Multi-dimensional dynamical decoupling based quantum sensing

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Nuclear magnetic resonance (NMR) has enormous applications [1-3]. Multi-dimensional NMR has been an essential technique to characterize correlations between nuclei and hence molecule structures [4]. Multi-dimensional spectroscopy has also been extended to optics to study correlations in molecules [5] and many-body effects in semiconductors [6, 7]. Towards the ultimate goal of single-molecule NMR, dynamical decoupling (DD) enhanced diamond quantum sensing [8, 9] has enabled detection of single nuclear spins [10-14] and nanoscale NMR [15, 16]. However, there is still lack of a standard method in DD-based quantum sensing to characterize correlations between nuclear spins in single molecules. Here we present a scheme of multi-dimensional DD-based quantum sensing, as a universal method for correlation spectroscopy of single molecules. We design multi-dimensional DD sequences composed of multiple sets of periodic DD sequences with different periods, which can be independently set to match different transition frequencies for resonant DD. We find that under resonant DD the sensor coherence patterns, as functions of multiple DD pulse numbers, can differentiate different types of correlations between nuclear spin transitions. This work offers a standard approach to correlation spectroscopy for single-molecule NMR.
Nuclear magnetic resonance (NMR) has important applications in analytical chemistry, structural biology and quantum computing [1]. Furthermore, multi-dimensional NMR [4], which employs composite control pulses and sequences on nuclear spins, provides information about the couplings between the nuclear spins and hence the molecule structures. However, the conventional NMR is generally performed for large ensembles of molecules (>10^{12} molecules) and at high magnetic fields (>1 Tesla). The top challenge of magnetic spectroscopy is NMR with atomic scale sensitivity and resolution [2].

A breakthrough toward single-spin NMR is dynamical decoupling (DD) based quantum sensing [8]. The basic principle is as follows. The quantum sensor (such as the electron spin of a nitrogen-vacancy (NV) center in diamond [17]) loses its coherence due to noises from weakly coupled nuclear spins (targets). The DD control (Fig. 1a) flips the sensor state periodically so as to cancel the effect of the background noises. The noises from the target nuclear spins have characteristic frequencies corresponding to their transitions. If the DD period matches the characteristic frequency, the noise from the target nuclear spins is resonantly amplified and the sensor coherence presents fingerprint sharp dips. Using this DD-based scheme, several groups have successfully detected single ^{13}\text{C} nuclear spins [10-12] and ^{13}\text{C} clusters in diamond [14]. Shallow NV centers near diamond surfaces have been used to sense NMR of single nuclear spins [13], nano-scale NMR of molecules [15, 16] and single protein molecules [18]. This DD-based quantum sensing, however, has important limitations: (i) it does not resolve nuclear spins of the same species; (ii) it does not distinguish nuclear spin clusters of different correlation types if the clusters produce the same noise spectrum. It is highly desirable to have a correlation spectroscopy in quantum sensing similar to that in
multi-dimensional NMR [19]. Recently we have found that for DD resonant with a specific target spin transition, the sensor coherence dip oscillates periodically as a function of DD pulse number, with the oscillation period inversely proportional to the coupling strength between the sensor spin and target spin [20]. Based on this finding, we proposed a scheme that is capable of resolving single nuclear spins of the same species and identifying correlations in nuclear spin clusters [20]. However, that scheme cannot fully differentiate different types of correlations in nuclear spin clusters.

In this work, we propose the concept of multi-dimensional DD-based quantum sensing. We demonstrate that the multi-dimensional quantum sensing is capable of fully characterizing the correlations between different transitions in single nuclear spin clusters. The multi-dimensional DD sequence is composed of multiple sets of periodic DD sub-sequences with different pulse intervals. When the pulse intervals match different nuclear spin transitions (resonant DD), the sensor coherence, as a function of the pulse numbers of the DD sub-sequences, shows distinct patterns depending on the correlation types of different nuclear spin transitions.

**Theoretical model**

We consider a spin-1/2 quantum sensor \( S = 1/2 \) weakly coupled to \( M \) target nuclear spin clusters (each representing a "molecule"). The general Hamiltonian is

\[
H = S_z \sum_{k=1}^{M} \beta^{(k)} + \sum_{k=1}^{M} H_0^{(k)},
\]

where \( H_0^{(k)} = \sum_{m=1}^{d_k} \epsilon_m^{(k)} |m\rangle_{kk} \langle m| \) is the nuclear spin Hamiltonian for the \( k \)-th "molecule" with \( d_k \) denoting the number of eigenstates \( \{|m\rangle_{kk}\} \),
\[
\beta^{(k)} = \frac{1}{2} \sum_{m,n} \left( \beta^{(k)}_{mn} |m\rangle_k \langle n| + \text{H.c.} \right) \text{ is the noise operator from the } k\text{-th "molecule"}
\]
which induces the nuclear spin transition \( |m\rangle_k \leftrightarrow |n\rangle_k \) with the transition frequency
\[
\omega^{(k)}_{mn} = \epsilon^{(k)}_m - \epsilon^{(k)}_n \quad \text{and the transition matrix element } \beta^{(k)}_{mn} = \langle m | \beta^{(k)} | n \rangle_k.
\]
In quantum sensing, the coupling to the target spins is weak, i.e. \( |\beta^{(k)}_{mn}| \ll |\epsilon^{(k)}_{mn}| \) for \( m \neq n \). We denote the eigenstates of the sensor operator \( S_z \) as \( |\pm\rangle \).

To suppress the background noise and selectively enhance the noise from the target spins [10], we apply DD [8, 9] to the sensor (consisting of a sequence of \( \pi \)-flips at times \( \{t_1, t_2, \ldots, t_N\} \) for the evolution from 0 to \( t \)). Under DD control, the target spin evolution conditioned on the sensor states \( |\pm\rangle \) is
\[
U^{(\pm)}_{k,N}(t) = \bigotimes_{k} U^{(\pm)}_{k,N}(t) \quad \text{with}
\]
\[
U^{(\pm)}_{k,N}(t) = e^{-i[\mathbf{H}^{(\pm)}_{k,N} (\mathbf{H}_{k}^{(\pm)} / 2 \mathbf{N}^{(\pm)}_{k,N} t)]} \ldots e^{-i[\mathbf{H}^{(\pm)}_{k,N} (\mathbf{H}_{k}^{(\pm)} / 2 \mathbf{N}^{(\pm)}_{k,N} t)]} e^{-i[\mathbf{H}^{(\pm)}_{k,N} (\mathbf{H}_{k}^{(\pm)} / 2 \mathbf{N}^{(\pm)}_{k,N} t)]}.
\]
The sensor spin decoherence is caused by the sensor-target entanglement,
\[
L(t) = \prod_{k=1}^{M} \text{Tr}_k \left[ \rho_k \left( U^{(-)}_{k,N} \right)^\dagger U^{(+)}_{k,N} \right],
\]
where the density matrix of the \( k \)-th target is assumed in a maximally mixed state
\[
\rho_k = d^{-1} \sum_{m} |m\rangle_k \langle m| \quad \text{as the temperature is usually much higher than the nuclear spin transition frequencies. In the following, we will omit the subscript } k \text{ when we consider only one target "molecule".}
\]

**One-dimensional DD-based quantum sensing**

One-dimensional DD sequence is just the conventional \( N_1 \)-pulse
Carr-Purcell-Meiboom-Gill (CPMG- \(N_i\)) sequence [21, 22]. The pulse interval is \(2\tau_i\) and \(t_p = (2 p_i - 1)\tau_i\) with \(p_i = 1, 2, \cdots, N_i\), as shown in Fig. 1a. The sensor coherence presents dips when then pulse interval matches the frequency of a transition \(|m\rangle \leftrightarrow |n\rangle\), i.e., \(2\tau_1 = \pi (2c - 1)/\omega_{mn}\) with \(c = 1, 2, \cdots\), as shown in Fig. 1d. In the following we always consider the first-order coherence dip \((c = 1)\). Under the resonant DD, the sensor spin coherence dip periodically oscillates as a function of the CPMG pulse number \(N_i\) [20], namely,

\[
L^{dp}(N_i) \approx \frac{1}{d} \text{Tr}[U_{mn}(2N_i)] = \frac{1}{d} \left[ d - 2 + 2 \cos(2N_i\delta_1) \right],
\]

where \(\delta_1 = |\beta_{mn}/\omega_{mn}|\) and

\[
U_{mn}(N_i) = \exp[-iN_i (\beta_{mn} |m\rangle\langle n| + \text{H.c.})/\omega_{mn}].
\]

The sensor coherence has quantized minima determined by the Hilbert space dimension \(d\) of the target nuclear spin cluster \(\min \left(L^{dp}(N_i)\right) = (d - 4)/d\) at \(N_i = \pi/(2\delta_1)\). The quantized minima can be understood from the probability for the target spin cluster remaining in its initial state after the evolution. If the target spin cluster is initially in the state \(|m\rangle\) or \(|n\rangle\), the DD control on the sensor drives the target spin cluster to periodically oscillate between \(|m\rangle\) and \(|n\rangle\), while if the target spin cluster is initially in other states, the DD control would keep the target spin states unchanged. Thus the depth of the sensor coherence oscillation is \(-2/d + (d - 2)/d = (d - 4)/d\).

Two-dimensional DD-based quantum sensing
The two-dimensional DD sequence contains two sets of CPMG sequences with pulse intervals $2\tau_1, 2\tau_2$ and pulse numbers $N_1, N_2$, respectively (Fig. 1b). The sensor spin is flipped at time $t_{p_i} = (2p_i - 1)\tau_1$, $t_{N_i+p_2} = 2N_i\tau_1 + (2p_2 - 1)\tau_2$ with $p_i = 1, 2, \ldots, N_i$ ($i = 1, 2$). The sensor coherence shows sharp dips when the two pulse intervals are matched to two different target spin transition frequencies, i.e. $2\tau_i = \pi/\omega_i$, as shown in Fig. 1e. Then the sensor coherence as a function of $N_1$ and $N_2$ contains information about the correlation between the two different transitions.

We first consider the case that the sensor spin is weakly coupled with two independent target nuclear spin “molecules” ($M = 2$). We choose a transition $|m\rangle_1 \leftrightarrow |n\rangle_1$ from target 1 and another transition $|p\rangle_2 \leftrightarrow |q\rangle_2$ from target 2. Under the double-resonance condition ($2\tau_1 = \pi/\omega_{mn}^{(i)}$, $2\tau_2 = \pi/\omega_{pq}^{(i)}$), the CPMG-$N_1$ sub-sequence resonantly amplifies the noise from the transition $|m\rangle_1 \leftrightarrow |n\rangle_1$ in target 1, and the CPMG-$N_2$ sub-sequence resonantly amplifies the noise from the transition $|p\rangle_2 \leftrightarrow |q\rangle_2$ in target 2. Then the sensor spin coherence as a function of the pulse numbers $N_1$ and $N_2$ is (see Supplementary Information)

$$L^\text{dp}_{i,N_1,N_2} (N_1, N_2) \approx \frac{1}{d_1d_2} \text{Tr} \left[ U_{1,mm} (2N_1) \right] \text{Tr} \left[ U_{2,pq} (2N_2) \right]$$

$$= \frac{1}{d_1d_2} \left[ d_1 - 2 + 2\cos(2N_1\delta_1) \right] \left[ d_2 - 2 + 2\cos(2N_2\delta_2) \right],$$

(6)

where $\delta_j = |\beta_{mn}^{(j)}/\omega_{mn}^{(j)}|$. As shown in Fig. 2, the sensor coherence as a function of the pulse numbers presents periodic patterns with the unit cell $N_{1c} \times N_{2c}$ ($N_{ic} = \pi/\delta_i$).
Now we consider the two different transitions $|m\rangle \leftrightarrow |n\rangle$ and $|p\rangle \leftrightarrow |q\rangle$ in the same target “molecule”. Under the double-resonance condition

\[(2\tau_1 = \pi / \omega_{mn}, 2\tau_2 = \pi / \omega_{pq}),\]

the sensor coherence is (see Supplementary Information)

\[L_{\text{dip}}^{\text{uncorr}}(N_1, N_2) \approx \frac{1}{d} \text{Tr}[U_{pq}(2N_2)U_{mn}(2N_1)],\]

(7)

where $U_{mn}(2N_1)$ and $U_{pq}(2N_2)$ are defined in Eq. (5). When the two transitions are uncorrelated (they do not share a state), the sensor spin coherence (see Supplementary Information) is

\[L_{\text{dip}}^{\text{uncorr}}(N_1, N_2) \approx \frac{1}{d}[d - 4 + 2 \cos(2N_1\delta_1) + 2 \cos(2N_2\delta_2)].\]

(8)

where $\delta_1 = |\beta_{mn} / \omega_{mn}|$ and $\delta_2 = |\beta_{pq} / \omega_{pq}|$. When the two transitions share one state (correlated), the sensor coherence dip is

\[L_{\text{dip}}^{\text{corr}}(N_1, N_2) \approx \frac{1}{d}[d - 3 + \cos(2N_1\delta_1) + \cos(2N_2\delta_2) + \cos(2N_1\delta_1) \cos(2N_2\delta_2)].\]

(9)

For both correlated and uncorrelated transitions the sensor coherence dip oscillates periodically in the two-dimensional space $(N_1, N_2)$ with the unit cell $N_{1c} \times N_{2c}$ ($N_{ic} = \pi / \delta_{N}$). But the sensor coherence minima are different for uncorrelated and correlated transitions, being

\[\min(L_{\text{dip}}^{\text{uncorr}}(N_1, N_2)) = (d - 8)/d, \quad d \geq 4,\]

(10)

\[\min(L_{\text{dip}}^{\text{corr}}(N_1, N_2)) = (d - 4)/d, \quad d \geq 3.\]

(11)

Figure 3 shows the two-dimensional sensor coherence as a function of $N_1, N_2$ for
three different cases, i.e. type-V transition \((d=3)\), correlated and uncorrelated transitions in ladder type transitions \((d=4)\). The two-dimensional sensor coherence presents different patterns for the correlated and uncorrelated transitions in the same target spin cluster. For correlated transitions in different target spin clusters, the sensor coherence shows similar patterns but with different coherence minima.

**Three-dimensional DD-based quantum sensing**

Higher dimensional sensing can be introduced to differentiate further different types of correlations in the nuclear spin clusters. We demonstrate this idea with the three-dimensional DD-based quantum sensing. The three-dimensional sequence contains three sets of CPMG sequences with pulse intervals \(2\tau_1, 2\tau_2, 2\tau_3\) and pulse numbers \(N_1, N_2, N_3\) (Fig. 1c). The sensor spin is flipped at time \(t_p = (2p_i - 1)\tau_1\), \(t_{N_i+N_j+p_p} = 2N_i\tau_1 + 2N_j\tau_2 + (2p_j - 1)\tau_3\) with \(p_i = 1, 2, \cdots, N_i\) \((i=1,2,3)\). When the three pulse intervals match the frequencies of three different target spin transitions \(2\tau_i = \pi/\omega_i\), the sensor coherence as a function of \(N_1, N_2\) and \(N_3\) directly reveals the multiple correlations between the transitions.

For example, we consider the case that the sensor spin is coupled with three independent target “molecules” \((M=3)\) and the DD is resonant with three transitions \(|m\rangle_1 \leftrightarrow |n\rangle_1, \ |p\rangle_2 \leftrightarrow |q\rangle_2, \ |r\rangle_3 \leftrightarrow |s\rangle_3\) from target 1,2,3 (with transition frequencies \(\omega_{mn}^{(1)}, \omega_{pq}^{(2)}, \omega_{rs}^{(3)}\) and transition matrix elements \(\beta_{mn}^{(1)}, \beta_{pq}^{(2)}, \beta_{rs}^{(3)}\) in turn). Under the resonance condition, the sensor spin coherence as a function of the pulse numbers \(N_1, N_2, N_3\) is (see Supplementary Information)
\[ L^\text{dp}_{1,2,3}(N_1, N_2, N_3) \approx \frac{1}{d_1d_2d_3} \left[ d_1 - 2 + 2 \cos(2N_1\delta_1) \right] \left[ d_2 - 2 + 2 \cos(2N_2\delta_2) \right] \times \left[ d_3 - 2 + 2 \cos(2N_3\delta_3) \right], \] (12)

where \( \delta_1 = \left| \beta_{mn}^{(1)} / \omega_{mn}^{(1)} \right|, \delta_2 = \left| \beta_{pq}^{(2)} / \omega_{pq}^{(2)} \right|, \) and \( \delta_3 = \left| \beta_{rs}^{(3)} / \omega_{rs}^{(3)} \right|. \)

For three transitions in the same target "molecule" \( |m\rangle \leftrightarrow |n\rangle, \ |p\rangle \leftrightarrow |q\rangle, \) and \( |r\rangle \leftrightarrow |s\rangle \) the sensor coherence under the resonant DD condition \((2\tau_1 = \pi / \omega_{mn}, 2\tau_2 = \pi / \omega_{pq}, 2\tau_3 = \pi / \omega_{rs})\) is (see Supplementary Information)

\[ L^\text{dp}(N_1, N_2, N_3) \approx \frac{1}{d} \text{Tr} \left[ U_{rs} (2N_3) U_{pq} (N_2) U_{mn} (2N_1) U_{pq} (N_2) \right]. \] (13)

The sensor coherence as a function of \( N_1, N_2, N_3 \) has different forms depending on the specific correlation types of the three transitions.

(a) **Uncorrelated transitions** - the three transitions share no common state \((d \geq 6)\),

\[ L^\text{dp}_\text{in} (N_1, N_2, N_3) \approx \frac{1}{d} \left[ d - 6 + 2 \cos(2N_1\delta_1) + 2 \cos(2N_2\delta_2) + 2 \cos(2N_3\delta_3) \right]. \] (14)

(b) **Ring-type correlation** - the three transitions are among three states \((d \geq 3\) and type-\( \Delta \) in Fig. 4a),

\[ L^\text{dp}_\Delta (N_1, N_2, N_3) \approx \frac{1}{d} \left[ d - 3 + \cos(2N_1\delta_1) \cos(2N_2\delta_2) \sin^2(N_2\delta_2) + \cos(2N_1\delta_1) \cos^2(N_2\delta_2) + \cos^2(N_2\delta_2) \cos(2N_3\delta_3) \right. \]
\[ + \cos(2N_1\delta_1) \sin^2(N_2\delta_2) \cos(2N_3\delta_3) \]
\[ \left. - \cos(2N_1\delta_1) \sin^2(N_2\delta_2) \cos(2N_3\delta_3) \right]. \] (15)

(c) **Star-type correlation** - the three transitions share one state \((d \geq 4)\). The sensor coherence has the same form as that for the ring-type correlation.
(d) **Linked ladder-type correlations** - the three transitions form a linked ladder \((d \geq 4)\) and ladder type \((\text{linked})\) in Fig. 4a,

\[
L_{\text{ladder(linked)}}(N_1, N_2, N_3) \approx \frac{1}{d} \left[ d - 4 + \cos(2N_1\delta_1) - \sin^2(N_2\delta_2) + \cos(2N_3\delta_3) \\
+ \cos(2N_1\delta_1)\cos^2(N_2\delta_2) + \cos^2(N_2\delta_2)\cos(2N_3\delta_3) \\
- \cos(2N_1\delta_1)\sin^2(N_2\delta_2)\cos(2N_3\delta_3) \right]. \tag{16}
\]

(e) **Unlinked ladder-type correlations** - the three transitions form an unlinked ladder type transition \((d \geq 5)\) and ladder type \((\text{unlinked})\) in Fig. 4a,

\[
L_{\text{ladder(unlinked)}}(N_1, N_2, N_3) \approx \frac{1}{d} \left[ d - 4 + 2\cos(2N_1\delta_1) + \cos(2N_2\delta_2) + \cos(2N_3\delta_3) \\
+ \cos(2N_2\delta_2)\cos(2N_3\delta_3) \right]. \tag{17}
\]

The sensor coherence for both correlated and uncorrelated transitions is periodic with the unit cell \(N_{1c} \times N_{2c} \times N_{3c}\) \((N_{1c} = \pi/\delta_1)\) in the three-dimensional space \((N_1, N_2, N_3)\). Note that the sensor coherence remains unchanged if one exchanges \(N_1, \delta_1\) and \(N_3, \delta_3\) due to the symmetry in Eq. (13). In Fig. 4b we show the sensor coherence as a function of \(N_1, N_2\) while keeping \(N_3\) constant for three types of correlations (b)(d)(e), and find that the coherence patterns are different for different correlation types.

Even higher-dimensional DD sequences can be constructed for correlation sensing. It should be pointed out that the correlations between different transitions can already be fully determined by repeatedly applying the two-dimensional DD sensing to different pairs of transitions.
References


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**Author Contributions** R.B.L. conceived the idea and designed the project, R.B.L and W.L.M. designed the pulse sequences, W.L.M. formulated the theory and carried out the numerical calculations, W.L.M. and R.B.L. wrote the paper.

**Author Information** The authors declare no competing financial interests. Correspondence and requests for materials should be addressed to R.B.L. (rbliu@phy.cuhk.edu.hk).
Figure 1 | Multi-dimensional DD sequences. a, One-dimensional DD sequence. b, Two-dimensional DD sequence. c, Three-dimensional DD sequence. Here the $l$-dimensional DD sequence contains $l$ CPMG sub-sequences with different pulse intervals $2\tau_i$ and pulse numbers $N_i$. The $l$-dimensional DD is resonant with $l$ different transitions when the pulse intervals are such that $2\tau_i = \pi/\omega_i$ for $i = 1, 2, \ldots, l$.

d, Sensor coherence as a function of the pulse interval $\tau_1$ under one-dimensional DD control with $N_1 = 20$. The spin-1/2 sensor is weakly coupled to a single target spin-1

$\mathbf{J}$ via the coupling Hamiltonian $H = \lambda S_z J_z + \left(\omega_a + \omega_b\right)J_z^2/2 + \left(\omega_a - \omega_b\right)J_z/2$ with $\lambda = 5$ kHz and the two transition frequencies $\omega_a = 0.20$ MHz and $\omega_b = 0.14$ MHz (see inset). The sensor coherence shows sharp dips when the pulse interval matches the nuclear spin transition frequencies $\tau_{a/b} = \pi/(2\omega_{a/b})$. e, Sensor coherence as a function of two pulse intervals $\tau_1, \tau_2$ under two-dimensional DD control with $N_1 = N_2 = 20$. Here the sensor is coupled to the same target spin as in d.
Figure 2 | Two-dimensional quantum sensing of two independent nuclear spin clusters. 

a, Schematic illustration of the sensor coupled to two target “molecules” and different types of transitions from the two targets. Type-II transitions represent two independent target spin-1/2’s, type-IV transitions represent a target spin-1/2 and a target spin-1, and type-VV transitions represent two independent target spin-1’s. The solid (dashed) arrows are nuclear spin transitions resonant (unresonant) with the DD. 

b, Sensor coherence dip as a function of two free CPMG pulse numbers $N_1$ and $N_2$. The parameters are such that $\omega_1 = 0.20$ MHz, $\omega_2 = 0.14$ MHz, $\delta_1 = 0.025$, and $\delta_2 = 0.036$. 
Figure 3 | Two-dimensional quantum sensing of correlations in a single nuclear spin cluster. **a**, Schematic illustration of different correlation types of two transitions in a single nuclear spin cluster. Type-II transitions represent two correlated transitions in a spin-1 cluster, ladder type (**corre**) transitions represent two correlated transitions in a spin-3/2 cluster, and ladder type (**uncorre**) transitions represent two uncorrelated transitions in a spin-3/2 cluster. The solid (dashed) arrows are nuclear spin transitions resonant (unresonant) with the DD. **b**, Sensor coherence dip as a joint function of two free CPMG pulse numbers $N_1$ and $N_2$. The parameters are such that $\omega_1 = 0.20$ MHz, $\omega_2 = 0.14$ MHz, $\delta_1 = 0.025$, and $\delta_2 = 0.036$. 
**Figure 4 | Three-dimensional correlation spectroscopy of a single nuclear spin cluster.**

**a,** Schematic illustration of different correlation types of three transitions from a single nuclear spin cluster. Type-Δ transitions represent three correlated transitions in a spin-1 cluster, ladder type (linked) transitions represent three correlated transitions in a spin-3/2 cluster, and ladder type (unlinked) transitions represent one uncorrelated transition and two correlated transitions in a spin-3/2 cluster. **b,** Sensor coherence dip as a function of two CPMG pulse numbers $N_1, N_2$ with $N_3 = 12$. The parameters are such that $\omega_1 = 0.20$ MHz, $\omega_2 = 0.14$ MHz, $\omega_3 = 0.06$ MHz, $\delta_1 = 0.025$, $\delta_2 = 0.036$, and $\delta_3 = 0.083$. 