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Dipolar coupling spin dynamics: perturbation approach

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Abstract
A perturbation method is presented which can be applied to the description of a wide range of physical problems that deal with dynamics of dipolarly coupled spins in solids. The method is based on expansion of $e^{\mathcal{A}+\mathcal{B}}$ in a perturbation series. As an example of the application of the method, the multiple-quantum coherence dynamics in three- and four-spin clusters are considered. The calculated 0Q and 2Q intensities versus the duration of the preparation period give close agreement with exact results and simulations data. The exact solutions for J_{0Q} and J_{2Q} coherences in four-spin systems are obtained.

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1. Introduction

Dipolar coupling spin dynamics is of great interest concerning the general problems of physics of many bodies and nuclear magnetic resonance (NMR) [1, 2]. In solids, the evolution of a spin system under the dipole–dipole interaction (DDI) involves many spins and leads to unsolvable problems. Even the analysis using the numerical calculation becomes difficult because the number of states $N = 2^n$ is growing exponentially with the increase of n. So in existing theories, only macroscopic characteristics such as spin–spin relaxation times, the second and the fourth moments of resonance lines were taken into account [1]. These difficulties are very clearly displayed in multiple-quantum (MQ) spin dynamics. The MQ phenomena involve various multiple-spin transitions between the Zeeman energy levels and form MQ coherence at times $t > \omega_d^{-1}$, where ω_d is the characteristic frequency of DDI [3]. The problem in analytical description of MQ is that the different modes of coherence grow at different times, with higher modes requiring longer excitation times than lower modes [4]. Hence, $\omega_d t > 1$ is not a small parameter, and, at first glance, perturbation theory methods cannot be used to study MQ dynamics. Indeed, only simple exactly solvable models of the spin system such as two and three dipolar coupling spin-1/2 [5, 6] or one-dimensional linear chain spin [7] system were analysed theoretically. The last achievement in this direction is the model with identical DDI coupling constant for all spin pairs [8, 9]. Note that the simplified calculations essential for the case of identical DDI coupling constant have been already mentioned [10]. Such approaches cannot describe MQ processes except for zeroth and second-order coherences. Thus, the development of a method that can successfully represent important features of MQ dynamics with coherences larger than 0Q and 2Q is needed.

Importance of the analytical description of the MQ processes in solids is that involving the excitation of collections of dipolar coupling spins can provide important structural as well as the spin dynamics information. Moreover, during the past few years, the NMR is considered as the best candidate [11–13] for experimental realization of quantum information processes. Recently, by using MQ techniques a new method of preparing pseudopure states in a cluster of dipolar-coupled nuclear spins was developed [14] and the dynamics of entanglement in solids was investigated [6]. Thus, investigation of the quantum information processes with MQ methods is of current interest.

We present a perturbation approach to the problem of dipolar coupling spin dynamics in solids. DDI of all spins was divided into several groups that are characterized by the identical DDI coupling constants. Since the magnitudes of the dipolar coupling constants vary inversely with the cube of internuclear distance, the coupling constants are different for these groups.

Our main idea is to take into account in MQ NMR dynamics influence of the groups with different degree of accuracy. Spin groups with smaller DDI coupling constants can be considered as perturbation (spins located far apart) while the nearest neighbours are taken into account exactly. As the result, we can develop a perturbation method that allows obtaining the description of the MQ with a large coherence evolution under DDI in an analytical form. The proposed approach will be a powerful method to describe wide ranges of physical problems that deal with dynamics of dipolar coupled spins in solid. On one hand, this approach uses the advantages of exactly solvable models [9, 15]. On the other hand, it simplifies calculations by using a perturbation technique. Results will show that the perturbation method can be applied to solve complex spin-dynamics problem and to obtain the solution in an analytical form. The method is based on the differential method [16, 17] that expresses $\mathrm{e}^{\mathcal{A}+\mathcal{B}}$ as an infinite product of exponential operators [17]. In the case where the norm of operator $\mathcal{B}$ is small then one of operator $\mathcal{A}$, $\|\mathcal{B}\| < \|\mathcal{A}\|$, we will try to obtain the perturbation series up to second-order approximation ($\|\mathcal{B}\|/\|\mathcal{A}\|$). Then the problems in the description of the MQ dynamics will be significantly simplified.

2. Theory

Let us consider a spin system with Hamiltonian, $\mathcal{H}$ which includes only two parts with different DDI constants α and β: $\mathcal{H} = \mathcal{A} + \mathcal{B}$, where $\mathcal{A} = \alpha A$ and $\mathcal{B} = \beta B$, $\alpha = \|\mathcal{A}\|$ and $\beta = \|\mathcal{B}\|$ are the norms of operators $\mathcal{A}$ and $\mathcal{B}$, respectively ($\alpha > \beta$ and $[A, B] \neq 0$). The evolution of the spin system is governed by propagator

$$\mathrm{e}^{-\mathrm{i}t\mathcal{H}} = \mathrm{e}^{-\mathrm{i}t(\alpha A+\beta B)}. \tag{1}$$

We seek to express (1) as a series such that

$$\mathrm{e}^{-\mathrm{i}t(\alpha A+\beta B)} = \mathrm{e}^{-\mathrm{i}t\beta B}\sigma(t), \tag{2}$$

where operator $\sigma(t)$ obeys the differential equation [17, 18]

$$\mathrm{i}\frac{\mathrm{d}\sigma(t)}{\mathrm{d}t} = \alpha A^{(0)}(t)\sigma(t), \tag{3}$$

with initial condition

$$\sigma(0) = 1 \tag{4}$$

and $A^{(0)}(t) = \mathrm{e}^{-\mathrm{i}t\beta B} A\, \mathrm{e}^{\mathrm{i}t\beta B}$. Assume that $\alpha t \geqslant 1$ and $\beta t < 1$. First, we will restrict ourselves by keeping only first-order terms that are linearly proportional to β. This leads to

$$\mathrm{i}\frac{\mathrm{d}\sigma^{(0)}(t)}{\mathrm{d}t} = \alpha(A + \mathrm{i}t\beta[B, A])\sigma^{(0)}(t). \tag{5}$$

Solving equation (5) we obtain

$$\sigma^{(0)}(t) = \mathrm{e}^{-\mathrm{i}\alpha\left(At+\mathrm{i}\frac{t^2}{2}\beta[B,A]\right)}. \tag{6}$$

However, equation (6) contains terms that are proportional to a power of β. Then we will continue the expansion:

$$\sigma^{(0)}(t) = \mathrm{e}^{\alpha\frac{t^2}{2}\beta[B,A]}\sigma^{(1)}(t). \tag{7}$$

Differentiating expressions (7) results in the differential equation for $\sigma^{(1)}(t)$:

$$\mathrm{i}\frac{\mathrm{d}\sigma^{(1)}(t)}{\mathrm{d}t} = \alpha A^{(1)}(t)\sigma^{(1)}(t) \tag{8}$$

with initial condition $\sigma^{(1)}(0) = 1$ and $A^{(1)}(t) = \mathrm{e}^{-\alpha\frac{t^2}{2}\beta[B,A]} A\, \mathrm{e}^{-\alpha\frac{t^2}{2}\beta[B,A]}$. Solving equation (8) and keeping only terms linearly in β, the following expression for the operator $\sigma^{(1)}(t)$ can be obtained:

$$\sigma^{(1)}(t) = \mathrm{e}^{-\mathrm{i}\left(\alpha At-\alpha^2\frac{t^3}{6}\beta[[B,A],A]\right)}. \tag{9}$$

Again, equation (9) contains terms that are not linear in β. Continuing a similar expansion procedure, after the N steps we obtain

$$\mathrm{e}^{-\mathrm{i}t(\alpha A+\beta B)} = \left(1 - \beta\sum_{n=0}^{N}\alpha^n\frac{(\mathrm{i}t)^{n+1}}{(n+1)}\sum_{j=0}^{n}\frac{(-1)^j}{j!(n-j)!}A^j B A^{n-j}\right)\mathrm{e}^{-\mathrm{i}\left(\alpha tA+\frac{\beta}{\alpha}\frac{(\mathrm{i}t\alpha)^{N+2}}{(N+2)!}[B,A]_{N+1}\right)}, \tag{10}$$

where $[B, A]_{N+1}$ denotes the repeated commutators $[[[\ldots[[B, A], A]\ldots A]_{N+1}$. After summation over j in equation (10) we have

$$\mathrm{e}^{-\mathrm{i}t(\alpha A+\beta B)} = \left(1 - \frac{\beta}{\alpha}\sum_{n=0}^{N}\frac{(\mathrm{i}\alpha t)^{n+1}}{(n+1)!}\{B, A^n\}\right)\mathrm{e}^{-\mathrm{i}\left(\alpha tA+\frac{\beta}{\alpha}\frac{(\mathrm{i}t\alpha)^{N+2}}{(N+2)!}\{B,A^{N+2}\}\right)}, \tag{11}$$

where

$$\{B, A^0\} = B \quad \text{and} \quad \{B, A^{n+1}\} = [\{B, A^n\}, A]. \tag{12}$$

In the limit as the number of steps $N \to \infty$ we obtain that $\lim_{N=\infty}\left(\frac{\beta}{\alpha}\frac{(\mathrm{i}t\alpha)^{N+2}}{(N+2)!}\right) = 0$. Consequently, the exponent in (11) can be presented in the limit as $N \to \infty$ in the following form: $\lim_{N=\infty}\mathrm{e}^{-\mathrm{i}\left(\alpha tA+\frac{\beta}{\alpha}\frac{(\mathrm{i}t\alpha)^{N+2}}{(N+2)!}[B,A]_{N+1}\right)} = \mathrm{e}^{-\mathrm{i}\alpha tA}$, which does not include any terms with B, and the summing over n up to indefinite results in

$$\mathrm{e}^{-\mathrm{i}t(\alpha A+\beta B)} = \left(1 - \mathrm{i}\frac{\beta}{\alpha}\int_0^{\alpha t}\mathrm{d}x\,\mathrm{e}^{-\mathrm{i}xA}B\,\mathrm{e}^{\mathrm{i}xA}\right)\mathrm{e}^{-\mathrm{i}\alpha tA}, \tag{13}$$

which is a well-known formula for expansion of an exponential operator in a perturbation series [18]. To obtain expansion containing only linear to $\frac{\beta}{\alpha}$ terms we have to require that $\frac{\beta}{\alpha}\frac{(t\alpha)^{N+2}}{(N+2)!} \ll 1$. This requirement also imposes restrictions on time: $t \ll \frac{1}{\alpha}\left(\frac{\alpha}{\beta}(N+2)!\right)^{\frac{1}{N+2}}$. So for the smallest of times $t \ll \frac{1}{\alpha}\left(\frac{\alpha}{\beta}(N+2)!\right)^{\frac{1}{N+2}}$ or $t \ll \frac{1}{\beta}$, equation (10) includes only terms linear in β. In an analogous way, we obtain the expansion up to second order in the ratio $\frac{\beta}{\alpha}$:

$$\mathrm{e}^{-\mathrm{i}t(\alpha A+\beta B)} = \left\{1 - \frac{\beta}{\alpha}\sum_{m=0}^{\infty}\sum_{k=0}^{\infty}\left[\frac{(-1)^m(\mathrm{i}x)^{k+m+1}}{m!k!(k+m+1)}A^m B A^k \right.\right.$$
$$\left.\left. + \left(\frac{\beta}{\alpha}\right)^2 \sum_{l=0}^{\infty}\sum_{p=0}^{\infty}\frac{(-1)^{m+p}(\mathrm{i}x)^{k+l+m+p+2}}{m!k!l!p!(l+p+1)}\frac{A^{l+m}BA^pBA^k}{(k+l+m+p+2)}\right]\right\}\mathrm{e}^{-\mathrm{i}xA}. \qquad (14)$$

The series expansion (14) can be used not only for the small parameter $\frac{\beta}{\alpha} < 1$, but also independently, for the parameter $x = \alpha t$. Formula (14) can be easily generalized for a case where the exponential operator contains arbitrary number of the non-commutative operators and can be extended to include various power of the operators. Equation (14) appears to be complex at glance, but in fact it is quite simple to use, as the following examples will illustrate.

3. Results and Discussion

Let us consider a cluster of three dipolar-coupled spin-$\frac{1}{2}$ nuclei. The MQ dynamics in the rotating frame is described by propagator (1), where the time-independent average Hamiltonian is given by

$$\mathcal{H} = -\frac{1}{2}\sum_{j<k} d_{jk}\left(I_j^+ I_k^+ + I_j^- I_k^-\right) \qquad (15)$$

and I_j^+ and I_j^- are the raising and lowering operators for spin j respectively. The dipolar coupling constant, d_{jk}, for any pair of nuclei j and k in the cluster, is given by

$$d_{jk} = \frac{\gamma^2\hbar}{2r_{jk}^3}(1 - 3\cos\theta_{jk}), \qquad (16)$$

where γ is the gyromagnetic ratio of the nuclei, r_{jk} is the internuclear spacing and θ_{jk} is the angle the vector $\vec{r}_{jk}$ makes with the external magnetic field. In the high-temperature approximation, the density matrix at the end of the preparation period is given by

$$\rho(t) = \mathrm{e}^{-\mathrm{i}\mathcal{H}t}\rho(0)\,\mathrm{e}^{\mathrm{i}\mathcal{H}t}, \qquad (17)$$

where $\rho(0)$ is the initial density matrix in the high-temperature approximation

$$\rho(0) = \sum_{j=1}^{3} I_j^z, \qquad (18)$$

I_j^z is the projection of the angular momentum operator on the direction of the external field for a spin j. The average Hamiltonian (15) can be divided into three parts according to the number of the different coupling constants $d_{12} > d_{23} > d_{13}$:

$$H = H_{12} + H_{23} + H_{13}, \qquad (19)$$

where

$$H_{jk} = -\frac{d_{jk}}{2}\left(I_j^+ I_k^+ + I_j^- I_k^-\right) \qquad \text{with} \quad j \neq k \quad \text{and} \quad j,k = 1,2,3. \qquad (20)$$

The experimentally observed values are the intensities, $J_{nQ}(t)$ of multiple-quantum coherences:

$$J_{nQ}(t) = \frac{1}{\mathrm{Tr}\rho^2(0)}\sum_{p,q}\rho_{pq}^2(t) \qquad \text{for} \quad n = m_{zp} - m_{zq}, \qquad (21)$$

where m_{zp} and m_{zq} are the eigenvalues of the initial density matrix (18). The perturbation method described above is used to calculate the time evolution of MQ coherences. Using

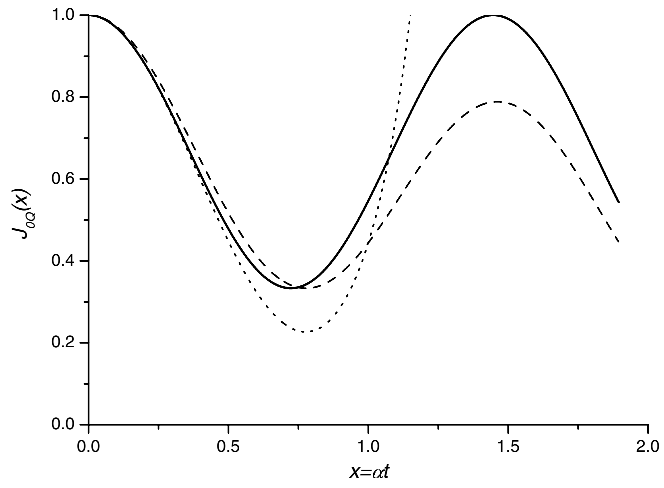

Figure 1. Time dependences (in units of $\frac{1}{\alpha}$) of the normalized intensities of 0Q coherence (solid-line is exact solution [5], dot-line is the calculation using equation (22) and dash-line is the calculation using equation (24)).

expansion (14) with $\alpha A = H_{12}$ and $\beta B = H_{23} + H_{13}$ and keeping terms up to eighth order in $x = \alpha t$, the normalized 0-quantum (J_{0Q}) and 2-quantum (J_{2Q}) intensities are given by

$$J_{0Q} = 1 - \frac{8x^2}{3} + \frac{32x^4}{9} - \frac{256x^6}{125} + \frac{512x^8}{945} + \left(\frac{\beta}{\alpha}\right)^2 \left(\frac{8x^2}{3} + \frac{40x^4}{9} - \frac{176x^6}{45} + \frac{1544x^8}{945}\right) \tag{22}$$

and

$$J_{2Q} = -\frac{4x^2}{3} + \frac{16x^4}{9} - \frac{128x^6}{135} + \frac{256x^8}{945} - \left(\frac{\beta}{\alpha}\right)^2 \left(\frac{4x^2}{3} + \frac{32x^4}{9} - \frac{128x^6}{45} + \frac{1024x^8}{945}\right), \tag{23}$$

where $\left(\frac{\beta}{\alpha}\right)^2 = \left(\frac{d_{23}}{d_{12}}\right)^2 + \left(\frac{d_{13}}{d_{12}}\right)^2$. Let us compare formulae (22) and (23) with results from equation (14) in which the terms with $x = \alpha t$ will be taken into account exactly. By summing over n, m, l and k up to infinity in (14), we obtain the analytical expressions of the intensities of 0-quantum

$$J_{0Q} = \frac{1}{3}\left\{\cos 4x - 2\left[\left(\frac{\beta}{\alpha}\right)^2 - 1\right] + 2\left(\frac{\beta}{\alpha}\right)^2 (\cos x + \cos 3x - \cos 4x - x \sin 4x)\right\} \tag{24}$$

and 2-quantum

$$J_{2Q} = -\frac{\sin 2x}{3}\left\{2x\left(\frac{\beta}{\alpha}\right)^2 \cos 2x + 2\left[\left(\frac{\beta}{\alpha}\right)^2 - \left[\left(\frac{\beta}{\alpha}\right)^2 - 1\right]\cos x\right]\sin x\right\}. \tag{25}$$

Now let us compare intensities (22)–(25) with the exact solution [5]. Figures 1 and 2 show the evolution of the normalized 0Q and 2Q coherences for three-spin cluster, where $\frac{\beta}{\alpha} = 0.3$ and at $t = 0$ the spin system is in thermal equilibrium (18). All approaches, perturbations (equations (22)–(25)) and exact [5], give closed agreement up to $x = 0.75$ (in unit of $\frac{1}{\alpha}$), both for 0Q- and 2Q-coherences. The exact account of influence of the nearest neighbours gives good agreements up to $x = 2$.

A second example we consider is a cluster consisting of four spins arranged in the corners of a square in an external magnetic field perpendicular to the square plane. In this case, the

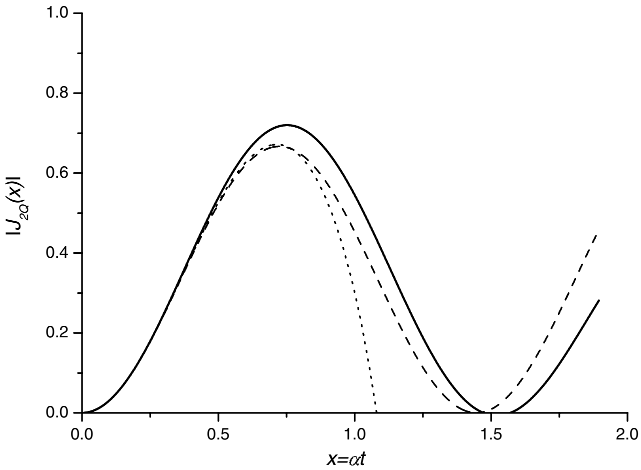

Figure 2. Time dependences (in units of $\frac{1}{\alpha}$) of the normalized intensities of 2Q coherence (solid-line is exact solution [5], dot-line is the calculation using equation (23) and dash-line is the calculation using equation (25)).

MQ spin dynamics is described by the average Hamiltonian

$$H = H_1 + H_2, \tag{26}$$

with two different dipolar coupling constants D_1 and D_2 , where D_1 and D_2 are the dipolar coupling constants between nearest neighbours and spins at opposite sites, respectively $\left(\frac{\beta}{\alpha} = \frac{D_2}{D_1} = \frac{1}{2\sqrt{2}}\right)$ where

$$H_1 = \left(-\frac{D_1}{2}\right)\sum_{j=1}^{4}\left(I_j^+ I_{j+1}^+ + I_j^- I_{j+1}^-\right) = \alpha A \tag{27}$$

and

$$H_2 = \left(-\frac{D_2}{2}\right)\sum_{j=1}^{2}\left(I_j^+ I_{j+2}^+ + I_j^- I_{j+2}^-\right) = \beta B. \tag{28}$$

Using the expansion (14) up to eighth order in $x = \alpha t$, the normalized 0-quantum (J_{0Q})

$$J_{0Q} = 1 - 2x^2 + \frac{7}{4}x^4 - \frac{13}{18}x^6 + \frac{5}{28}x^8 - \left(\frac{\beta}{\alpha}\right)^2\left(x^2 - \frac{13}{6}x^4 + \frac{121}{60}x^6 - \frac{599}{630}x^8\right) \tag{29}$$

and 2-quantum (J_{2Q})

$$J_{2Q} = -\frac{x^2}{4} + \frac{x^4}{4} - \frac{1}{9}x^6 + \frac{1}{35}x^8 - \left(\frac{\beta}{\alpha}\right)^2\left(\frac{x^2}{8} - \frac{x^4}{3} + \frac{11x^6}{30} - \frac{58x^8}{315}\right) \tag{30}$$

intensities can be determined. Formulae (29) and (30) will be compared with results from equation (14) in which the terms describing interaction of the neighbour spins will be taken into account exactly. By summation over n, m, l and k up to infinity in equation (14), we obtain the analytical expressions of the intensities of 0-quantum

$$J_{0Q} = \frac{1}{4}\left(1 + \sin^2 2x + 2\sin^2\sqrt{2}x\right) + \frac{x^2}{8}\left(\frac{\beta}{\alpha}\right)^2\left(2\sin^2 2x - \frac{\sqrt{2}}{x}\sin 2\sqrt{2}x\right) \tag{31}$$

and 2-quantum

$$J_{2Q} = -\frac{1}{8}\left(\sin^2 2x + 2\sin^2\sqrt{2}x\right) + \frac{x^2}{16}\left(\frac{\beta}{\alpha}\right)^2\left(2\sin^2 2x - \frac{\sqrt{2}}{x}\sin 2\sqrt{2}x\right) \tag{32}$$

coherences.

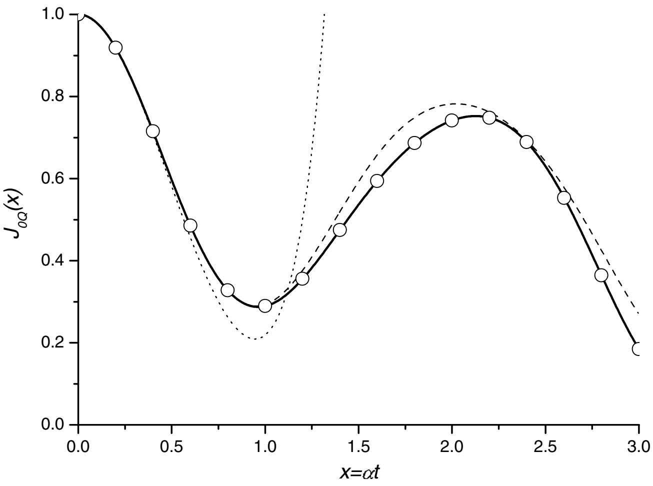

Figure 3. Time dependences (in units of $\frac{1}{\alpha}$) of the normalized intensities of 0Q coherence in four-spin cluster. Solid-line is exact solution (equation (33), dot-line is the calculation using equation (29), dash-line is the calculation using equation (31) and open circle is computer simulations.

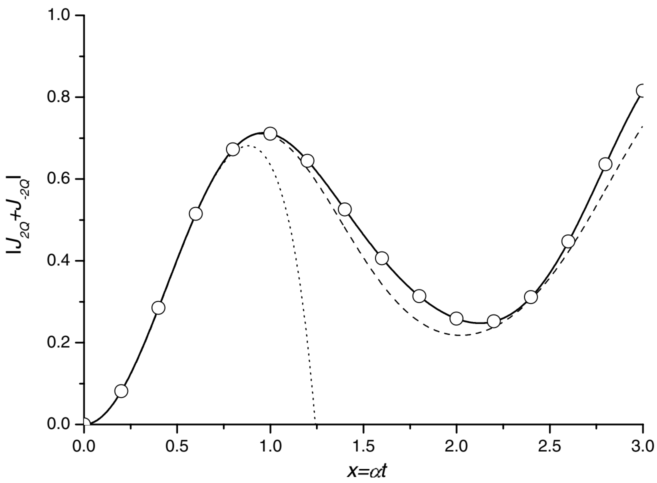

Figure 4. Time dependences (in units of $\frac{1}{\alpha}$) of the normalized intensities of 2Q coherence in four-spin cluster. Solid-line is exact solution (equation (34), dot-line is the calculation using equation (30), dash-line is the calculation using equation (32) and open circle is computer simulations.

To control the perturbation results (29)–(32), we obtained the exact solution for J_{0Q}

$$J_{0Q} = \frac{9}{4} - \frac{1}{2}\cos\left(2x\sqrt{2+\left(\frac{\beta}{\alpha}\right)^2}\right) + \frac{1}{4}\cos 4x \cos\left(2x\frac{\beta}{\alpha}\right) - \cos^2\left(x\frac{\beta}{\alpha}\right) \tag{33}$$

and for J_{2Q}

$$J_{2Q} = -\frac{3}{4} + \frac{1}{4}\cos\left(2x\sqrt{2+\left(\frac{\beta}{\alpha}\right)^2}\right) + \frac{1}{4}\sin^2 2x \cos\left(2x\frac{\beta}{\alpha}\right) + \frac{1}{2}\cos^2\left(x\frac{\beta}{\alpha}\right) \tag{34}$$

coherences and fulfilled the numerical analysis of the MQ dynamics. The exact solutions (33) and (34) and computer simulation of the MQ coherences of four-spin cluster have been obtained with a PC using the MATLAB package.

Figures 3 and 4 show that perturbation results (29) and (30) are in agreement with (31) and (32) and the exact solutions (33) and (34) and with the simulation data one up to $x = 1$ (in unit of $\frac{1}{\alpha}$). Calculations in which the interaction between the nearest neighbours is taken into account exactly (equations (31) and (32)) are in close agreement with the exact solutions (equations (33) and (34)) and simulation data up to $x = 3$.

4. Conclusion

In conclusion, a perturbation method was developed which is based on the expansion of operator exponent in a perturbation series. Then the perturbation approach was applied to the description of the MQ spin dynamics in solids. The analytical expressions for 0Q and 2Q dynamics in three- and four-spin clusters in solids were obtained. In the four-spin cluster the exact solution was obtained. The calculated 0Q and 2Q intensities versus the duration of the preparation period agree well with exact solutions for three- [5] and four-spin clusters (equations (33) and (34)).

The developed method can be extended to include various power of the operators with small norm and applied to the description of a wide range of physical problems that deal with dynamics of dipolar coupling spins in solids. The results in an analytical form can be used to extract from experimental data the dipolar constants and the molecular structure information.

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