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Exact results on spin dynamics and multiple quantum NMR dynamics in alternating spin-1/2 chains with XY-Hamiltonian at high temperatures

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We extend the picture of a transfer of nuclear spin-1/2 polarization along a homogeneous one-dimensional chain with the XY-Hamiltonian to the inhomogeneous chain with alternating nearest neighbour couplings and alternating Larmor frequencies. To this end, we calculate exactly the spectrum of the spin-1/2 XY-Hamiltonian of the alternating chain with an odd number of sites. The exact spectrum of the XY-Hamiltonian is also applied to study the multiple quantum (MQ) NMR dynamics of the alternating spin-1/2 chain. MQ NMR spectra are shown to have the MQ coherences of zero and ± second orders just as in the case of a homogeneous chain. The intensities of the MQ coherences are calculated.

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1. Introduction. The discovery of the exact solution of spin-1/2 homogeneous one-dimensional chains with the XY-Hamiltonian [1, 2] gives the observable and measurable features unraveling the NMR dynamics of spin-1/2 homogeneous one-dimensional chains [3]. Although most of the NMR experiments for one-dimensional spin chains appear to be well-explained by means of the nuclear spin dynamics on the homogeneous spin chains, NMR spin dynamics beyond the homogeneous chains has attracted the attention recently. For example, the experiments [4] demonstrate the propagation of spin wave excitations along the inhomogeneous spin chains, the mesoscopic echo has been observed [5] due to the reflections of the spin waves at the boundaries of the chain.

The presented paper is aimed at exploring the key differences of NMR of the homogeneous spin-1/2 chain from the inhomogeneous spin-1/2 chain. It may be difficult, however, to calculate the NMR responses for an inhomogeneous spin chain with a random variation of the nearest neighbour (NN) dipolar coupling. Thus, as a first step to unravel of the inhomogeneous effects, we treat the one-dimensional chain with an alternating spin-1/2 NN dipolar coupling and alternating Larmor frequencies.

In the approach [1, 2], the basic tool in exploring the 1D spin-1/2 XY-Hamiltonian is the Jordan-Wigner transformation of the original spin-1/2 XY-Hamiltonian to the Hamiltonian of the free fermions. In this way, the exact thermodynamics for alternating infinite chains with the spin-1/2 XY-Hamiltonian was explored in [6]. The report [7] presents the exact spectrum of the XY-Hamiltonian with alternating couplings on the finite rings. However, to our knowledge, the spectrum of the XY-Hamiltonian with alternating couplings on the open chains is lacking by now. This is what will be addressed in section 2. The derived exact spectrum of the spin-1/2 XY-Hamiltonian of the alternating open chains permits one to explain the transfer of the nuclear polarization along the alternating chains in section 3 and to calculate the MQ intensities of alternating spin-1/2 chains in section 4. The concluding section 5 draws the distinction of the NMR dynamics on alternating chains from that on the homogeneous ones.

2. Exact spectrum of spin-1/2 XY-Hamiltonian with alternating couplings on open chains. In this section, we derive the exact spectrum of the spin-1/2 XY-Hamiltonian

$$H = \sum_{n=1}^{N} \omega_n I_{nz} + \sum_{n=1}^{N-1} D_{n,n+1}\Big(I_{n,x}I_{n+1,x} + I_{n,y}I_{n+1,y}\Big) \qquad (1)$$

of the open chain of the odd number, N, of sites with the alternating NN coupling constants D_1 and D_2 and alternating Larmor frequencies ω_1 and ω_2, see Fig.1.

The nuclear spins are specified by the spin-1/2 operators $I_{n\alpha}$ at sites $n = 1, \ldots, N$ with projections $\alpha = x, y, z$. The Jordan-Wigner transformation [1]

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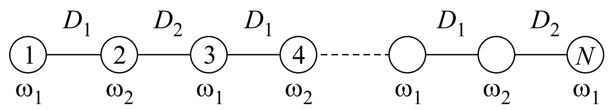

Fig.1. An open chain of the odd number, N, of spins with the alternating NN coupling constants D_1 and D_2 and alternating Larmor frequencies ω_1, ω_2

$$
\begin{gathered}
I_{n,-} = I_{n,x} - iI_{n,y} = (-2)^{n-1}\left(\prod_{l=1}^{l=n-1} I_{l,z}\right) c_n, \\
I_{n,+} = I_{n,x} + iI_{n,y} = (-2)^{n-1}\left(\prod_{l=1}^{l=n-1} I_{l,z}\right) c_n^+, \\
I_{n,z} = c_n^+ c_n - 1/2,
\end{gathered} \tag{2}
$$

from spin-1/2 operators $I_{n\alpha}$ to the creation (annihilation) operators c_n^+ (c_n) of the spinless fermions takes the Hamiltonian (1) into the Hamiltonian

$$
\begin{gathered}
H = \sum_{n=1}^{N} \omega_n \left(c_n^+ c_n - 1/2\right) + \\
+\frac{1}{2}\sum_{n=1}^{N-1} D_{n,n+1}\left\{c_n^+ c_{n+1} + c_{n+1}^+ c_n\right\},
\end{gathered} \tag{3}
$$

or in the matrix notations as

$$
H = \frac{1}{2}\mathbf{c}^+ (D + 2\Omega)\,\mathbf{c} \; - \; \frac{1}{2}\sum_{n=1}^{N}\omega_n. \tag{4}
$$

In Eq.(4), we denote the row vector $\mathbf{c}^+ = (c_1^+, \ldots, c_N^+)$, the column vector $\mathbf{c} = (c_1, \ldots, c_N)^t$ (the superscript t represents the transpose) and specify the matrices Ω and D as

$$
\Omega = \begin{bmatrix}
\omega_1 & 0 & 0 & \ldots & 0 & 0 \\
0 & \omega_2 & 0 & \ldots & 0 & 0 \\
0 & 0 & \omega_1 & \ldots & 0 & 0 \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\
0 & 0 & 0 & \ldots & \omega_2 & 0 \\
0 & 0 & 0 & \ldots & 0 & \omega_1
\end{bmatrix},
$$

$$
D = \begin{bmatrix}
0 & D_1 & 0 & \ldots & 0 & 0 \\
D_1 & 0 & D_2 & \ldots & 0 & 0 \\
0 & D_2 & 0 & \ldots & 0 & 0 \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\
0 & 0 & 0 & \ldots & 0 & D_2 \\
0 & 0 & 0 & \ldots & D_2 & 0
\end{bmatrix}. \tag{5}
$$

Diagonalization of the matrix $D + 2\Omega$ is performed by the unitary transformation

$$
D + 2\Omega = U\Lambda U^+, \quad \Lambda = \mathrm{diag}\{\lambda_1, \ldots, \lambda_N\}, \tag{6}
$$

so that the new fermion operators γ_k^+ and γ_k introduced by the relations

$$
c_n^+ = \sum_{n=1}^{N} u_{n,k}^* \gamma_k^+, \quad c_n = \sum_{n=1}^{N} u_{n,k}\gamma_k \tag{7}
$$

bring the Hamiltonian (4) into the Hamiltonian

$$
H = \frac{1}{2}\sum_{k=1}^{N} \lambda_k \gamma_k^+ \gamma_k - \frac{1}{2}\sum_{n=1}^{N}\omega_n \tag{8}
$$

with energies $\frac{1}{2}\lambda_\nu$ of the free fermion waves.

To go further in explicit calculations it is necessary to find the eigenvalues λ_ν and eigenvectors $|u_\nu\rangle = (u_{1\nu}, u_{2\nu}, \ldots, u_{N\nu})^t$ of the matrix $D + 2\Omega$,

$$
(D + 2\Omega)|u_\nu\rangle = \lambda_\nu |u_\nu\rangle. \tag{9}
$$

In Eq. (9), the components $u_{n,\nu}$ at the even sites $n = 2, 4, \ldots, N-1$ obey the equations

$$
\begin{gathered}
D_1 u_{2k-1,\nu} + 2\omega_2 u_{2k,\nu} + D_2 u_{2k+1,\nu} = \lambda_\nu u_{2k,\nu}, \\
k = 1, 2, \ldots, \frac{N-1}{2}.
\end{gathered} \tag{10}
$$

We now fix the indices k and ν in Eq. (10) and eliminate from Eq. (10) the components $u_{2k-1,\nu}$ and $u_{2k+1,\nu}$ at the odd sites $(2k-1)$ and $(2k+1)$ by writing down the equations for the $u_{2k-1,\nu}$, $u_{2k+1,\nu}$ from Eq. (9) as

$$
\begin{gathered}
D_2 u_{2k-2,\nu} + 2\omega_1 u_{2k-1,\nu} + D_1 u_{2k,\nu} = \lambda_\nu u_{2k-1,\nu}, \\
D_2 u_{2k,\nu} + 2\omega_1 u_{2k+1,\nu} + D_1 u_{2k+2,\nu} = \lambda_\nu u_{2k+1,\nu}.
\end{gathered} \tag{11}
$$

Substituting (11) into (10), we get the relations for the amplitudes $u_{n,\nu}$ at the even sites,

$$
\begin{gathered}
g u_{2k-2,\nu} + g u_{2k+2,\nu} = u_{2k,\nu}, \\
k = 1, 2, \ldots, (N-1)/2,
\end{gathered} \tag{12}
$$

with the spatially independent coupling constant

$$
g = \frac{D_1 D_2}{(\lambda_\nu - 2\omega_1)(\lambda_\nu - 2\omega_2) - D_1^2 - D_2^2}. \tag{13}
$$

The derivation of (12), (13) is a characteristic way in the theory of the real space renormalization [8] which eliminates the half of the degrees of freedom belonging to the odd sites yielding the field equations on the (even) lattice sites with doubled lattice constant. Eqs. (12), (13) govern the amplitudes $u_{2,\nu}, u_{4,\nu}, \ldots, u_{N-1,\nu}$ (including the amplitudes $u_{2,\nu}$, $u_{N-1,\nu}$ at the border sites 2 and $N-1$) if we introduce the additional sites $n=0$ and $n=N+1$ and put there

$$u_{0,\nu}=0, \quad u_{N+1,\nu}=0. \tag{14}$$

Eq. (14) implies the cutting off the lattice at the sites 1 and N, thus, preventing the fermions from escaping the lattice shown on Fig.1. The solution of Eq. (12) conditioned by Eq. (14) read

$$u_{2k,\nu}=A_\nu \sin\left(\frac{2\pi k\nu}{N+1}\right), \quad k,\nu=1,2,\dots,\frac{N-1}{2}, \tag{15}$$

with eigenvalues

$$\lambda_\nu^{(\pm)}=\omega_1+\omega_2\pm\sqrt{(\omega_1-\omega_2)^2+D_1^2\,\Delta_\nu},$$
$$\nu=1,2,\dots,\frac{N-1}{2}, \tag{16}$$
$$\Delta_\nu=1+2\delta\cos\left(\frac{2\pi\nu}{N+1}\right)+\delta^2, \quad \delta=D_2/D_1.$$

Eq. (16) gives $2[(N-1)/2]=N-1$ eigenvalues λ_ν since for each index $\nu=1,2,\dots,N$ and for each superscript $(+)$ and $(-)$ the relationship $\lambda_\nu^{(\pm)}=\lambda_{N+1-\nu}^{(\pm)}$ holds. It is convenient to arrange $(N-1)$ distinct eigenvalues λ_ν as

$$\lambda_\nu=\begin{cases}\lambda_\nu^{(+)}, & \text{for } \nu=1,2,\dots,(N-1)/2\\ \lambda_\nu^{(-)}, & \text{for } \nu=(N+3)/2,(N+5)/2,\dots,N.\end{cases} \tag{17}$$

In accord with the enumeration of the eigenvalues λ_ν in Eq. (17), the missed N-th eigenvalue λ_ν stands for the index $\nu=(N-1)/2+1=(N+1)/2$ (recall that N is odd). To find the eigenvalue $\lambda_{(N+1)/2}$ and the eigenvector $u_{n,(N+1)/2}$ at even sites n, the use is made of the properties (see Eq. (15))

$$u_{n,(N+1)/2}=0, \quad n=0,2,4,\dots,N-1,N+1. \tag{18}$$

By the condition $u_{N-1,(N+1)/2}=0$, the N-th equation from Eq. (9) gives immediately the sought eigenvalue

$$\lambda_{(N+1)/2}=2\omega_1. \tag{19}$$

It remains to find the amplitudes $u_{k,(N+1)/2}$ at the odd sites. The sought amplitudes obey the relations

$$D_1u_{2k-1,\nu}+D_2u_{2k+1,\nu}=0,$$
$$\nu=\frac{N+1}{2},\ k=1,2,\dots,\frac{N-1}{2}. \tag{20}$$

Eq. (20) gives the components of the eigenvector $u_{n,(N+1)/2}$ belonging to odd sites n (up to the normalization coefficient B),

$$u_{n,(N+1)/2}=B\cdot(-\delta)^{(N-n)/2}, \quad n=1,3,\dots,N. \tag{21}$$

Calculating the normalization coefficients A_ν in Eq. (15) and the coefficient B in Eq. (21), one finds all N eigenvectors $|u_\nu\rangle$ and all N eigenvalues λ_ν of the Hamiltonian (8),

$$\lambda_\nu=\begin{cases}\omega_1+\omega_2+\sqrt{(\omega_1-\omega_2)^2+D_1^2\Delta_\nu}, & \nu=1,2,\dots,\frac{N-1}{2}\\ 2\omega_1, & \nu=\frac{N+1}{2}\\ \omega_1+\omega_2-\sqrt{(\omega_1-\omega_2)^2+D_1^2\Delta_\nu}, & \nu=\frac{N+3}{2},\frac{N+5}{2},\dots,N\end{cases} \tag{22}$$

For all indices $\nu=1,\dots,N$ except the index $\nu=(N+1)/2$, the eigenvector $|u_\nu\rangle$ has the elements

$$u_{j,\nu}=\begin{cases}A_\nu\frac{D_1}{\lambda_\nu-2\omega_1}\left[\delta\sin\left(\frac{\pi\nu(j-1)}{N+1}\right)+\sin\left(\frac{\pi\nu(j+1)}{N+1}\right)\right], & j=1,3,5,\dots,N\\ A_\nu\sin\left(\frac{\pi\nu j}{N+1}\right), & j=2,4,\dots,N-1\end{cases} \tag{23}$$

and the normalization coefficient

$$A_\nu=\frac{2|\lambda_\nu-2\omega_1|}{\sqrt{N+1}}\frac{1}{\sqrt{(\lambda_\nu-2\omega_1)^2+D_1^2\,\Delta_\nu}}. \tag{24}$$

By Eq. (21), the elements of the eigenvector $|u_{(N+1)/2}\rangle$ read

$$u_{j,(N+1)/2}=\begin{cases}B\cdot(-\delta)^{(N-j)/2}, & j=1,3,5,\dots,N\\ 0, & j=2,4,\dots,N-1\end{cases} \tag{25}$$

with the normalization coefficient

$$B=\left(\frac{\delta^2-1}{\delta^{N+1}-1}\right)^{1/2}. \tag{26}$$

3. Spin-wave propagation in open chains with alternating couplings. The spectrum of Eqs. (22)–(26) of the XY Hamiltonian (8) can now be applied to describe how the polarization at a single site of the alternating chain with equal Larmor frequencies ω_n changes over the time.

Let the initial polarization is on the single site j, hence, the spin dependent part of the initial density matrix at the high temperature approximation is, see [9],

$$\rho(0) = I_{j,z} = c_j^+ c_j - 1/2. \tag{27}$$

Given the initial density matrix (27) and the Hamiltonian H (8), the Liouville-von Neumann equation ($\hbar = 1$)

$$i\frac{\partial \rho}{\partial t} = \left[H, \rho\right], \tag{28}$$

is solved in terms of the fermion operators (7) as follows,

$$\rho(t) = e^{-iHt} I_{j,z} e^{iHt} = -\frac{1}{2} + \sum_{1 \le l,m \le N} u_{j,l}^* u_{j,m} e^{-\frac{i}{2}(\lambda_l - \lambda_m)t} \gamma_l^+ \gamma_m. \tag{29}$$

Denoting the polarization at j'-th spin at the time moment t by $\langle I_{j',z}\rangle(t)$ and invoking Eq. (27), we get

$$\frac{\langle I_{j',z}\rangle(t)}{\langle I_{j,z}\rangle(0)} = \frac{tr\{\rho(t) I_{j',z}\}}{tr\{I_{j,z}^2\}} = \left| \sum_{1 \le \nu \le N} u_{j,\nu}^* u_{j',\nu} e^{-\frac{i}{2}\lambda_\nu t} \right|^2. \tag{30}$$

Fig.2 shows the time course of the polarization $\langle I_{1,z}\rangle(t)/\langle I_{1,z}\rangle(0)$ at site $j' = 1$ when the initial polarization is also at site 1 and $D_2 = \frac{3}{2}D_1$. Just as in the case of the polarization dynamics on the homogeneous chain with equal couplings, $D_2 = D_1$, [3], the dynamics of the polarization of the alternating chain can be regarded as the propagation of the spin wave packet starting at site 1 and bouncing back and forth at the chain ends To calculate the return time t_R for the wave packet to reappear at site 1, we, first, determine the group velocity of the waves described by the Hamiltonian H (8) with the dispersion law $\frac{1}{2}\lambda_\nu$ (16) and equal Larmor frequencies, $\omega_1 = \omega_2$. By specifying the wave vector $p = 2\pi\nu/(N+1)$, $\nu = 1, 2, \ldots, (N-1)/2$, the dispersion law of Eq. (16) written down as $\frac{1}{2}\lambda_\nu = \frac{1}{2}D_1\sqrt{1 + 2\delta\cos(p) + \delta^2}$ allows one to calculate the sought group velocity

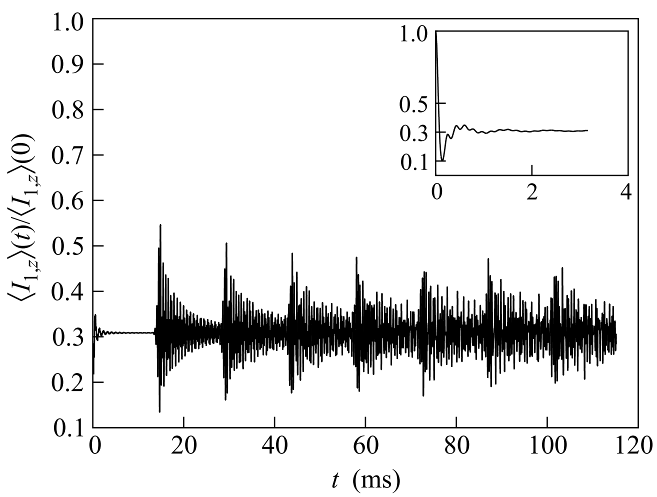

Fig.2. Time course of the polarization $\langle I_{1,z}\rangle(t)/\langle I_{1,z}\rangle(0)$ of the first spin, Eq. (30), for the chain of 201 spins interacting with coupling coefficients $D_1 = 2\pi \cdot 4444\ \mathrm{s}^{-1}$ and $D_2 = 2\pi \cdot 6666\ \mathrm{s}^{-1}$ and equal Larmor frequencies. The initial polarization is on the first spin. Insert shows the time course of the polarization on an earlier time interval $0 \le t < 4\,\mathrm{ms}$

$$v = \max_p \left\{ a\,\frac{d\lambda(p)}{2\,dp} \right\} = \frac{aD_1}{2}, \tag{31}$$

where a is the lattice constant and the coupling constant D_1 is the minimal coupling constant among the two coupling constants D_1 and D_2. Thus, for $N = 201$-chain with coupling constants $D_1 = 2\pi \cdot 4444\,\mathrm{s}^{-1}$ and $D_2 = 2\pi \cdot 6666\,\mathrm{s}^{-1}$, the time of the first returning of the wave packet to site 1 becomes, see Fig.2,

$$t_R = \frac{2(N-1)}{D_1} \approx 14.5\,\mathrm{ms}. \tag{32}$$

The traveling waves of the spin polarization $\langle I_{j,z}\rangle(t)/\langle I_{1,z}\rangle(0)$ are shown on Fig.3.

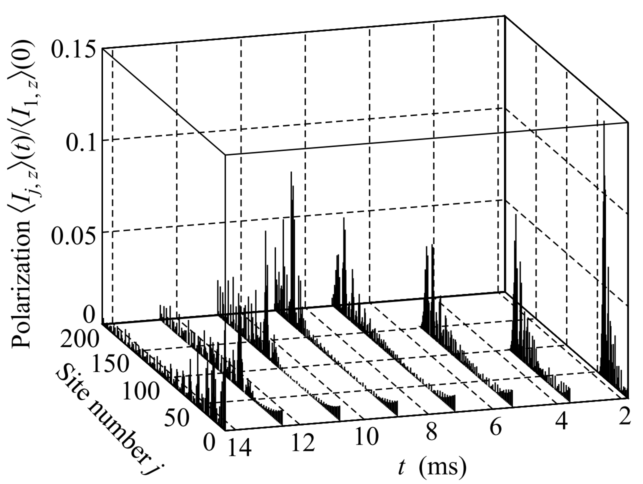

Fig.3. Propagation of the spin-wave packet along the 201-site open chain. The packet starts at boundary site $j = 1$

4. Intensities of MQ coherences in open chains with alternating couplings. The exact spectrum of Eqs. (22)–(26) of the Hamiltonian H (8) provides us with the technique for determining of the multi-quantum dynamics in an alternating open chain. Again, we take the initial density matrix $\rho(0)$ (27) and calculate how the MQ coherences develop in the spin system of the alternating spin-1/2 chain. MQ NMR dynamics of the nuclear spins coupled by nearest neighbour dipolar interactions are described by the Hamiltonian [10, 11],

$$H_{\rm MQ} = \frac{1}{2}\sum_{n=1}^{N-1} D_{n,n+1}\left\{I_{n,+}I_{n+1,+} + I_{n,-}I_{n+1,-}\right\}. \quad (33)$$

The Hamiltonian H (33) takes the form of the exactly solvable Hamiltonian H (1) (with the Larmor frequencies $\omega_n = 0$ for all sites) by making use of the unitary transformation [11] acting on the even sites,

$$Y = \exp(-i\pi I_{2,x})\exp(-i\pi I_{4,x})\cdots\exp(-i\pi I_{N-1,x}), \quad (34)$$

so that $YH_{\rm MQ}Y^+ = H(\text{ Eq. (1)}; \{\omega_n = 0\})$. In addition, the transformation Y brings the initial density matrix (27) to the form

$$\bar\rho(0) = YI_zY^+ = \sum_{n=1}^{N}(-1)^{n-1}I_{n,z}, \quad (35)$$

where we introduce the total polarization $I_z = \sum_{n=1}^{N} I_{n,z}$. The Liouville-von Neumann Eq. (28) with Hamiltonian H (33) and the initial density matrix (27) gives the intensities $G_n(t)$ of $n = 0$ and $n = \pm 2$ orders, just as in the case of the homogeneous chain with the conservation condition [11, 12],

$$G_0(t) + G_2(t) + G_{-2}(t) = 1, \quad (36)$$

and

$$G_0(t) = \frac{1}{N}\sum_{n=1}^{N}\cos^2(\lambda_\nu t), \qquad G_{\pm 2}(t) = \frac{1}{2N}\sum_{n=1}^{N}\sin^2(\lambda_\nu t). \quad (37)$$

Fig.4 demonstrates the development of the 2Q coherence on the alternating $N = 201$-chain with couplings $D_1 = 2\pi\cdot 4444\ {\rm s}^{-1}$ and $D_2 = 2\pi\cdot 6666\ {\rm s}^{-1}$, thus $\delta = D_2/D_1 = 1.5$. As time proceeds, the regular course of the intensities $G_{\pm 2}(t)$ is transformed to the erratic temporary behaviour just as in the case of the homogeneous lattice with $D_1 = D_2$ [13]. For $D_1 = D_2$, Eq. (37) reproduces exactly the results for the intensities of the 2Q coherence of Ref. [11].

5.Conclusion. Sensitivity of the NMR spin polarization dynamics to the spatially periodic short-distance inhomogeneity of the lattice has been explored in the previous sections relying on the exact spectrum of the spin-1/2 XY-Hamiltonian on the alternating open chain with an odd number of sites.

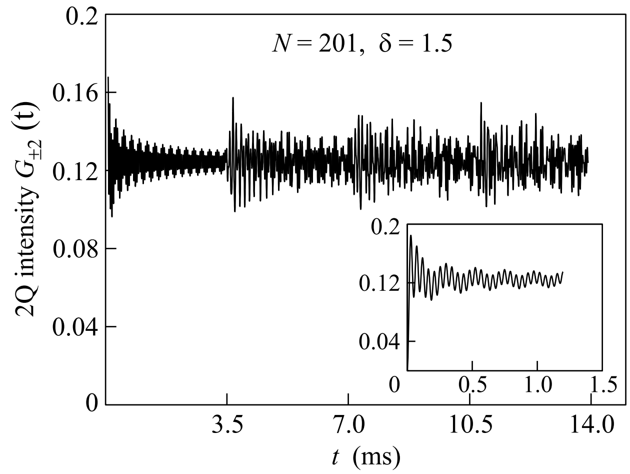

Fig.4. Time course of the intensities of the 2Q coherence, Eq. (37), in the chain of $N = 201$ spins interacting with coupling constants $D_1 = 2\pi\cdot 4444{\rm s}^{-1}$ and $D_2 = 2\pi\cdot 6666{\rm s}^{-1}$ and zero Larmor frequencies. Insert shows an early development of 2Q coherence over times $t < 1.5$ ms

If the spin polarization is prepared initially on the single site, the spin dynamics on the non-homogeneous lattice is represented as the propagation of the spin wave packet. The velocity of the wave packet on the non-homogeneous lattice is controlled by the minimal coupling constant $\min\{D_1, D_2\}$. In the extreme situation $D_1 \ll D_2$ ($D_1, D_2 \neq 0$), the spin polarization stays fixed on the initial site.

As time proceeds, the regular spin propagation along the alternating chain is transformed to the erratic one.

The time scale of the regular behaviour of the 2Q intensities $G_{\pm 2}(t)$ is 4 times shorter than the time scale of the regular behaviour of the polarization $\langle I_{j,z}\rangle(t)/\langle I_{1,z}\rangle(0)$, as it can be seen by a comparison of Fig.4 with Fig.2. The same effect happens in the case of the spin dynamics on the homogeneous chain [13]. The effect is caused by two reasons. First, the velocity of the propagation of the 2Q coherence is doubled as compared with the velocity of the propagation of the spin polarization, as it is obvious by the comparison of the dispersion law in Eq. (37) with the dispersion law in Eq. (30). Secondly, the two-spin local excitations of the 2Q coherence travel the path N before the two-spin excitations return back on the N-site chain. The path N should be compared with the path $2N$ traveled by the single local spin excitation before it reappears on the initial site of the chain.

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