsymbol{S}_{i-1,2} \cdot \boldsymbol{S}_{i,1}, f_3^{\alpha^2}]_-$ has the square brackets for 3 spin alone. We have $\Delta_4 = (20a/3)\alpha^2$ and $f_5 = a\alpha^4 f_5^{\alpha^4}$, where $f_5^{\alpha^4} = i[\sum_i \boldsymbol{S}_{i-1,2} \cdot \boldsymbol{S}_{i,1}, f_4^{\alpha^3}]_- + (20/3) f_3^{\alpha^2}$ has the square brackets for 2 and 4 spin. This leads to $\Delta_5 = (1148/135)a\alpha^2$. Different from the XY model for this case, we expect to have $f_6 = O(\alpha^5)$. Such a term as in Eq. (12) is of next leading order. No dip in the $\{\Delta_n\}$ at $n = 6$ is expected. We again observe a *dip* at $n = 3$ in the $\{\Delta_n\}$ such as $\{50.0, 51.0, 2.25, 167, \ldots\}$ for $\alpha^2 = 100$. We use Eq. (10) with $l = 3$ and calculate Eq. (1) to obtain $\mathrm{Re}\overline{a}_0(-i\omega^+)$ with each spectrum at $\omega = 0$ and around $\omega = \pm|\alpha|$ had nearly the same intensity and linewidth $\simeq |\alpha|^{-1}$. The spectra converge to $\mathrm{Re}\overline{a}_0(-i\omega^+) = (\pi/4)[\delta(\omega \pm |\alpha|) + 2\delta(\omega)]$ as $|\alpha| \to \infty$ when the alternation infinitely strengthens.

Equation (2) at $T = \infty$ for $\alpha = 0$, $\langle A(t)A\rangle/\langle A^2\rangle = 1$, decreases with an envelope of $\pm\exp(-t/\tau)$ for $\alpha^2 \ll 1$ both in the XY and the Heisenberg models. For $\alpha^2 \gg 1$, while it has an envelope of $+\exp(-t/\tau')$ in the Heisenberg model and converges to $(1 + \cos\alpha t)/2$, it has a slower tail in the XY model and converges to $\cos\omega_0 t(1 + \cos\alpha t)/2$.

In summary, we have investigated the dynamics of $S = 1/2$ alternating chains in limiting cases at $T = \infty$ in terms of the continued fraction formalism with recurrence relations, to obtain the well-defined delocalization models of J_{AF} and $2J_{AF}$. In this system very short-ranged and very weak correlations might survive dynamically at $T = \infty$. We have also raised the possibility of observing these modes in neutron and Raman scattering at high temperatures, $k_B T \gg J_{AF}$. These features are manifest in the dip in the continued fraction coefficients $\{\Delta_n\}$.

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[1] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983).
[2] F. D. M. Haldane, Phys. Rev. Lett. **50**, 1153 (1983).
[3] M. Hase, I. Terasaki, and K. Uchinokura, Phys. Rev. Lett. **70**, 3651 (1993).
[4] J. J. Borrás-Almenar, E. Coronado, J. Curely, R. Georges, and J. C. Gianduzzo, Inorg. Chem. **33**, 5171 (1994).
[5] H. Manaka, I. Yamada, and K. Yamaguchi, J. Phys. Soc. Jpn. **66**, 564 (1997).
[6] M. Hagiwara, Y. Narumi, K. Kindo, T. C. Kobayashi, H. Yamakage, K. Amaya, and G. Schumauch, J. Phys. Soc. Jpn. **66**, 1792 (1997).
[7] H. Manaka and I. Yamada, J. Phys. Soc. Jpn. **66**, 1908 (1997).
[8] M. Aïn, J. E. Lorenzo, L. P. Regnault, G. Dhalenne, A. Revcolevschi, B. Hennion, and Th. Jolicoeur, Phys. Rev. Lett. **78**, 1560 (1997).
[9] W. Duffy, Jr. and K. P. Barr, Phys. Rev. **165**, 647 (1968).
[10] M. C. Cross and D. S. Fisher, Phys. Rev. B **19**, 402 (1979).
[11] T. Nakano and H. Fukuyama, J. Phys. Soc. Jpn. **46**, 1679 (1980).
[12] K. Hida, Phys. Rev. B **45**, 2207 (1992).
[13] S. Kokado and N. Suzuki, J. Phys. Soc. Jpn. **66**, 3605 (1997).
[14] S. Kokado and N. Suzuki, J. Magn. Magn. Mater. **196–197**, 566 (1999).
[15] J. H. H. Perk and H. W. Capel, Physica (Amsterdam) **100A**, 1 (1980).
[16] J. H. Taylor and G. Müller, Physica (Amsterdam) **130A**, 1 (1985).
[17] K. Kawasaki, N. Maya, A. Kouzuki, and K. Nakamura, J. Phys. Soc. Jpn. **66**, 839 (1997).
[18] H. Mori, Prog. Theor. Phys. **34**, 399 (1965).
[19] H. Mori, Prog. Theor. Phys. **33**, 423 (1965).
[20] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[21] H. Okamoto, K. Nagano, T. Karasudani, and H. Mori, Prog. Theor. Phys. **66**, 53 (1981).
[22] M. H. Lee, Phys. Rev. B **26**, 2547 (1982).
[23] M. H. Lee, Phys. Rev. Lett. **49**, 1072 (1982).
[24] M. H. Lee and J. Hong, Phys. Rev. Lett. **48**, 634 (1982).
[25] J. Hong and M. H. Lee, Phys. Rev. Lett. **70**, 1972 (1993).
[26] M. H. Lee, I. M. Kim, and R. Dekeyser, Phys. Rev. Lett. **52**, 1579 (1984).
[27] J. Florencio, Jr. and M. H. Lee, Phys. Rev. B **35**, 1835 (1987).
[28] C. Lee and S. I. Kobayashi, Phys. Rev. Lett. **62**, 1061 (1989).
[29] S. Sen, Proc. R. Soc. London A **441**, 169 (1993).
[30] V. S. Viswanath and G. Müller, *The Recursion Method* (Springer, Berlin, 1994).
[31] S. Sen, Physica (Amsterdam) **222A**, 195 (1995).
[32] S. Sen and T. D. Blersch, Physica (Amsterdam) **253A**, 178 (1998).
[33] J. Hong and H.-Y. Kee, Phys. Rev. B **52**, 2415 (1995).
[34] H.-Y. Kee and J. Hong, Phys. Rev. B **55**, 5670 (1997).
[35] Equation (5) is also written in the literature as $H = J_{AF}\sum_i \boldsymbol{S}_{2i} \cdot \boldsymbol{S}_{2i+1} - \alpha\sum_i \boldsymbol{S}_{2i-1} \cdot \boldsymbol{S}_{2i}$, where Eq. (6) takes the form $A = S_{2l}^z + S_{2l+1}^z$.