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Investigation of the Effect of Finite Pulse Errors on BABA Pulse Sequence Using Floquet-Magnus Expansion Approach

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Abstract

This paper presents the study of finite pulse widths for the BABA pulse sequence using the Floquet-Magnus expansion (FME) approach. In the FME scheme, the first order F_1 is identical to its counterparts in average Hamiltonian theory (AHT) and Floquet theory (FT). However, the timing part in the FME approach is introduced via the $A_1(t)$ function not present in other schemes. This function provides an easy way for evaluating the spin evolution during “the time in between” through the Magnus expansion of the operator connected to the timing part of the evolution. The evaluation of $A_1(t)$ is useful especially for the analysis of the non-stroboscopic evolution. Here, the importance of the boundary conditions, which provides a natural choice of $A_1(0)$ is ignored. This work uses the $A_1(t)$ function to compare the efficiency of the BABA pulse sequence with δ -pulses and the BABA pulse sequence with finite pulses. Calculations of $A_1(t)$ and F_1 are presented.

Keywords

BABA Pulse Sequence; Finite Pulse Widths; Dipolar Coupling; Floquet-Magnus Expansion

I. INTRODUCTION

Average Hamiltonian theory (AHT) [1, 2] and Floquet theory (FT) [3] are the commonly used methods to treat theoretical problems in solid-state nuclear magnetic resonance (NMR), respectively in static and rotating samples. In recent years, there has been a sustained effort using the AHT to improve the line-narrowing efficiency of a multiple-pulse cycle by constructing supercycles to compensate for any pulse errors and by designing symmetries into the toggling frame Hamiltonian [4]. For example, the celebrated WAHUA cycle as well as many other multiple-pulse sequences were significantly improved when finite pulse widths were properly accounted for [5]. About three decades ago, Vega [6, 8, 9] and Maricq

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[7] introduced to NMR, the FT which provides a more universal approach for the description of the full time system. The FT also allows the computation of the full spinning sideband pattern that is of importance in many magic angle spinning (MAS) experimental circumstances to obtain information on anisotropic sample properties.

Recently, the Floquet Magnus Expansion (FME) approach was introduced to solid-state NMR spectroscopy [10, 11, 12, 13]. The method of FME is a viable scheme for controlling the complex spin dynamics for an ensemble of dipolar coupled spins. The approach was compared to other series expansions such as AHT, FT, and a generalized FME scheme was presented based on the importance of the boundary conditions, which provides a natural choice of $A_n(0)$ to simplify calculation of higher order terms and allows FT to be managed in the Hilbert space. In the recent work on the efficient theory of dipolar recoupling in solid state NMR of rotating solids [14], we applied the FME approach on BABA [15] and C7 radiofrequency pulse sequences [16]. The application of the method on BABA sequences cases ignored the spin system's evolution during the RF pulse sequences and approximated the RF pulses as ideal δ -function perturbations.

Pulses in NMR spectrometers have a finite length, but the usual hard pulse assumption ignores finite pulse width and treats the pulse as a rotation of the frame of reference about the direction of the radiofrequency (RF) magnetic field [17]. This assumption implies that the interaction with the RF magnetic field dominates all other terms in the Hamiltonian. For liquids, this approximation works well, but for dipolar nuclei (spin-1/2), the RF magnetic field is commonly compared to the offset of the signal from resonance. Modern high-field spectrometers permit offsets of the dipolar interactions. The situation with quadrupolar nuclei is much worse, with resonances hundreds of kHz off-resonance. Therefore, accounting for finite pulse width is important due to the evolution of the spin system under the dipolar or quadrupolar interaction [4, 18, 19, 20, 21, 22]. Hence, a good understanding of the spin system behavior during the finite pulses and theory therein is important.

Previous work associated with effects due to finite pulse widths have been reported by Bloom et al. [23], Henrichs et al. [24], Mananga et al. [18], and Gregory et al. [25]. The first three works showed that one can remove distortions due to finite pulse widths for example in deuterium (spin-1) nuclear magnetic resonance spectroscopy respectively by scaling the experimental data by a multiplicative factor determined a priori, and by cycling the phases of both the receiver and the transmitter. Gregory et al. reported substantial distortions in the phase of the signal for a spin-1/2, at frequency offsets comparable to the size of the RF field. On the other hand, Sergeev [26] reported the effects of finite pulse width on free induction decay using Liouville superoperators. The effects of the finite pulse width on the shape of the FID were also discussed by Barnaal et al. [27, 28], and Henrichs et al. [24].

Jaroniec did work for REDOR [40, 42] that involved use of the average Hamiltonian theory in the straightforward manner in obtaining the magnitude of the double quantum Hamiltonian as a function of the pulse length relative to the rotor period. The initial density operator and the spin evolution under an average Hamiltonian result in the observable signal. This development does not consider the behavior of the spin system during the finite pulse width which cannot be described by the AHT. In the theory of AHT, the spin system is

considered to be evolving under an average Hamiltonian. Unfortunately, the spin system evolves in a more complex way as shown in this article. Therefore, the FME approach that offers the possibility to study the spin dynamics at all times including the multiple period times ($t = nT$) as described by the AHT can also be used as a viable approach in control theory of spin systems. Only under a particular case [10] that the Floquet Magnus Expansion gives the AHT result as provided by the Magnus expansion. This new approach of FME provides new opportunities to control the evolution of the spin systems in between the stroboscopic detection points. More recently, Saalwachter [41] advocate the use of normalized double quantum (DQ) build-up curves for a quantitative assessment of weak average dipole-dipole couplings and even distributions thereof.

In this work, we use the FME approach to investigate the effects of finite pulse width on the BABA pulse sequence. The interaction Hamiltonian used in this study is the dipolar Hamiltonian. In practice, the performance of rotor-synchronized pulse sequences is sensitive to chemical-shielding interaction and to resonance offset, making use of nuclei such as ^{13}C , ^{15}N , and ^{31}P impractical in high magnetic fields, where the isotropic and anisotropic part of the chemical shielding commonly dominates over the dipolar interaction and often exceeds the spinning frequency. In the present article, we ignore effects of the off-resonance, chemical shift anisotropy and J couplings. We only consider and use the dipole-dipole coupling to generate double quantum coherence. To the best of our knowledge, this is the first report highlighting the application of the FME scheme to investigate the effect of finite pulse widths on recoupling pulse sequences such as BABA. We have limited our effort to the first order term. In our recent work [14], we showed that $\mathcal{A}_2(t) \ll \mathcal{A}_1(t)$. This work shows how complex equations describing the complexity of the spins dynamics can be reduced to simple equations.

An outline of the paper is as follows. In Sec. II, we describe the Floquet-Magnus expansion with a brief illustration of the Magnus expansion (ME) and the celebrated Floquet theorem, which ensures the factorization of the solution in a periodic part and a purely exponential factor. We elucidate explicitly, the first contributions to the Floquet-Magnus expansion. Sec. III, is the discussion of the results obtained. We briefly discuss the results and we compare them to those found in previous work and other authors [14, 15] in an analogue treatment. Sec. IV of the paper summarizes our conclusions and the final sections V, VI and VII are reserved respectively to the acknowledgments, appendix and references.

II. THEORY

In the following, we analyze the spin dynamics of a system of dipolar coupled spins evolving under the effect of finite pulse widths of BABA pulse sequence shown in Fig. 1. Ignoring any J-coupling, quadrupolar coupling and chemical shift, the nuclear spin Hamiltonian is given by the familiar dipolar interaction, which is written as

$$H_D(t) = \frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) T_{20}^{ij} \quad (1)$$

where the second-rank irreducible tensor operators is

$$T_{20}^{ij} = \frac{1}{\sqrt{6}} [2I_{ZZ}^{ij} - I_{XX}^{ij} - I_{YY}^{ij}]. \quad (2)$$

In eq. (1), we have

$$\omega_D^{ij}(t) = b_{ij} \sum_{n=-2}^2 C_n^{ij}(\alpha^{ij}, \beta^{ij}, \gamma^{ij}) e^{-in\omega_R t} = b_{ij} \sum_{n=-2}^2 C_n^{ij}(\alpha^{ij}, \beta^{ij}) e^{-in(\omega_R t + \gamma^{ij})} \quad (3)$$

where α , β and γ are Euler angles describing the orientation of a given molecule or crystallite in the MAS rotor. b_{ij} is the coupling constant [14, 28]. The coefficients C_n^{ij} can be expressed as

$$C_n^{ij}(\alpha^{ij}, \beta^{ij}) = d_{0,n}^2(\theta_M) \sum_{n'=-2}^2 (-1)^{n'} Y_{2n'}^{ij} e^{-in\alpha^{ij}} d_{n-n'}^2(\beta^{ij}) \quad (4)$$

where Y_{2n}^{ij} are second-rank spherical harmonic functions. $d_{nm}^2(\beta^{ij})$ are second-rank reduced Wigner rotation matrix elements and θ_M is the magic angle, with $d_{00}^2(\theta_M) = 0$ and $C_0^{ij} = 0$ [29].

We consider the evolution of the spin system under the BABA pulse sequence with finite pulse width errors shown in Fig. 1, developed by Demco et al. [15]. This sequence resembles the pulse sequence proposed by Meier and Earl [30, 31] which is not completely synchronized for producing a pure double-quantum Hamiltonian. The BABA cycle with ideal δ -pulses is already well known in the NMR community to give more efficient recoupling compared to many recoupling sequences such as DRAMA, HORROR or C7 pulse sequences [16]. The BABA strength for generating a pure double-quantum Hamiltonian is twice that of the DRAMA and C7 pulse sequences, and has been used for broadband high-resolution multiple-quantum NMR spectroscopy in rotating dipolar solids. The sequence with finite pulse width errors consists of a 90° pulse about the x axis, a period of free evolution of time $(\frac{\tau_R}{2} - 2\tau_P)$, a 90° pulse about the (-x) axis, followed by a 90° pulse about the (y) axis, next a period of free evolution of time $(\frac{\tau_R}{2} - 2\tau_P)$, ending with the application of a 90° pulse about the (-y) axis. All pulse widths are of duration (τ_P) . The BABA pulse sequence is used during the excitation period to generate a pure double-quantum Hamiltonian. It has been proven that a further improvement of the signal-to-noise ratio is achieved by sign inversion of all pulse phases after two rotor periods for the compensation of radiofrequency pulse imperfections [15]. Different variants of the BABA sequence are possible for the excitation of double quantum coherences [30, 31, 33, 34, 35], and can also be the subject of a similar treatment used in this article.

Consider the following representation for free evolution time in Fig. 1

The toggling frame dipolar Hamiltonian, $H_D^{\sim}$, during each stage of the pulse cycle can be computed. During the first pulse about (x), the toggling Hamiltonian is writing as

$$\tilde{H}_D^X(t) = \frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) \tilde{T}_{20}^{ijX}(t). \quad (5)$$

We have,

$$T_{20}^{ijX}(t) = e^{-i\omega_{RF} I_X t} T_{20}^{ij} e^{i\omega_{RF} I_X t} = e^{-i\omega_{RF} I_X t} \frac{1}{\sqrt{6}} [2I_{ZZ}^{ij} - I_{XX}^{ij} - I_{YY}^{ij}] e^{i\omega_{RF} I_X t}. \quad (6)$$

Using the following sandwich formula [32]

$$e^{-i\theta A} B e^{i\theta A} = B \cos \theta + C \sin \theta \quad (7)$$

(A, B, and C are three operators that cyclically commute), the dipolar operator during the first interval: $0 \leq t < \tau_P$, can be calculated as following

$$\tilde{T}_{20}^{ijX}(t) = e^{-i\alpha I_X} \frac{1}{\sqrt{6}} [2I_{ZZ}^{ij} - I_{XX}^{ij} - I_{YY}^{ij}] e^{i\alpha I_X} = \frac{1}{\sqrt{6}} [2e^{-i\alpha I_X} I_{ZZ}^{ij} e^{i\alpha I_X} - e^{-i\alpha I_X} I_{XX}^{ij} e^{i\alpha I_X} - e^{-i\alpha I_X} I_{YY}^{ij} e^{i\alpha I_X}] \quad (8)$$

which gives

$$\tilde{T}_{20}^{ijX}(t) = \frac{1}{\sqrt{6}} [(3\cos^2 \alpha - 1) I_{ZZ}^{ij} + (3\sin^2 \alpha - 1) I_{YY}^{ij} - I_{XX}^{ij} - 3\cos \alpha \sin \alpha (I_{ZY}^{ij} + I_{YZ}^{ij})]. \quad (9)$$

with,

$$\alpha = \omega_{RF} t, \quad (10)$$

For $\alpha = \frac{\pi}{2}$,

$$\tilde{T}_{20}^{ijX}\left(\alpha = \frac{\pi}{2}\right) = \frac{1}{\sqrt{6}} [2I_{YY}^{ij} - I_{ZZ}^{ij} - I_{XX}^{ij}] = H_{YY}^{ij} \quad (11)$$

During the second time interval: $\tau_P \leq t < \frac{\tau_R}{2} - \tau_P$, we have

$$H_{\alpha\alpha}(t) = \frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) H_{\alpha\alpha}^{ij} \quad (12)$$

with $\alpha = x, y$, and z .

Considering the picture of Fig. 2, the time-dependent function $\theta(t)$ can be expressed in the form of the Fourier expansion

$$\theta(t) = \sum_{k=-\infty}^{+\infty} a_k \exp(-ik\omega_R t) \quad (13)$$

with a_k representing the time-independent Fourier coefficients corresponding to the Fourier index k . The coefficient a_k can be obtained

$$a_k = \frac{1}{\tau_R} \int_0^{\tau_R} \theta(t) e^{ik\omega_R t} dt \quad (14)$$

The coefficients in the first half of the sequence are

$$a_k^X = \frac{1}{\tau_R} \int_{\tau_P}^{\tau_R/2 - \tau_P} e^{ik\omega_R t} dt \quad (15)$$

which are given explicitly by

$$a_o^X = \frac{1}{2} - \varphi, \quad (16)$$

$$a_k^X = \frac{1}{2\pi ik} e^{ik\pi\varphi} [e^{ik\pi(1-2\varphi)} - 1] \quad (17)$$

with

$$\varphi = \frac{2\tau_P}{\tau_R}. \quad (18)$$

In the second half of the sequence, we have

$$a_k^Y = \frac{1}{\tau_R} \int_{\frac{\tau_R}{2} + \tau_P}^{\tau_R - \tau_P} e^{ik\omega_R t} dt \quad (19)$$

which can also be written explicitly as

$$a_o^Y = \frac{1}{2} - \varphi, \quad (20)$$

$$a_k^Y = \frac{1}{2\pi ik} e^{ik\pi(1+\varphi)} [e^{ik\pi(1-2\varphi)} - 1] \quad (21)$$

Globally, we can summarize the following

$$a_o^X = a_o^Y = \frac{1}{2} - \varphi = a_0, \quad (22)$$

$$a_k^Y = e^{ik\pi} a_k^X = a_k \quad (23)$$

For the δ -pulse corresponding to $\phi = 0$, we retrieve the identical results found in [14]. Table 1 gives the toggling frame Hamiltonians during each stage of the system evolution that was developed knowing the transformations of I_x , I_y and I_z .

The FME is an illuminating approach that can be used to provide a more intuitive understanding of processes in spin dynamics. This approach is essentially distinguished from AHT with its function $\Lambda_n(t)$ which provides an easy and alternative way for evaluating the spin behavior in between the stroboscopic observation points. Following the procedure described in reference [10], the general formula of the approach is given by:

$$\Lambda_n(t) = \int_0^t G_n^t(\tau) d\tau - tF_n + \Lambda_n(0) \quad (24)$$

$$F_n = \frac{1}{T} \int_0^T G_n^T(\tau) d\tau \quad (25)$$

where $G_n(t)$ functions are constructed using the FME recursive generation scheme [10, 11, 12, 13]. The first order explicit formula gives

$$\Lambda_1(t) = \Lambda_1(0) + \int_0^t G_1^t(\tau) d\tau - tF_1 \quad (26)$$

with

$$G_1(\tau) = H(\tau). \quad (27)$$

$$\Lambda_1(T) = \Lambda_1(0) \quad (28)$$

and

$$F_1 = \frac{1}{T} \int_0^T H^T(\tau) d\tau \quad (29)$$

In the above expressions, F_n represents the time independent Hamiltonian that governs the evolution of the propagator $U(t)$ expressed as

$$U(t) = e^{-i \sum_{n=1}^{\infty} \Lambda_n(t)} e^{-it \sum_{n=1}^{\infty} F_n} \quad (30)$$

Via the $\Lambda_n(t)$ function, the FME approach is used to provide insight into how the finite pulse widths affect the dipolar spin system evolution. We have limited our efforts to the first

order contribution. The FME method will show how the BABA pulse sequence, with the finite pulse widths taken into consideration, are less robust in generating a pure double-quantum Hamiltonian compared to the BABA sequence when removing the contribution of finite pulse widths i.e. considering the δ -function RF pulses.

The resulting first order term of the dipolar Hamiltonian in the FME is calculated as following:

$$F_1 = \frac{1}{T} \int_0^T \tilde{H}_D(\tau) d\tau = \frac{1}{T} \left[\int_0^{\tau_P} \tilde{H}_D(\tau) d\tau + \int_{\tau_P}^{\tau_R/2 - \tau_P} \tilde{H}_D(\tau) d\tau + \int_{\tau_R/2 - \tau_P}^{\tau_R/2} \tilde{H}_D(\tau) d\tau \right. \\ \left. + \int_{\tau_R/2}^{\tau_R/2 + \tau_P} \tilde{H}_D(\tau) d\tau + \int_{\tau_R/2 + \tau_P}^{\tau_R - \tau_P} \tilde{H}_D(\tau) d\tau + \int_{\tau_R - \tau_P}^{\tau_R} \tilde{H}_D(\tau) d\tau \right] \quad (31)$$

Note that the sum

$$\frac{1}{T} \int_{\tau_P}^{\tau_R/2 - \tau_P} \tilde{H}_D(\tau) d\tau + \frac{1}{T} \int_{\tau_R/2 + \tau_P}^{\tau_R - \tau_P} \tilde{H}_D(\tau) d\tau \quad (32)$$

corresponds to the average Hamiltonian of BABA with δ -function RF pulses [14].

The resulting toggling frame Hamiltonians were integrated over their respective time intervals and are provided as follows

$$\int_0^{\tau_P} \tilde{H}_D(t) dt = \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^2 C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3(2\omega_{RF} \sin(2\tau_P \omega_{RF})) - 3(\omega_R \sin \cos(2\tau_P \omega_{RF}))}{2e^{(\tau_P \omega_R \sin)} (4\omega_{RF}^2 - \omega_R^2 n^2)} + \frac{1 - e^{-(\tau_P \omega_R \sin)}}{2\omega_R \sin} \right] I_{ZZ} \\ + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^2 C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3\omega_R \sin}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} I_{ZZ} + \right. \\ \left. \left(\frac{-3(2\omega_{RF} \sin(2\tau_P \omega_{RF})) - \omega_R \sin \cos(2\tau_P \omega_{RF}))}{2e^{(\tau_P \omega_R \sin)} (4\omega_{RF}^2 - \omega_R^2 n^2)} \right) I_{YY} \right] \\ + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^2 C_n^{ij} e^{-in\gamma^{ij}} \left[\left(\frac{1 - e^{-(\tau_P \omega_R \sin)}}{2\omega_R \sin} - \frac{3\omega_R \sin}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \right) I_{YY} + \right. \\ \left. \frac{e^{-(\tau_P \omega_R \sin)} - 1}{\omega_R \sin} I_{XX} \right] + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^2 C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{-3\omega_{RF}}{(4\omega_{RF}^2 - \omega_R^2 n^2)} + \right. \\ \left. \frac{-3(2\omega_{RF} \cos(2\tau_P \omega_{RF})) + \omega_R \sin \sin(2\tau_P \omega_{RF}))}{2e^{(\tau_P \omega_R \sin)} (4\omega_{RF}^2 - \omega_R^2 n^2)} \right] (I_{ZY} + I_{YZ}) \quad (33)$$

$$\int_{\tau_P}^{\tau_R - \tau_P} \tilde{H}_D(t) dt = \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{\tau_R}{2} - 2\tau_P \right) (2I_{YY}^{ij} - I_{XX}^{ij} - I_{ZZ}^{ij}) \quad (34)$$

$$\int_{\tau_R/2 - \tau_P}^{\tau_R/2} \tilde{H}_D(t) dt = \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\tau_P}{2} - \frac{3}{4\omega_{RF}} (\sin(\omega_{RF} \tau_R) + \sin(2\omega_{RF} (\tau_P - \frac{\tau_R}{2}))) \right] I_{ZZ}^{ij} \right\} \\ + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\tau_P}{2} + \frac{3}{4\omega_{RF}} (\sin(\omega_{RF} \tau_R) + \sin(2\omega_{RF} (\tau_P - \frac{\tau_R}{2}))) \right] I_{YY}^{ij} - \tau_P I_{XX}^{ij} \right\} \\ + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{3}{4\omega_{RF}} (\cos(\omega_{RF} \tau_R) - \cos(2\omega_{RF} (\tau_P - \frac{\tau_R}{2}))) \right] (I_{ZY}^{ij} + I_{YZ}^{ij}) \right\} \quad (35)$$

$$\begin{aligned}
& \int_{\frac{\tau_R}{2}}^{\frac{\tau_R}{2}+\tau_P} \tilde{H}_D(t) dt = \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\tau_P}{2} - \frac{3}{4\omega_{RF}} (\sin(\omega_{RF}\tau_R) - \sin(2\omega_{RF}(\tau_P + \frac{\tau_R}{2}))) \right] I_{ZZ}^{ij} \right\} \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\tau_P}{2} + \frac{3}{4\omega_{RF}} (\sin(\omega_{RF}\tau_R) - \sin(2\omega_{RF}(\tau_P + \frac{\tau_R}{2}))) \right] I_{XX}^{ij} - \tau_P I_{YY}^{ij} \right\} \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{3}{4\omega_{RF}} (\cos(\omega_{RF}\tau_R) - \cos(2\omega_{RF}(\tau_P + \frac{\tau_R}{2}))) \right] (I_{XZ}^{ij} + I_{ZX}^{ij}) \right\} \quad (36) \\
& \int_{\frac{\tau_R}{2}+\tau_P}^{\tau_R-\tau_P} \tilde{H}_D(t) dt = \frac{-1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{\tau_R}{2} - 2\tau_P \right) (2I_{XX}^{ij} - I_{YY}^{ij} - I_{ZZ}^{ij}) \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^2 C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{-1}{in\omega_R} \right) (e^{-in\omega_R\tau_R(1-\frac{\tau_P}{\tau_R})} - e^{in\omega_R\tau_R(\frac{1}{2}+\frac{\tau_P}{\tau_R})}) (2I_{XX}^{ij} - I_{YY}^{ij} - I_{ZZ}^{ij})
\end{aligned}$$

$$\begin{aligned}
& \int_{\tau_R-\tau_P}^{\tau_R} \tilde{H}_D(t) dt = \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} [I_1 I_{XX}^{ij} + I_2 I_{ZZ}^{ij} - I_3 I_{YY}^{ij} + 3I_4 (I_{XZ}^{ij} + I_{ZX}^{ij})] \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\tau_P}{2} + \frac{3}{4\omega_{RF}} (\sin(2\omega_{RF}\tau_R) + \sin(2\omega_{RF}(\tau_P - \tau_R))) \right] I_{XX}^{ij} \right\} \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\tau_P}{2} + \frac{3}{4\omega_{RF}} (\sin(2\omega_{RF}\tau_R) + \sin(2\omega_{RF}(\tau_P - \tau_R))) \right] I_{ZZ}^{ij} - \tau_P I_{YY}^{ij} \right\} \quad (37) \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{-3}{4\omega_{RF}} (\cos(2\omega_{RF}\tau_R) - \cos(2\omega_{RF}(\tau_P - \tau_R))) \right] (I_{XZ}^{ij} + I_{ZX}^{ij}) \right\}
\end{aligned}$$

where the integrals I_1 , I_2 , I_3 and I_4 are given by

$$\begin{aligned}
I_1 = & \int_{\tau_R-\tau_P}^{\tau_R} e^{-in\omega_R t} (3\cos^2(\omega_{RF}t) - 1) dt = \frac{3(2\omega_{RF}\sin(2\tau_R\omega_{RF})) - 3(\omega_R in\cos(2\tau_R\omega_{RF}))}{2e^{(\tau_R\omega_R in)}(4\omega_{RF}^2 - \omega_R^2 n^2)} \\
& - (e^{in\omega_R\tau_R}) \frac{(1 - e^{(\tau_P\omega_R in)})}{2\omega_R in} - \frac{3(2\omega_{RF}\sin(2\omega_{RF}(\tau_R - \tau_P))) - 3(\omega_R in\cos(2\omega_{RF}(\tau_R - \tau_P)))}{2e^{in\omega_R(\tau_R - \tau_P)}(4\omega_{RF}^2 - \omega_R^2 n^2)} \quad (38)
\end{aligned}$$

$$\begin{aligned}
I_2 = & \int_{\tau_R-\tau_P}^{\tau_R} e^{-in\omega_R t} (3\sin^2(\omega_{RF}t) - 1) dt = \frac{3(2\omega_{RF}\sin(2\omega_{RF}(\tau_R - \tau_P))) - 3(\omega_R in\cos(2\omega_{RF}(\tau_R - \tau_P)))}{2e^{\omega_R in(\tau_R - \tau_P)}(4\omega_{RF}^2 - \omega_R^2 n^2)} \\
& - (e^{in\omega_R\tau_R}) \frac{(1 - e^{(\tau_P\omega_R in)})}{2\omega_R in} - \frac{3(2\omega_{RF}\sin(2\omega_{RF}\tau_R)) - 3(\omega_R in\cos(2\omega_{RF}\tau_R))}{2e^{in\omega_R\tau_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \quad (39)
\end{aligned}$$

$$I_3 = \int_{\tau_R-\tau_P}^{\tau_R} e^{-in\omega_R t} dt = - (e^{-in\omega_R\tau_R}) \frac{(1 - e^{(\tau_P\omega_R in)})}{\omega_R in} \quad (40)$$

$$\begin{aligned}
I_4 = & \int_{\tau_R-\tau_P}^{\tau_R} e^{-in\omega_R t} \cos(\omega_{RF}t) \sin(\omega_{RF}t) dt = \frac{2\omega_{RF}\cos(2\omega_{RF}(\tau_R - \tau_P)) + \omega_R in\sin(2\omega_{RF}(\tau_R - \tau_P))}{2e^{\omega_R in(\tau_R - \tau_P)}(4\omega_{RF}^2 - \omega_R^2 n^2)} \\
& - \frac{2\omega_{RF}\sin(2\omega_{RF}\tau_R) + \omega_R in\sin(2\omega_{RF}\tau_R)}{2e^{in\omega_R\tau_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \quad (41)
\end{aligned}$$

From the above results, the first-order term of the FME F_1 is obtained

(42)

Two important findings of this result should be recognized. First, when $\tau_P = 0$, the above first-order expression F_1 of the FME is reduced to the following build-up DQ coherence expression

$$F_1 = \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} \right) (2I_{YY}^{ij} - I_{XX}^{ij} - I_{ZZ}^{ij}) + \\ - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} \right) (2I_{XX}^{ij} - I_{ZZ}^{ij} - I_{YY}^{ij}) + \\ - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{1}{in\omega_R\tau_R} \right) (e^{-in\omega_R\tau_R} - e^{-i\frac{n}{2}\omega_R\tau_R}) (2I_{XX}^{ij} - I_{ZZ}^{ij} - I_{YY}^{ij}) \quad (43)$$

The last double summation in the above F_1 expression is shown to be equal to zero (see Appendix). Therefore, the above first order expression F_1 is reduced to the following form

$$F_1 = \overline{H}^0 = \frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} \right) (I_{YY}^{ij} - I_{XX}^{ij}) \quad (44)$$

This is what one would expect in the case of δ -function RF pulses corresponding to $\tau_P = 0$ [14, 15]. Second, the result indicates that the contribution of finite pulse widths to the system evolution becomes less important for large values of τ_R . In practice, the timing of the BABA pulse sequence is known from broadband high-resolution multiple-quantum NMR spectroscopy to be essential for full synchronization in rotating dipolar solids [15]. This is important for generating a pure double-quantum Hamiltonian or for producing the maximum strength of the DQ Hamiltonian.

The expression of F_1 that describes not only build-up but also destruction of DQ coherence can also be obtained as

$$\begin{aligned}
F_1 = & -\frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3}{4\pi e^{(in\pi\varnothing)}} \left(\frac{\omega_R^2 in}{4\omega_{RF}^2 - \omega_R^2 n^2} \right) + \frac{e^{(-in\varnothing\pi)} - 1}{4\pi in} \right] I_{YY}^{ij} + \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{-3}{4\pi} \left(\frac{\omega_R^2 in}{4\omega_{RF}^2 - \omega_R^2 n^2} \right) I_{YY}^{ij} + \left(\frac{e^{(-in\varnothing\pi)} - 1}{2\pi in} \right) I_{XX}^{ij} \right] + \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} - \varnothing \right) (2I_{YY}^{ij} - I_{XX}^{ij}) + \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\varnothing}{4} + \frac{3\varnothing}{4\pi} \left(\sin \left(\frac{\pi}{\varnothing} \right) + \sin \left(\pi \left(1 - \frac{1}{\varnothing} \right) \right) \right) \right] I_{YY}^{ij} - \frac{\varnothing}{2} I_{XX}^{ij} \right\} + \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left\{ \left[\frac{\varnothing}{4} + \frac{3\varnothing}{4\pi} \left(\sin \left(\frac{\pi}{\varnothing} \right) - \sin \left(\pi \left(1 + \frac{1}{\varnothing} \right) \right) \right) \right] I_{XX}^{ij} - \frac{\varnothing}{2} I_{YY}^{ij} \right\} + \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{-1}{in2\pi} \right) [e^{-in2\pi(1-\frac{\varnothing}{2})} - e^{-in2\pi(\frac{1}{2}+\frac{\varnothing}{2})}] (2I_{XX}^{ij} - I_{YY}^{ij}) \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} - \varnothing \right) (2I_{XX}^{ij} - I_{YY}^{ij}) + \\
& + \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{-1}{in2\pi} \right) [e^{-in2\pi(1-\frac{\varnothing}{2})} - e^{-in2\pi(\frac{1}{2}+\frac{\varnothing}{2})}] \left[\frac{\varnothing}{4} + \frac{3\varnothing}{4\pi} \sin \left(\frac{2\pi}{\varnothing} \right) \right] I_{XX}^{ij} + \\
& + \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{-1}{in2\pi} \right) [e^{-in2\pi(1-\frac{\varnothing}{2})} - e^{-in2\pi(\frac{1}{2}+\frac{\varnothing}{2})}] \left[\frac{3\varnothing}{4\pi} \sin \pi \left(1 - \frac{2}{\varnothing} \right) \right] I_{XX}^{ij} + \\
& + \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{-1}{in2\pi} \right) [e^{-in2\pi(1-\frac{\varnothing}{2})} - e^{-in2\pi(\frac{1}{2}+\frac{\varnothing}{2})}] \left[\frac{-\varnothing}{2} \right] I_{YY}^{ij} + \\
& - \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} - \varnothing \right) \left[\frac{\varnothing}{4} + \frac{3\varnothing}{4\pi} \left(\sin \left(\frac{2\pi}{\varnothing} \right) + \sin \pi \left(1 - \frac{2}{\varnothing} \right) \right) \right] I_{XX}^{ij} + \\
& + \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} - \varnothing \right) \frac{\varnothing}{2} I_{YY}^{ij}
\end{aligned} \tag{45}$$

with

$$\omega_{RF} \tau_P = \frac{\pi}{2}; \tag{46}$$

$$\omega_R = \frac{2\pi}{\tau_R}; \tag{47}$$

$$\varnothing = \frac{2\tau_P}{\tau_R}. \tag{48}$$

Considering only the expressions with double-quantum terms F_1 is reduced to the following terms:

$$F_1 = \overline{H}^0 = \frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{1}{2} - \varnothing \right) (I_{YY}^{ij} - I_{XX}^{ij}) \tag{49}$$

which corresponds to the above result (Eq. (44)) for $\varnothing = 0$.

Next, the first order of eq. (24) can be computed as follows

$$\Lambda_1(t) = \Lambda_1(0) + \int_0^t G_1(\tau) d\tau - tF_1 \quad (26)$$

with

$$G_1(\tau) = H(\tau). \quad (27)$$

$$\Lambda_1(T) = \Lambda_1(0) = 0 \quad (50)$$

$$\Lambda_1(t) = \int_0^t \tilde{H}_D^X(t') dt' + \int_0^t H_{YY}(t') \theta_Y(t') dt' + \int_0^t \tilde{H}_D^{\bar{X}}(t') dt' + \int_0^t \tilde{H}_D^Y(t') dt' + \int_0^t H_{XX}(t') \theta_X(t') dt' + \int_0^t \tilde{H}_D^{\bar{Y}}(t') dt' - tF_1 \quad (51)$$

After integration of each term, we obtain

$$\begin{aligned}
\Lambda_1(t) = & \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3(2\omega_{RF} \sin(2t\omega_{RF}) - \omega_R \operatorname{incos}(2t\omega_{RF}))}{2e^{int\omega_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{ZZ} + \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[-\frac{e^{-in\omega_R t} - 1}{2\omega_R in} + \frac{3in\omega_R}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{ZZ} + \\
& \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{-3(2\omega_{RF} \sin(2t\omega_{RF}) - \omega_R \operatorname{incos}(2t\omega_{RF}))}{2e^{int\omega_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{YY} + \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[-\frac{e^{-in\omega_R t} - 1}{2\omega_R in} + \frac{3in\omega_R}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{YY} \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{e^{-in\omega_R t} - 1}{\omega_R in} \right] I_{XX} \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{-3\omega_{RF}}{4\omega_{RF}^2 - \omega_R^2 n^2} \right] (I_{ZY} + I_{YZ}) \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3(2\omega_{RF} \cos(2t\omega_{RF}) + \omega_R \operatorname{insin}(2t\omega_{RF}))}{2e^{int\omega_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] (I_{ZY} \\
& + I_{YZ}) + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-1} C_n^{ij} t(2I_{YY}^{ij} - I_{XX}^{ij} - I_{ZZ}^{ij}) \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left[\left(\frac{t}{2} - \frac{1}{4\omega_{RF}} (3\sin(2\omega_{RF}t)) \right) I_{ZZ} \right. \\
& + \left. \left(\frac{t}{2} + \frac{1}{4\omega_{RF}} (3\sin(2\omega_{RF}t)) \right) I_{YY} - tI_{XX} - \left(\frac{3}{2\omega_{RF}} \sin^2(\omega_{RF}t) - t \right) (I_{ZY} + I_{YZ}) \right] \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left[\left(\frac{t}{2} + \frac{1}{4\omega_{RF}} (3\sin(2\omega_{RF}t)) \right) I_{ZZ} \right. \\
& + \left. \left(\frac{t}{2} - \frac{1}{4\omega_{RF}} (3\sin(2\omega_{RF}t)) \right) I_{XX} + \left(\frac{3}{2\omega_{RF}} \sin^2(\omega_{RF}t) - t \right) (I_{XZ} + I_{ZX}) - tI_{YY} \right] \\
& + \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} (e^{-in\omega_R t} - 1) H_{XX}^{ij} - \frac{1}{2} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} t H_{XX}^{ij} \\
& + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3(2\omega_{RF} \sin(2t\omega_{RF}) - \omega_R \operatorname{incos}(2t\omega_{RF}))}{2e^{int\omega_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{XX} + \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{e^{-in\omega_R t} - 1}{2\omega_R in} + \frac{3in\omega_R}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{XX} + \\
& \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{-3(2\omega_{RF} \sin(2t\omega_{RF}) - \omega_R \operatorname{incos}(2t\omega_{RF}))}{2e^{int\omega_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{ZZ} + \\
& - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{e^{-in\omega_R t} - 1}{2\omega_R in} + \frac{3in\omega_R}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] I_{ZZ} + \\
& \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{e^{-in\omega_R t} - 1}{\omega_R in} \right] I_{YY} + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{-3\omega_{RF}}{4\omega_{RF}^2 - \omega_R^2 n^2} \right] (I_{XZ} + \\
& I_{ZX}) + \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left[\frac{3(2\omega_{RF} \cos(2t\omega_{RF}) + \omega_R \operatorname{insin}(2t\omega_{RF}))}{2e^{int\omega_R}(4\omega_{RF}^2 - \omega_R^2 n^2)} \right] (I_{XZ} + I_{ZX}) - \\
& \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left(\frac{t}{2} + \frac{1}{4\omega_{RF}} (3\sin(2\omega_{RF}t)) \right) I_{XX} - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left[\left(\frac{t}{2} - \right. \right. \\
& \left. \left. \frac{1}{4\omega_{RF}} (3\sin(2\omega_{RF}t)) \right) I_{ZZ} - tI_{YY} \right] - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} \left[\frac{3}{2\omega_{RF}} \sin(\omega_{RF}t) \right] (I_{XZ} + I_{ZX}) - tF_1
\end{aligned} \tag{52}$$

These series of overwhelming equations (Eqs. 42, 45, and 52) gives an insight of the complexity of the dynamics of the spin systems during the pulse pulse sequence.

Considering the expression with only double quantum terms, we obtain the following form

$$\begin{aligned}\Lambda_1(t) = & \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} t (2I_{YY} - I_{XX} - I_{ZZ}) \\ & - \frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} t (2I_{XX} - I_{YY} - I_{ZZ}) \\ & - \frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} t \left(\frac{1}{2} - \emptyset\right) (I_{YY} - I_{XX})\end{aligned}\quad (53)$$

which is reduced to the expression

$$\Lambda_1(t) = \frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} a_{-n} C_n^{ij} t \left(\frac{1}{2} + \emptyset\right) (I_{YY} - I_{XX}) \quad (54)$$

Note that the above expression is proportional to t . The function $\Lambda_1(t)$ depends on both, the evolution time t and the parameter $\emptyset$. In the previous studies of BABA pulse sequence with δ -pulse sequences, the expression of the function $\Lambda_1(t)$ with only double quantum terms was found to be proportional to $e^{-im\omega_R t}$ as following [14]

$$\begin{aligned}\Lambda_1(t) = & \frac{1}{4\sqrt{6}} \sum_{i \neq j} b_{ij} [C_{-2}^{ij}(\alpha^{ij}, \beta^{ij}) e^{2i\gamma^{ij}} \left(\frac{1}{2i\omega_R}\right) (e^{2i\omega_R t} - 1)] (I_{XX}^{ij} + I_{YY}^{ij} - 2I_{ZZ}^{ij}) + \\ & + \frac{1}{4\sqrt{6}} \sum_{i \neq j} b_{ij} [-C_2^{ij}(\alpha^{ij}, \beta^{ij}) e^{-2i\gamma^{ij}} \left(\frac{1}{2i\omega_R}\right) (e^{-2i\omega_R t} - 1)] (I_{XX}^{ij} + I_{YY}^{ij} - 2I_{ZZ}^{ij}) \\ & + \frac{1}{4\sqrt{6}} \sum_{i \neq j} b_{ij} [-C_1^{ij}(\alpha^{ij}, \beta^{ij}) e^{-i\gamma^{ij}} \left(\frac{1}{i\omega_R}\right) (e^{-i\omega_R t} - 1) + \\ & C_{-1}^{ij}(\alpha^{ij}, \beta^{ij}) e^{i\gamma^{ij}} \left(\frac{1}{i\omega_R}\right) (e^{i\omega_R t} - 1)] (I_{XX}^{ij} + I_{YY}^{ij} - 2I_{ZZ}^{ij}) \\ & - \sum_{m=-\infty}^{+\infty} \frac{1}{im\omega_R} (e^{-im\omega_R t} - 1) \left[\frac{3}{2\sqrt{6}} \sum_{i \neq j} b_{ij} e^{-im\gamma^{ij}} \sum_{n=-2}^{+2} a_{m-n} C_n^{ij} (I_{YY}^{ij} - I_{XX}^{ij}) \right]\end{aligned}\quad (55 - a)$$

This tells us that the BABA pulse sequences with finite pulses produce less double quantum terms comparatively to the BABA pulse sequences with δ -pulse sequences.

III NUMERICAL ANALYSIS OF BABA

For $m = 1$, consider a system of 2 spins. Only DQ terms are considered for the function $\Lambda_1(t)$. We consider the simple case as in the reference [14] where the rotations are: $\alpha^{ij} = \beta^{ij} = \gamma^{ij} = 0$

The coefficient C_n^{ij} are $C_1 = -\frac{1}{\sqrt{3}} \sin(\theta) \cos(\theta) e^{-i\emptyset}$, $C_{-1} = 0$, $C_2 = \frac{1}{2\sqrt{6}} (\sin^2(\theta) e^{-2i\emptyset})$, $C_{-2} = 0$. For example, with $\theta = \pi/4$ and, $\phi = 0$, the coefficients are $C_1 = -\frac{1}{2\sqrt{3}}$ and $C_2 = \frac{1}{4\sqrt{6}}$. The function $\Lambda_1(t)$ is written as:

$$\Lambda_1(t) = \frac{3}{2\sqrt{6}} b_{ij} \left[a_{-1} C_1 \left(\frac{\tau_R + 4\tau_p}{2} \right) \psi + a_{-2} C_2 \left(\frac{\tau_R + 4\tau_p}{2} \right) \psi \right] (I_{YY} - I_{XX}) \quad (55 - b)$$

where

$$\psi = \frac{t}{\tau_R}, \quad (55 - c)$$

$$\varphi = \frac{2\tau_P}{\tau_R}. \quad (18)$$

$$a_{-1} = \frac{-1}{2\pi i} e^{-i\pi(1+\varphi)} [e^{i\pi(1-2\varphi)} - 1], \quad (55 - d)$$

and

$$a_{-2} = \frac{-1}{4\pi i} e^{-i2\pi(1+\varphi)} [e^{-i2\pi(1-2\varphi)} - 1]. \quad (55 - e)$$

After substitution of the Eqs. (55-c), (55-d), and (55-e) into (55-b), we obtain

$$\frac{\Lambda_1(\psi, \varphi)}{b_{ij}\tau_R} = \frac{3}{2\sqrt{6}} (-a_{-1} + \frac{1}{2\sqrt{2}} a_{-2}) \left(\frac{1}{2} + \varphi \right) \psi (I_{YY} - I_{XX}) \quad (55 - f)$$

We consider the case, $0.1 \leq \varphi \leq 0.606$, that corresponds to the spinning frequencies $\frac{\omega_R}{2\pi} = 5-10$ kHz, and to the recoupling RF fields $\frac{\omega_{RF}}{2\pi} = 25-50$ kHz. We generated two types of plots from the Eq. (55 - f). First, the plot of $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ versus $\psi = \frac{t}{\tau_R}$, while keeping $\varphi = \frac{2\tau_P}{\tau_R}$ constant corresponding to the Figure 3. The plot of $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ versus $\varphi = \frac{2\tau_P}{\tau_R}$, while keeping the time t constant corresponding to Figure 4.

Analysis of the Figures

Fig. 3 a) shows the dimensionless function $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ for BABA pulse sequence with finite pulse widths versus the dimensionless number $\psi = \frac{t}{\tau_R}$, for $\varphi = 0.1$. Fig. 3 b) shows the plot of the same function $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ versus $\psi = \frac{t}{\tau_R}$, for the two cases: $\varphi = 0.1$ and $\varphi = 0.606$. Due to the complexity of the function $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$, real, imaginary, and absolute parts are plotted separately as function of ψ . In Fig. 3 b), the symbols “square”, “plus”, and “hexagram” represent respectively the real, imaginary, and absolute parts of the function $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ for $\varphi = 0.606$. These functions depend on the DQ terms. Therefore, the study of the amplitude of DQ terms can be considered as a viable approach for controlling the complex spin dynamics of a spin system evolving under the dipolar interaction of BABA pulse sequence with finite widths. The plot can be considered as a quantitative representation of the amplitude of the DQ coherence as a function of ψ . The size of $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ determine the amplitude of the DQ coherence, which indicates the degree of efficiency of the scheme. A closer look at Fig. 3 (BABA with finite pulse widths) compared to Fig. 4 (BABA with delta-pulse sequences) [14] shows that the magnitude of the DQ terms of BABA with finite pulses is small compared to the magnitude of BABA with δ -pulse sequences, $\left| \frac{\Lambda_1(t)}{b_{ij}\tau_R} \right|_{Finite\ Pulse} < \left| \frac{\Lambda_1(t)}{b_{ij}\tau_R} \right|_{\delta-Pulse}$ as expected.

Fig. 4 a) shows the plot of the function $\frac{\Lambda_1(t)}{b_{ij}}$ for BABA pulse sequence with finite pulse widths versus the dimensionless number $\varphi = \frac{2\tau_P}{\tau_R}$, for $t = 1$ ms. Fig. 4 b) shows the plot of the same function $\frac{\Lambda_1(t)}{b_{ij}}$ versus $\varphi = \frac{2\tau_P}{\tau_R}$, for the two cases: $t = 1$ ms and $t = 2$ ms. As with Fig. 3, due to the complexity of the function $\frac{\Lambda_1(t)}{b_{ij}}$, real, imaginary, and absolute parts are plotted separately as function of φ . In Fig. 4 b), the symbols “square”, “plus”, and “hexagram” represent respectively the real, imaginary, and absolute parts of the function $\frac{\Lambda_1(t)}{b_{ij}}$ for $t = 2$ ms. These functions depend on the DQ terms. It can easily be seen that, when $\varphi = \frac{2\tau_P}{\tau_R}$ increases, the magnitude of the double quantum terms decreases, as expected. When $\varphi \rightarrow 0$, the magnitude of the DQ term $\rightarrow$ maximum corresponding to the delta-pulse sequence. However, when $\varphi = 0.5$ corresponding to $\tau_P = \frac{\tau_R}{4}$, we have $\frac{\Lambda_1(t)}{b_{ij}} = 0$. The strength of the DQ terms decreases, cancel and build up again. This dynamic predict that a full decoupling is possible, which occurs at $\varphi = 0.5$.

IV DISCUSSION OF THE RESULTS OBTAINED

The FME approach is essentially distinguished from AHT with its function $\Lambda_1(t)$ which provides an easy and alternative way for evaluating the spin behavior in between the stroboscopic observation points. The approach provides the option to evaluate the spin evolution between the time points of detection. However, the AHT and FT result respectively in average and effective Hamiltonians that are expanded in a set of terms of increasing orders. These Hamiltonians are in general connected with stroboscopic detection schemes. The FME approach has the advantage to overcome the limitations of the stroboscopic detection schemes. We have limited our investigation to the first order F_1 of the effective Hamiltonian. This order is identical to its counterparts in AHT and FT. However, the $\Lambda_1(t)$ function is connected to the appearance of features like spinning sidebands in MAS. The evaluation of $\Lambda_1(t)$ is useful in this study especially to measure the level of productivity of double quantum terms between δ -pulse sequences and sequences when finite widths are taken into consideration. The FME provides a quick means to calculate higher order terms allowing the disentanglement of the stroboscopic observation and effective Hamiltonian that will be useful to describe spin dynamics in solid-state NMR. Note that, the FME scheme is not restricted to dipolar or quadrupolar interaction, and can be applied to any case. The FME approach is unique through its expression for $\Lambda_1(t)$. Here, we did not considered the importance of the boundary conditions (origin of time), which provides a natural choice of $\Lambda_1(0)$ to simplify calculation of higher order terms that we neglected. From Eqs. (54) and (55), it can be seen that the BABA with δ -pulse sequences produces more DQ terms than the BABA with finite pulse width.

The plot of the magnitude of the double quantum term of $\Lambda_1(t)$ as a function of the pulse length gives a basics understanding of the experiment such as how to select robust finite pulse widths, how to select finite pulse widths that maximize or minimize double quantum terms. The study of this function could be helpful to predict the conditions of decoupling such as the one shown in the particular case described in part III.

V CONCLUSION

This article calculates the first order term of the Floquet-Magnus expansion approach for the spin system evolving under the dipolar interaction and subject to BABA with finite pulse width. The first order term F_1 is identical to the first order average Hamiltonian. This F_1 term in the BABA scheme with finite pulse sequence is similar to the result obtained with the BABA scheme when considering the delta-function RF pulse, i. e. $\tau_p \rightarrow 0$. The obtained Eq. (44) of F_1 is valid for the multispin systems with the dipolar interactions and can also be extended to quadrupolar interactions. Eqs. (54) and (55) are very significant results in this article. Using the FME scheme, we describe theoretically the fact that the BABA sequence performance is related to double quantum excitation. This result basically means that this pulse sequence allows a scaling factor for the dipolar interaction that depends on how long the finite pulses are. In the future, the impact of the resulting formula against experimental data of the spin dynamics will be checked. Also, the perturbations due to simultaneously excited zero quantum coherences will be considered, essentially for providing suggestions for optimum setup conditions. With the quite advanced micro-coil technology, the possibility to set a limit on applicable pulse lengths to render the practical relevance of the obtained equations rather deserves further attention. This analysis is the subject of a forthcoming paper. Another interesting treatment that is amenable to similar results obtained here consists in using a free mathematica script developed by Brinkmann and Levitt [36, 37, 38, 39].

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42. We thanks the anonymous referee for pointing this out.

VII APPENDIX

A1

$$\int_0^{\tau_P} e^{-in\omega_R t} (3\cos^2(\omega_{RF} t) - 1) dt = \frac{3(2\omega_{RF} \sin(2\tau_P \omega_{RF})) - 3(\omega_R \sin(2\tau_P \omega_{RF}))}{2e^{(\tau_P \omega_R in)} (4\omega_{RF}^2 - \omega_R^2 n^2)} + \frac{(1 - e^{(-\tau_P \omega_R in)})}{2\omega_R in} + \frac{3\omega_R in}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \quad (56)$$

$$\int_0^{\tau_P} e^{-in\omega_R t} (3\sin^2(\omega_{RF} t) - 1) dt = -\frac{3(2\omega_{RF} \sin(2\tau_P \omega_{RF})) - 3\omega_R \sin(2\tau_P \omega_{RF})}{2e^{(\tau_P \omega_R in)} (4\omega_{RF}^2 - \omega_R^2 n^2)} + \frac{(1 - e^{(-\tau_P \omega_R in)})}{2\omega_R in} + \frac{3\omega_R in}{2(4\omega_{RF}^2 - \omega_R^2 n^2)} \quad (57)$$

$$\int_0^{\tau_P} e^{-in\omega_R t} dt = \frac{(1 - e^{(-\tau_P \omega_R in)})}{\omega_R in} \quad (58)$$

$$\int_0^{\tau_P} e^{-in\omega_R t} \cos(\omega_{RF} t) \sin(\omega_{RF} t) dt = \frac{\omega_{RF}}{\omega_{RF}^2 - \omega_R^2 n^2} - \frac{2\omega_{RF} \cos(2\omega_{RF} \tau_P) + \omega_R \sin(2\omega_{RF} \tau_P)}{2e^{in\omega_R \tau_P} (4\omega_{RF}^2 - \omega_R^2 n^2)} \quad (59)$$

$$\int_{\frac{\tau_R}{2} - \tau_P}^{\frac{\tau_R}{2}} (3\sin^2(\omega_{RF} t) - 1) dt = \frac{\tau_P}{2} - \frac{1}{4\omega_{RF}} [3(\sin(\omega_{RF} \tau_R) + \sin(2\omega_{RF} \tau_P - \omega_{RF} \tau_R))] \quad (60)$$

$$\int_{\frac{\tau_R}{2} - \tau_P}^{\frac{\tau_R}{2}} (3\cos^2(\omega_{RF} t) - 1) dt = \frac{\tau_P}{2} + \frac{1}{4\omega_{RF}} [3(\sin(\omega_{RF} \tau_R) + \sin(2\omega_{RF} \tau_P - \omega_{RF} \tau_R))] \quad (61)$$

$$\int_{\frac{\tau_R}{2} - \tau_P}^{\frac{\tau_R}{2}} \cos(\omega_{RF} t) \sin(\omega_{RF} t) dt = -\frac{1}{4\omega_{RF}} [\cos(\omega_{RF} \tau_R) - \cos(2\omega_{RF} \tau_P - \omega_{RF} \tau_R)] \quad (62)$$

$$\int_{\frac{\tau_R}{2}}^{\frac{\tau_R}{2} + \tau_P} (3\sin^2(\omega_{RF} t) - 1) dt = \frac{\tau_P}{2} + \frac{1}{4\omega_{RF}} [3(\sin(\omega_{RF} \tau_R) - \sin(2\omega_{RF} \tau_P + \omega_{RF} \tau_R))] \quad (63)$$

$$\int_{\frac{\tau_R}{2}}^{\frac{\tau_R}{2} + \tau_P} (3\cos^2(\omega_{RF} t) - 1) dt = \frac{\tau_P}{2} - \frac{1}{4\omega_{RF}} [3(\sin(\omega_{RF} \tau_R) - \sin(2\omega_{RF} \tau_P + \omega_{RF} \tau_R))] \quad (64)$$

$$\int_{\frac{\tau_R}{2}}^{\frac{\tau_R}{2} + \tau_P} \cos(\omega_{RF} t) \sin(\omega_{RF} t) dt = \frac{1}{4\omega_{RF}} [\cos(\omega_{RF} \tau_R) - \cos(2\omega_{RF} \tau_P + \omega_{RF} \tau_R)] \quad (65)$$

$$\int_{\tau_R - \tau_P}^{\tau_R} (3\sin^2(\omega_{RF} t) - 1) dt = \frac{\tau_P}{2} - \frac{1}{4\omega_{RF}} [3(\sin(2\omega_{RF} \tau_R) + \sin(2\omega_{RF} \tau_P - 2\omega_{RF} \tau_R))] \quad (66)$$

$$\int_{\tau_R - \tau_P}^{\tau_R} (3\cos^2(\omega_{RF} t) - 1) dt = \frac{\tau_P}{2} + \frac{1}{4\omega_{RF}} [3(\sin(2\omega_{RF} \tau_R) + \sin(2\omega_{RF} \tau_P - 2\omega_{RF} \tau_R))] \quad (67)$$

$$\int_{\tau_R - \tau_P}^{\tau_R} \cos(\omega_{RF} t) \sin(\omega_{RF} t) dt = -\frac{1}{4\omega_{RF}} [\cos(2\omega_{RF} \tau_R) - \cos(2\omega_{RF} \tau_P - 2\omega_{RF} \tau_R)] \quad (68)$$

A2

Using the relation

$$\omega_R \tau_R = 2\pi \quad (69)$$

We have

$$\begin{aligned}
 & -\frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{1}{in\omega_R \tau_R} \right) \left(e^{-in\omega_R \tau_R} - e^{-i\frac{n}{2}\omega_R \tau_R} \right) \\
 & = -\frac{1}{2\sqrt{6}} \sum_{i \neq j} b_{ij} \sum_{n=-2}^{+2} C_n^{ij} e^{-in\gamma^{ij}} \left(\frac{1}{i2\pi} \right) \underbrace{\frac{1}{n} (1 - e^{-i\pi n})}_{S_n} \equiv S \quad (70)
 \end{aligned}$$

Next,

$$C_0^{ij} = 0 \quad (71)$$

$$S_1 = 2 \quad (72)$$

$$S_{-1} = -2 \quad (73)$$

$$S_2 = 0 \quad (74)$$

$$S_{-2} = 0 \quad (75)$$

Lead to

$$S = 0 \quad (76)$$

Fig. 1.
BABA pulse sequence with δ -pulse width

Fig. 2.
BABA pulse sequence with finite pulse width

Fig. 3.

Numerical Functions of Finite Pulse BABA Sequence with $\frac{\Lambda_1(t)}{b_{ij}\tau_R}$ versus $\psi = \frac{t}{\tau_R}$

a) For $t = 1 \text{ ms}$ b) For $t = 1 \text{ ms}$ and $t = 2 \text{ ms}$

Fig. 4.

Numerical Functions of Finite Pulse BABA Sequence with $\frac{\Lambda_1(t)}{b_{i,j}}$ versus $\phi = \frac{2\tau_P}{\tau_R}$

Table 1

Interval	Time	$H_D^{\sim}(t)$
δ_1	$0 \leq t \leq \tau_P$	$\frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) \frac{1}{\sqrt{6}} [(3\cos^2\alpha - 1)I_{ZZ}^{ij} + (3\sin^2\alpha - 1)I_{YY}^{ij}]$ $- \frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) \frac{1}{\sqrt{6}} [I_{XX}^{ij} + 3\sin\alpha \cos\alpha (I_{ZY}^{ij} + I_{YZ}^{ij})]$
δ_2	$\tau_P \leq t \leq \frac{\tau}{2} - \tau_P$	$A_{ij} \frac{1}{\sqrt{6}} (2I_{YY}^{ij} - I_{ZZ}^{ij} - I_{XX}^{ij})$
δ_3	$\frac{\tau}{2} - \tau_P \leq t \leq \frac{\tau_R}{2}$	$A_{ij} \frac{1}{\sqrt{6}} [(3\sin^2\alpha - 1)I_{ZZ}^{ij} + (3\cos^2\alpha - 1)I_{YY}^{ij}]$ $- A_{ij} \frac{1}{\sqrt{6}} [I_{XX}^{ij} + 3\sin\alpha \cos\alpha (I_{ZY}^{ij} + I_{YZ}^{ij})]$
δ_4	$\frac{\tau_R}{2} \leq t \leq \frac{\tau_R}{2} + \tau_P$	$A_{ij} \frac{1}{\sqrt{6}} [(3\cos^2\alpha - 1)I_{ZZ}^{ij} + (3\sin^2\alpha - 1)I_{XX}^{ij}]$ $+ A_{ij} \frac{1}{\sqrt{6}} [-I_{YY}^{ij} + 3\sin\alpha \cos\alpha (I_{XZ}^{ij} + I_{ZX}^{ij})]$
δ_5	$\frac{\tau_R}{2} + \tau_P \leq t \leq \tau_R - \tau_P$	$\frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) (1 - \theta) \frac{1}{\sqrt{6}} (2I_{XX}^{ij} - I_{ZZ}^{ij} - I_{YY}^{ij})$
δ_6	$\tau_R - \tau_P \leq t \leq \tau_R$	$\frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) (1 - \theta) \frac{1}{\sqrt{6}} [(3\cos^2\alpha - 1)I_{XX}^{ij} + (3\sin^2\alpha - 1)I_{ZZ}^{ij}]$ $+ \frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) \frac{1}{\sqrt{6}} (1 - \theta) [-I_{YY}^{ij} + 3\sin\alpha \cos\alpha (I_{XZ}^{ij} + I_{ZX}^{ij})]$

Where

$$a = \omega_R F t, A_{ij} = \frac{1}{2} \sum_{i \neq j} \omega_D^{ij}(t) \sum_{k=-\infty}^{k=+\infty} a_k e^{-ik(\omega_R t + \gamma^{ij})}$$

$$\text{and } \theta(t) = \sum_{k=-\infty}^{+\infty} a_k \exp(-ik\omega_R t)$$