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Effects of finite pulse width on free induction decay

N.A. Sergeev [1]

Higher Teacher Education School, Słupsk, 76-200, Poland

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Abstract

The shape of the free induction decay for the case in which the amplitude B_1 ($B_1 = \omega_1/\gamma$) of the RF pulse is comparable with local magnetic fields at sites of the nuclei is considered. It is shown that the shape of the FID $G(t+\tau)$ (where τ is the width of a RF pulse, t is the time after a RF pulse) depends on the shape of the FID $G_0(t)$ following a hard, delta function RF pulse and on the shape of the function $F_0(\tau)$, which describes the evolution of the longitudinal nuclear magnetization M_z under the effect of the hamiltonian $(H_0 - \omega_1 I_Y)$, where H_0 is the interaction hamiltonian of the nuclear spin system. Calculations of $F_0(\tau)$ and $G(t+\tau)$ for different interaction hamiltonians H_0 are presented.

Keywords: Free induction decay; RF pulse with finite width; Superoperatory; Dipolar interactions; Quadrupolar interactions

1. Introduction

The free induction decay signal (FID) is the signal observed in the nuclear spin system after the radio frequency (RF) pulse. If a state of nuclear spin system at time $t = 0$ can be described by a spin temperature and a RF field is much larger than the NMR linewidth, so that $\omega_1\tau = \text{const}$ at $\tau \to 0$ and $\omega_1 \to \infty$, the shape of the FID following a RF pulse does not depend on the angle by which the pulse has rotated the magnetization [1]. In this case the Fourier transform of the FID is identical to the shape of the CW spectrum obtained by the slow passage of a continuous RF field [2]. At the present time the approximation of a hard, delta function RF pulse (delta RF pulse) is widely applied in NMR of a liquids and solids. For liquids this approximation works well, but for solids the experimental RF pulse has the strength and the duration, which have comparable values with the linewidth of the NMR spectrum and the time of the FID decay. The effects of the finite pulse width on the shape of the FID were discussed in [3–7]. Barnaal and Lowe [3] calculated the FID of the two identical spin $-1/2$ nuclei subject to the dipolar interaction and demonstrated that the FID has its origin at approximately the center of the RF pulse. Bloom, Davis and Valic [6] obtained the same result for the nuclei ^2H subject to the quadrupolar interaction and introduced the spectral distortion factor which allows the correction of the spectrum NMR obtained by the Fourier transform of the FID. Their results are quite adequate for the case of the deuterium NMR but at the present time it is not obvious how to determined similar correction factors for the cases when the nuclear spin $I > 1$ and when there are no isolated pairs of spin $1/2$ nuclei in solids.

[1] Corresponding address: Institute of Physics, University of Szczecin, ul. Wielkopolska 15, 70-451 Szczecin, Poland.

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In this paper we consider the effects of the finite pulse width on the FID shape for the general cases, when there are no isolated spin groups and $I > 1$.

2. Theory

Consider an ensemble of a nuclear spins subject to a strong static magnetic field B_0k. The reduced equilibrium density matrix operator at $t = 0$ can be expressed in the high-temperature approximation as [1]

$$\rho(0) = I_Z$$

After the RF pulse (RF field in a coordinate frame rotating with the Larmor frequency $\omega_0 = -\gamma B_0$ about B_0 is B_1j; γ is the magnetogyric ratio of the nuclei) the density operator in the rotating coordinate frame is given at $t = \tau$ (τ is the width of a RF pulse) by

$$\rho(\tau) = R(\tau)\rho(0)R^{-1}(\tau),$$

where

$$R = \exp\{-i\tau(H_0 - \omega_1 I_Y)\}$$

and H_0 is the interaction hamiltonian ($\hbar = 1$) of the nuclear spin system in the rotating frame; $\omega_1 = \gamma B_1$.

After the RF pulse the evolution of the density matrix operator is described by the hamiltonian H_0 and at the time t the density matrix operator is

$$\rho(t+\tau) = U(t)R(\tau)\rho(0)R^{-1}(\tau)U^{-1}(t),$$

where

$$U(t) = \exp\{-itH_0\}.$$

The transient response of the ensemble of spins in the rotating frame—the signal of the free induction decay, is given by [1]

$$G(t+\tau) = \frac{\mathrm{Tr}[\varrho(t+\tau)I_X]}{\mathrm{Tr}(I_X^2)}. \quad (1)$$

Introducing the Liouville superoperators [8–11]

$$L_0 = [H_0, \ldots],$$

$$L_r = [\ldots, (H_0 - \omega_1 I_Y)], \quad (2)$$

expression (1) can be written as

$$G(t+\tau) = \frac{\langle I_Z | \exp(i\tau L_r) \cdot \exp(itL_0) | I_X \rangle}{\langle I_X | I_X \rangle}, \quad (3)$$

where [8–11]

$$\langle A|B\rangle = \mathrm{Tr}(A^+ B),$$

and A^+ is the operator hermitian conjugate of A.

As was shown in [9] the ket-vector $\exp(itL_0)|I_X\rangle$ in the Liouville space can be expressed as

$$\exp(itL_0)\cdot|I_X\rangle = \sum_n G_n(t)|n\rangle, \quad (4)$$

where

$$|0\rangle = I_X,$$

$$|n\rangle = L_0^n|0\rangle - \sum_{k=0}^{n-1} \frac{\langle k|L_0^n|0\rangle}{\langle k|k\rangle}|k\rangle, \quad (5)$$

and

$$\langle n|k\rangle = 0, \quad n \neq k \quad (6)$$

The functions $G_n(t)$ are the solutions of the system of equations [9,10]

$$-i\frac{d}{dt}G_0(t) = \nu_0^2 \cdot G_1(t),$$

$$-i\frac{d}{dt}G_n(t) = G_{n-1}(t) + \nu_n^2 \cdot G_{n+1}(t), \quad (7)$$

where

$$\nu_n^2 = \frac{\langle n+1|n+1\rangle}{\langle n|n\rangle}. \quad (8)$$

From Eq. (4) and Eq. (5) we see that the function

$$G_0(t) = \frac{\langle I_X|\exp(itL_0)|I_X\rangle}{\langle I_X|I_X\rangle} \quad (9)$$

is the function, which describes the shape of the FID after a hard delta 90°-pulse (ω_1 $\tau = 90°$, $\omega_1 \to \infty$, $\tau \to 0$) [1,9].

We can establish the following properties of $G_n(t)$ at time $t = 0$ from Eq. (7) and Eq. (9)

$$G_0(0) = 1,$$

$$G_n(0) = 0, \quad n > 0.$$

The values ν_n^2 in Eq. (7) are simply related to the Van-Vleck moments M_n of NMR spectrum [9–11]

$$\nu_0^2 = M_2,$$

$$\nu_1^2 = \frac{M_4 - M_2^2}{\nu_0^2}, \ldots$$

By analogy with Eq. (4) and Eq. (5), we can write the bra vector $< I_z|\exp(\mathrm{i}\tau L_r)$ as

$$\langle I_Z|\exp(\mathrm{i}\tau L_r) = \sum_\beta F_\beta(\tau)\langle\beta|, \qquad (10)$$

where

$$\langle 0_r| = I_Z,$$

$$\langle\beta| = \langle 0_r|L_r^\beta - \sum_{\alpha=0}^{\beta-1}\frac{\langle 0_r|L_r^\beta|\alpha\rangle}{\langle\alpha|\alpha\rangle}\langle\alpha|, \qquad (11)$$

and

$$\langle\alpha|\beta\rangle = 0, \quad \alpha\neq\beta.$$

The functions $F_\beta(\tau)$ are determined by the system of equations

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}\tau}F_0(\tau) = \Omega_0^2 F_1(\tau),$$

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}\tau}F_\beta(\tau) = F_{\beta-1}(\tau) + \Omega_\beta^2 F_{\beta+1}(\tau), \qquad (12)$$

where

$$\Omega_\beta^2 = \frac{\langle\beta+1|\beta+1\rangle}{\langle\beta|\beta\rangle}.$$

From Eq. (10) and Eq. (11) we see that the function

$$F_0(\tau) = \frac{\langle I_Z|\exp(\mathrm{i}\tau L_r)|I_Z\rangle}{\langle I_Z|I_Z\rangle} \qquad (13)$$

describes the evolution of the longitudinal magnetization $M_Z = \gamma\hbar I_Z$ in a rotating coordinate frame under the effect of the hamiltonian $(H_0 - \omega_1 I_Y)$.

At time $\tau = 0$, from Eq. (12) and Eq. (13) it follows that

$$F_0(0) = 1,$$

$$F_n(0) = 0, \quad n > 0.$$

The power series expansion of $\exp(\mathrm{i}\tau L_r)$ in Eq. (13) leads to

$$F_0(\tau) = \sum_{p=0}^{\infty}\frac{(\mathrm{i})^p}{p!}C_p\tau^p,$$

where

$$C_p = \frac{\langle I_Z|L_r^p|I_Z\rangle}{\langle I_Z|I_Z\rangle} \qquad (14)$$

are the moments of the curve obtained by the Fourier transform of $F_0(\tau)$.

The values Ω_β^2 in Eq. (12) are related to the moments C_p

$$\Omega_0^2 = C_2 = \omega_1^2, \quad \Omega_1^2 = \frac{C_4 - C_2^2}{C_2},$$

$$\Omega_2^2 = \frac{1}{C_2}\frac{C_6C_2 - C_4^2}{C_4 - C_2^2}, \ldots \qquad (15)$$

By inserting Eq. (4) and Eq. (10) into Eq. (3) we obtain the following general expression for the shape of the FID after the RF pulse

$$G(t+\tau) = \sum_{n,\beta}\frac{\langle\beta|n\rangle}{\langle 0|0\rangle}G_n(t)F_\beta(\tau). \qquad (16)$$

In a strong static magnetic field B_0, the hamiltonian H_0 commutes with I_Z (secular approximation [1])

$$[H_0, I_Z] = 0. \qquad (17)$$

Using Eq. (5), Eq. (11) and Eq. (17) we can calculate $<\beta|n>/<0|0>$. These calculations lead to the following expression for the shape of the FID

$$G(t+\tau) = -\frac{1}{\omega_1}G_0(t)\frac{\mathrm{d}}{\mathrm{d}t}F_0(\tau) +$$

$$+\frac{\omega_1}{M_2}\frac{\mathrm{d}}{\mathrm{d}t}G_0(t)\left[\frac{1}{\omega_1^2}\frac{\mathrm{d}^2}{\mathrm{d}\tau^2}F_0(\tau) + F_0(\tau)\right] + \cdots \qquad (18)$$

In Eq. (18) we retain only the functions $G_n(t)$ and $F_\beta(\tau)$ with $n = 0, 1$; $\beta = 0, 1, 2$. From Eq. (12) we see that this approximation of $G(t+\tau)$ is a good approximation, when $\|H_0\|\tau \ll 1$.

3. Discussion of the results obtained

In this section we discuss the evaluations of the functions $F_0(\tau)$ and $G(t+\tau)$ for the different RF pulses and the different interaction hamiltonians H_0.

3.1. Delta RF pulse ($\omega_1\tau = const$, $\omega_1 \to \infty$, $\tau \to 0$)

If the RF pulse is the delta RF pulse then the interaction hamiltonian H_0 in the superoperator L_r can be ignored and from Eq. (14) and Eq. (15) we obtain

$$C_{2p} = \omega_1^{2p}.$$

$$\Omega_0^2 = \omega_1^2;\ \Omega_1^2 = \Omega_2^2 = \ldots = \Omega_\alpha^2 = 0. \qquad (19)$$

Substituting Eq. (19) into Eq. (12) we obtain

$$F_0(\tau) = \cos(\omega_1\tau) \tag{20}$$

and from Eq. (18) we have for $G(t+\tau)$ the well known result [1]

$$G(t+\tau) = G_0(t)\sin(\omega_1\tau).$$

3.2. The interaction hamiltonian is $H_0 = \Delta I_Z$

For this case from Eq. (14) we have

$$C_2 = \omega_1^2,\ C_4 = \omega_1^4 + \Delta^2\omega_1^2,$$

$$C_6 = \omega_1^6 + 2\omega_1^4\Delta^2 + \omega_1^2\Delta^4,$$

and

$$\Omega_0^2 = \omega_1^2,\ \Omega_1^2 = \Delta^2,\ \Omega_2^2 = 0.$$

The system of Eq. (12) can be written as

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}\tau}F_0(\tau) = \omega_1^2 F_1(\tau);$$

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}\tau}F_1(\tau) = F_0(\tau) + \Delta^2 F_2(\tau),$$

$$-\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}\tau}F_2(\tau) = F_1(\tau).$$

The solution of this system of equations for $F_0(\tau)$ is

$$F_0(\tau) = \frac{\Delta^2}{\omega_1^2 + \Delta^2} + \frac{\omega_1^2}{\omega_1^2 + \Delta^2}cos\left(\sqrt{\omega_1^2 + \Delta^2}\,\tau\right). \tag{21}$$

For the hamiltonian $H_0 = \Delta I_z$ the FID $G_0(t)$ is described from Eq. (9) by

$$G_0(t) = \cos(\Delta t). \tag{22}$$

By inserting Eq. (21) and Eq. (22) into Eq. (18) we obtain the well known result also [1]

$$G(t+\tau) = \cos\theta_{ef}\sin(\omega_{ef}\tau)\cos(\Delta t) - \sin(2\theta_{ef})\sin^2\left(\frac{1}{2}\omega_{ef}\tau\right)\sin(\Delta t), \tag{23}$$

where

$$\omega_{ef} = \sqrt{\omega_1^2 + \Delta^2},$$

and

$$\cos\theta_{ef} = \frac{\omega_1}{\omega_{ef}}.$$

3.3. The interaction hamiltonian is $H_0 = aI_Z^2$ and the nuclear spin is 1 ($I = 1$)

For this case from Eq. (14) we have

$$\Omega_0^2 = \omega_1^2,\ \Omega_1^2 = a^2,\ \Omega_2^2 = \omega_1^2,\ \Omega_3^2 = 0$$

and from Eq. (9) for $G_0(t)$ we obtain

$$G_0(t) = \cos(at). \tag{24}$$

For $F_0(\tau)$ we have from Eq. (12)

$$F_0(\tau) = \frac{a}{\sqrt{a^2 + 4\omega_1^2}}\sin\left(\frac{a\tau}{2}\right)\sin\left(\frac{\sqrt{a^2 + 4\omega_1^2}\,\tau}{2}\right) + \cos\left(\frac{a\tau}{2}\right)\cos\left(\frac{\sqrt{a^2 + 4\omega_1^2}\,\tau}{2}\right). \tag{25}$$

Insertion of Eq. (24) and Eq. (25) into Eq. (18) also yields the well known result [3,6]

$$G(t+\tau) = \frac{2\omega_1}{\sqrt{a^2 + 4\omega_1^2}}\cos\left[a\left(t + \frac{\tau}{2}\right)\right] \times \sin\left(\frac{\sqrt{a^2 + 4\omega_1^2}\,\tau}{2}\right). \tag{26}$$

If we replace time t(t is the time after a RF pulse) with $t_1 = t + \tau$ and assume that $\omega_1 > a$, then from Eq. (26) we see that the signal of the FID has a maximum at $t_1 = (\tau/2)$[3,6].

3.4. The interaction hamiltonian is the dipolar hamiltonian

If H_0 is the truncated dipolar hamiltonian [1]

$$H_{dZ} = \sum_{i>j} b_{ij}(2I_{iZ}I_{jZ} - I_{iX}I_{jX} - I_{iY}I_{jY}), \tag{27}$$

then from Eq. (14) we obtain the following expressions for the first moments C_p

$$C_2 = \omega_1^2,$$

$$C_4 = \omega_1^4 + \omega_1^2 M_2,$$

$$C_6 = \omega^6 + 3M_2\omega_1^4 + M_4\omega_1^2. \tag{28}$$

Using Eq. (28) we find from Eq. (15)

$$\Omega_0^2 = \omega_1^2,$$
$$\Omega_1^2 = M_2,$$
$$\Omega_2^2 = \frac{M_4 - M_2^2}{M_2} + \omega_1^2, \cdots. \quad (29)$$

The system of an differential Eq. (12) may be transformed into the system of an algebraic equations by the Laplace transformation method [9,10]

$$f(s) = \int_0^\infty f(t) e^{-st} \mathrm{d}t.$$

The transformed system of algebraic equations is

$$\mathrm{i} s F_0(s) + \Omega_0^2 F_1(s) = \mathrm{i},$$
$$F_{\alpha-1}(s) + \mathrm{i} s F_\alpha(s) + \Omega_\alpha^2 F_{\alpha+1}(s) = 0. \quad (30)$$

The formal solution of the system of Eq. (30) for $F_0(s)$ is given by [9]

$$F_0(s) = \cfrac{1}{s + \cfrac{\Omega_0^2}{s + \cfrac{\Omega_1^2}{s + \cfrac{\Omega_2^2}{s + \ldots}}}}. \quad (31)$$

If $\Omega_1^2 = \Omega_2^2 = \ldots = 0$, then from Eq. (31) we obtain

$$F_0(s) = \frac{s}{s^2 + \omega_1^2}. \quad (32)$$

The opposite Laplace transformation of Eq. (32) gives Eq. (20). If $\Omega_1^2 = M_2 = \Delta^2$, $\Omega_2^2 = \Omega_3^2 = \ldots = 0$, then from Eq. (31) follows the Eq. (21). The sum of the infinite fraction in Eq. (31) may be calculated assuming that: (a) $\Omega_n^2 = \text{const}$ at $n > k$ and (b) $\Omega_n^2 = 0$ at $n > k$ [9,10].

Let $\Omega_3^2 = \Omega_4^2 = \ldots = 0$. Using Eq. (31) and performing the Laplace transformation of $F_0(s)$ we obtain

$$F_0(\tau) = \frac{1}{2}[\cos(\lambda_1\tau) + \cos(\lambda_2\tau)] + \frac{1}{2}\frac{M_4}{M_2}\frac{\cos(\lambda_1\tau) - \cos(\lambda_2\tau)}{\lambda_2^2 - \lambda_1^2}, \quad (33)$$

where

$$\lambda_{1,2}^2 = \frac{(2\omega_1^2 M_2 + M_4) \pm \sqrt{M_4^2 + 4\omega_1^2 M_2^3}}{2M_2}.$$

If $M_4 = M_2^2 = a^4$, then from Eq. (33) follows the function (25) for the isolated two-spin system with dipole–dipole interactions.

3.5. The hard RF pulse with the finite width. The interaction hamiltonian is the dipolar hamiltonian

For this case we write the hamiltonian (27) in the form

$$H_{\mathrm{dZ}} = -\frac{1}{2}H_{\mathrm{dY}} + \frac{3}{2}\sum_{i>j} b_{ij}(I_{iZ}I_{jZ} - I_{iX}I_{jX}). \quad (34)$$

where

$$H_{\mathrm{dY}} = \sum_{i>j} b_{ij}(2I_{iY}I_{jY} - I_{iX}I_{jX} - I_{iZ}I_{jZ}). \quad (35)$$

It is now assumed that $\omega_1 > \|H_{\mathrm{dZ}}\|$, but the RF pulse is not the delta-pulse. In this case the elements noncommuted with I_Y in the hamiltonian H_{dZ} can be ignored [3,4] and the effective superoperator L_r becomes equal

$$L_r = \left[\ldots, -\omega_1 I_Y - \frac{1}{2}H_{\mathrm{dY}}\right]. \quad (36)$$

Insertion of Eq. (36) into Eq. (13) yields

$$F_0(\tau) = \cos(\omega_1\tau) G_0\left(-\frac{1}{2}\tau\right), \quad (37)$$

where the function $G_0(t)$ describes the FID after a hard 90° delta-pulse (Eq. (9)). Using Eq. (37) we obtain from Eq. (18) that at $\omega_1\tau = 90°$

$$G(t+\tau) = G_0(t) G_0\left(-\frac{1}{2}\tau\right) + \frac{1}{M_2}\frac{\mathrm{d}}{\mathrm{d}t} \times G_0(t) \frac{\mathrm{d}}{\mathrm{d}\left(-\frac{1}{2}\tau\right)} G_0\left(-\frac{1}{2}\tau\right) + \ldots, \quad (38)$$

From Eq. (9) for the function $G_0(t)$ we obtain (see Appendix A)

$$G_0(t_1 + t_2) = G_0(t_1)G_0(t_2) + \nu_0^2 G_1(t_1) G_1(t_2) + \nu_0^2\nu_1^2 G_2(t_1) G_2(t_2) + \ldots. \quad (39)$$

Using Eq. (39), Eq. (7) and Eq. (A4) we have from Eq. (38)

$$G(t+\tau) = G_0\left(t + \frac{1}{2}\tau\right) + \ldots. \quad (40)$$

It follows from Eq. (40), that for the case of the hard RF pulse with the finite width and at $\omega_1\tau = 90°$ the FID of the nuclear multispin system subject to the dipolar interaction has its origin, as in the case of the two-spin system, at the center of RF pulse.

3.6. The hard RF pulse with the finite width. The interaction hamiltonian is the quadrupolar hamiltonian ($I \geq 1$)

The truncated quadrupolar hamiltonian is [1]

$$H_{QZ} = \sum_{i=1}^{N} \omega_{Qi}\left[3I_{iZ}^2 - (I_i \cdot I_i)\right]. \quad (41)$$

It is convenient to write the hamiltonian (41) in the form

$$H_{QZ} = -\frac{1}{2}H_{QY} + \frac{3}{2}\sum_{i=1}^{N} \omega_{Qi}\left(I_{iZ}^2 - I_{iX}^2\right), \quad (42)$$

where

$$H_{QY} = \sum_{i=1}^{N} \omega_{Qi}\left[3I_{iY}^2 - I(I+1)\right]. \quad (43)$$

From Eq. (42) and Eq. (43) it follows

$$[H_{QY}, I_Y] = 0, \quad \left[\left(I_{iZ}^2 - I_{iX}^2\right), I_Y\right] \neq 0.$$

It is now again assumed that $\omega_1 > \|H_{QZ}\|$, but the RF pulse is not the delta-pulse. In this case in the hamiltonian (42) only $-H_{QY}/2$ can remain and

$$L_r = \left[\ldots, -\omega_1 I_Y - \frac{1}{2}H_{QY}\right]. \quad (44)$$

By inserting Eq. (44) into Eq. (13) we obtain Eq. (37), whence, according to Eq. (38), we have Eq. (40). Thus, for the quadrupolar nuclei with $I \geq 1$ the FID after the hard RF pulse with the finite width at $\omega_1\tau = 90°$ has its origin, so as in the case of the multispin dipolar system, at the middle of the RF pulse.

4. Conclusions

We have calculated the shape of the FID for the case of any ratio of the local magnetic field to the amplitude of the RF pulse. The obtained Eq. (18) for the shape of the FID is valid for the multispin systems with the dipolar and quadrupolar interactions. The most important result of our consideration is that for the multispin dipolar system and for the quadrupolar nuclei with $I \geq 1$, the FID after the hard 90° RF pulse with the finite amplitude and duration has its origin at the center of RF pulse.

Appendix A

In this appendix we present a derivation of Eq. (39). It follows from Eq. (9) that at $t = t_1 + t_2$

$$G_0(t_1 + t_2) = \frac{\langle I_X | \exp(i t_1 L_0^+) \cdot \exp(i t_2 L_0) | I_X \rangle}{\langle I_X | I_X \rangle}. \quad (A1)$$

Insertion of Eq. (4) into Eq. (A1) yields

$$G_0(t_1 + t_2) = \sum_{n,m} \frac{\langle n | m \rangle}{\langle 0 | 0 \rangle} G_n^+(t_1) G_m(t_2). \quad (A2)$$

It follows from Eq. (7) that

$$G_n^+(t) = G_n(t) \quad (A3)$$

and

$$G_{2n}(t) = G_{2n}(-t), \quad G_{2n+1}(t) = -G_{2n+1}(-t). \quad (A4)$$

Using Eq. (A3) and Eq. (6) from Eq. (A2) we obtain

$$G_0(t_1 + t_2) = G_0(t_1)G_0(t_2) + \nu_0^2 G_1(t_1)G_1(t_2) + \nu_0^2\nu_1^2 G_2(t_1)G_2(t_2) + \nu_0^2\nu_1^2\nu_2^2 G_3(t_1)G_3(t_2) + \ldots \quad (A5)$$

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