Fading entanglement near equilibrium state

Gregory B. Furman, Victor M. Meerovich, and Vladimir L. Sokolovsky

Physics Department, Ben Gurion University of the Negev, Beer Sheva, 84105 Israel

Abstract

It was recently shown that entanglement in quantum systems being in a non-equilibrium state can appear at much higher temperatures than in an equilibrium state. However, any system is subject to the natural relaxation process establishing equilibrium. The work deals with the numerical study of entanglement dynamics in a dipolar coupled spin-1/2 system under the transition from a non-equilibrium state to an equilibrium state. The spin system is characterized by a two-temperature density matrix, and the process of the establishment of equilibrium is in the equalization of these temperatures. The method of the non-equilibrium statistical operator is used to describe the evolution of the system. The process of establishing an equilibrium state in the homonuclear spin systems at low temperature was first considered. It was shown that the time dependences of the inverse temperatures of the spin subsystems are given by a solution of non-linear equations in contrast to the linear equations in the well-known high temperature approximation. It was first studied the entanglement dynamics during the equilibrium state establishment and found that, during establishing equilibrium, the concurrence changes non-monotonically with time and temperatures. Entanglement fades long before equilibrium is established in the system. It was shown that the entanglement dynamics depends strongly on the ratio of the Zeeman energy to the dipolar energy. At a high ratio, the concurrence in the system decreases quickly for time about 100 $\mu$s while, at a low ratio, establishment of equilibrium and fading entanglement take prolonged time up to 1 ms.
I. INTRODUCTION

Entanglement is a term used in quantum theory to describe the fact that the particles can be correlated with each other regardless of how far they are from each other [1–3]. Many current studies [1–3] focus on how to harness the potential of entanglement in the development of quantum cryptography, quantum communication [4], quantum computation [5], and quantum metrology [6, 7]. The growth in interest in entanglement applications has stimulated intensive qualitative and quantitative research of entanglement in various physical systems and in both equilibrium [2, 3, 8–12] and non-equilibrium [13–17, 22], including the pseudo-pure state [18–20] as an initial state [21].

It was shown that quantum systems in thermal equilibrium became entangled, if the interaction energy between them is larger than thermal energy due to their coupling to the environment. This condition leads to that entanglement appears only at very low temperature.

For example, entanglement in dipolar coupled spin system in equilibrium state is achieved by the application of a low external magnetic field in which the Zeeman interaction energy is the order of or even less than the dipolar interaction energy, \( \omega_0 \leq \omega_d \) [10, 11]. For a proton spin systems, the dipolar energy is \( \omega_d = 15 \text{ kHz} \). That results in the temperature of the entanglement appearance of \( T \sim \frac{\omega_d}{k_B} = 0.7\mu K \) [9–12]. Here \( k_B \) is the Boltzmann constant.

Recently, it was proved that this relationship between temperature and entanglement is not justified for systems that are not in thermal equilibrium [23]. In contrast to the equilibrium cases, as it was shown that entanglement in quantum systems being in a non-equilibrium state can appear at much higher temperatures [23, 24]. Investigations of spin systems demonstrate this statement: e.g. a spin system, which is in a separated state at equilibrium, is irradiated by a radiofrequency field and goes out from the equilibrium, and entanglement appears in the system simultaneously [13–17, 22].

The present paper deals with the opposite case when the system being initially in a non-equilibrium state evolves to an equilibrium state. This entanglement dynamics during the natural process of establishing equilibrium is an actual problem which, according to our knowledge, does not studied yet. We study entanglement dynamics in a dipolar coupled spin-1/2 system under the transition from non-equilibrium state to equilibrium state. We consider a spin system, which can be divided into two thermodynamic subsystems which are
in the equilibrium state and characterized by two different temperatures (so-called a quasi-equilibrium state [25, 26]). The process of establishing equilibrium in the whole system can be represented as equalization of their temperatures. Such quasi-equilibrium states were prepared experimentally using various methods [25–27]. These states can be considered as the last stage in relaxation process from non-equilibrium state to equilibrium one. Existence of such states can be explained by using the fundamental principles of non-equilibrium statistical mechanics first formulated by Bogolyubov [28]. In non-equilibrium statistical mechanics, a fundamental role is played hierarchy of the relaxation times and reducing in the number of parameters requested for the description of a non-equilibrium process. As shown by Bogolyubov, after the lapse of a small interval of time, of the order of the duration of a collision, all higher distribution functions are completely determined by a single-particle function, and kinetic equations can be found for this stage. This means it is possible to use an abbreviated description of a non-equilibrium state in which the complete distribution function depends on few parameters.

Bogolyubov’s idea about hierarchy of the relaxation times has been widely used for description of relaxation processes in spin systems [25, 29–31]. In particular, if the correlation times are much shorter than the time $T_2$, the lifetime of the free precession signal, and the time $T_2$ is shorter then the characteristic time of an investigated process, then the equilibrium state is established in parts of the spin system before the whole system reaches the total equilibrium.

The structure of the paper is as follows: in the next section, we describe the Hamiltonian for a spin system in an external field at the equilibrium and quasi-equilibrium states and principale of the Provotorov saturation theory [25, 31]. According to this theory in a dipolar coupled spin system in a quasi-equilibrium state, two spin temperatures $T_z$ and $T_d$ can be assigned to the Zeeman and dipolar thermodynamic subsystems [25, 31]. In section 3 establishment of the equilibrium state is analyzed. Then we consider dynamics of the pairwise entanglement in a quasi-equilibrium state which is characterized by these two temperatures. Discussion of the results is given in the final section. Numerical calculations of evolution of the spin temperature and concurrence $C_{mn}$ between the $m$-th and $n$-th spins are performed using the software based on the MatLab package. To demonstrate dependence of the entanglement dynamics on the relative spin position we will consider three four-spin systems: linear chain, ring, and rectangle. The choice of these systems is due to the following reasons:
i) these structures exist in real molecules and used in experiments and ii) as dipole-dipole interactions decrease inversely to the third power of the distance between the spins, the effect of the system geometry is manifested more clearly at a small number of spins.

II. SPIN SYSTEM IN QUASI-EQUILIBRIUM STATE

We consider the system of identical spin-1/2 particles with the dipole-dipole interaction (DDI) in an external magnetic field $\vec{H}_0$ along the $z$-axis. In the equilibrium state the system is characterized by a density matrix

$$\rho = \frac{\exp(-\beta H)}{Z(\beta)}, \quad (1)$$

with a single parameter $\beta = \frac{1}{k_B T}$ denotes the inverse spin temperature, $T$ is the spin temperature, and $Z(\beta) = Tr\{\exp(-\beta H)\}$ is the partition function. The Hamiltonian consists of the two parts, $H = H_Z + H_{dd}$, the Zeeman part

$$H_z = -\gamma \vec{H}_0 \sum_j \vec{I}_j \quad (2)$$

and the dipolar part

$$H_{dd} = -\gamma^2 \hbar \sum_{jk} \left( \frac{3}{r_{jk}^5} (\vec{I}_j \vec{r}_{jk}) (\vec{I}_k \vec{r}_{jk}) - \frac{1}{r_{jk}^3} \vec{I}_j \vec{I}_k \right). \quad (3)$$

Here $\gamma$ is the gyromagnetic ratio, $\vec{I}_j$ is the angular momentum operator of the $j$-th spin ($j = 1, 2, \ldots N$), $\vec{r}_{jk}$ is the radius vector from the $j$-th to $k$-th spins with the spherical coordinates $r_{jk}, \theta_{jk}$ and $\varphi_{jk}$. $\theta_{jk}$ is the angle between the radius vector $\vec{r}_{jk}$ and direction of the external magnetic field.

In low magnetic field, when the Zeeman energy splitting $\omega_0 = \gamma H_0$, and DDI energy $\omega_d = \sqrt{\frac{Tr(H_{dd}^2)}{Tr(\sum_j I_{zj})^2}}$ are commensurable quantities, the time for reaching equilibrium state describing by Eq.(1) is determined by $\omega_d^{-1}$ which is of the order of $T_2 \quad (26)$.

In high magnetic field, $\omega_0 >> \omega_d$, the time $t$ for reaching equilibrium becomes very long, $t >> T_2$, due to the fact that the energy spectra of the Zeeman energy and the DDI energy are very different \[26\].

We will consider the spin system placed in high magnetic field as two thermodynamic subsystems, the Zeeman and dipole-dipole ones, each of them is at thermodynamic equi-
librium with own spin temperature and between which there is an energy transfer. This approach is based on the following reasoning.

The total Hamiltonian can be presented as the sum of two terms each commuting with other, \( H_z \) and \( H_d \) ([\( H_z, H_d \) = 0]), and the term \( H_{nd} = H_{dd} - H_d \), which does not commute with both \( H_z \) and \( H_d \). \( H_d \) is the secular part of dipolar Hamiltonian (3)

\[
H_d = -\gamma^2 \hbar \sum_{jk} r_{jk}^{-3} P_2(\cos \theta_{jk}) \left( 3I_{zj}I_{zk} - \vec{I}_j \cdot \vec{I}_k \right),
\]

where \( P_2(\cos \theta_{jk}) = \frac{1}{2}(1 - 3 \cos^2 \theta_{jk}) \), \( I_{\alpha j} \) is the projection of the angular momentum operator of the \( j \)-th spin \( (j = 1, 2, \ldots, N) \) on the \( \alpha \)-axis (\( \alpha = x, y, z \)). At \( \omega_0 >> \omega_d \) the effect of the dipolar interaction is small in comparison with the Zeeman interaction and, in the first-order perturbation, the secular part \( H_d \) (4) plays the main role in establishing equilibrium in each subsystem. This process is characterized by flip-flop terms in the secular part \( H_d \), that ensures the density matrix in the following approximate form

\[
\rho = \frac{\exp(-\beta_z H_z - \beta_d H_d)}{Z(\beta_z, \beta_d)}.
\]

where \( \beta_z = \frac{1}{k_B T_z} \) and \( \beta_d = \frac{1}{k_B T_d} \) are the inverse Zeeman and dipolar temperatures, respectively, \( Z(\beta_z, \beta_d) = Tr \{\exp(-\beta_z H_z - \beta_d H_d)\} \) is the partition function that encodes the statistical properties of the system in the quasi-equilibrium state. Separation of the spin system into the Zeeman and dipolar subsystems is consequence of the fact that the Zeeman and dipolar energies

\[
\langle H_z \rangle = \omega_0 \langle I_z \rangle = -\frac{\partial \ln Z(\beta_z, \beta_d)}{\partial \beta_z}
\]

and dipolar energies

\[
\langle H_d \rangle = -\frac{\partial \ln Z(\beta_z, \beta_d)}{\partial \beta_d}
\]

are separately constants of the motion, while both \( \langle H_z \rangle \) and \( \langle H_d \rangle \) depend on both thermodynamics parameters, \( \beta_z \) and \( \beta_d \) [26]. Results (5-7) have been conformed by experiments [26, 27].

The time for reaching the quasi-equilibrium state described by Eq. (5) is of the order of \( T_2 \). It is assumed that further relaxation process takes more time \( t > T_2 \). This process consists in equalization of the temperature \( \beta_z^{-1} \) and \( \beta_d^{-1} \) and results in establishing of the equilibrium state which is characterized by density matrix (1) with \( \beta_z = \beta_d = \beta \).

The approximate form of the density matrix does not include the non-secular term of the dipolar Hamiltonian, \( H_{nd} \). To take into account this term and to describe the relaxation at time \( t > T_2 \), we will use below the method of the non-equilibrium statistical operator [32].
III. ESTABLISHMENT OF EQUILIBRIUM

During the time $T_2$ the system loses the quantum coherence features and achieves a quasi-equilibrium state due to the flip-flop terms including in the Hamiltonian $H_d$. For the spin system at low temperature, the decoherence time $T_2$ typically ranges between nanoseconds and seconds [33]. A typical value of $T_2$ in a nuclear dipolar coupled spin system is $100$ $\mu$s [34] which is much shorter than the spin-lattice relaxation times of the Zeeman, $T_1z$ and dipolar, $T_1d$ energies. $T_1z$ and $T_1d$ take values in the range from minutes to hours [26]. We will study spin dynamics at $T_2 < t \ll \min\{T_1z, T_1d\}$ when each subsystem can be considered as being at thermal equilibrium, and the spin-lattice relaxation process can be ignored.

To derive an equation describing the establishment of equilibrium state (1), we take the density matrix in the general form [32]

$$\rho_{ne}(t) = Z_{ne}^{-1} \exp\left(-\sum_n \left(\beta_n(t) H_n - \beta_n(t) \int_{-\infty}^{0} d\tau e^{\varepsilon \tau} K_n(\tau)\right)\right), \ n = z, dd \quad (8)$$

where

$$Z_{ne}(t) = Tr \left\{ \exp\left(-\sum_n \left(\beta_n(t) H_n - \beta_n(t) \int_{-\infty}^{0} d\tau e^{\varepsilon \tau} K_n(\tau)\right)\right)\right\}, \quad (9)$$

$K_n(\tau)$ is the operator of the energy flux

$$K_n(\tau) = \frac{dH_n(\tau)}{d\tau} \quad (10)$$

and transition to the limit $\varepsilon \to 0$ should be made after the calculation of the integral. According to the method of the non-equilibrium statistical operator, the operators under the integral in Eq. (10) are taken in the Heisenberg representation to be $H_n(\tau) = e^{i\tau H} H_n e^{-i\tau H}$ [32].

Accounting that $\sum_n K_n(\tau) = 0$, we rewrite density matrix (8) in the following form [32]

$$\rho_{ne} = Z_{ne}^{-1} \exp\left(-\sum_k \beta_k(t) H_k - (\beta_d(t) - \beta_z(t)) \int_{-\infty}^{0} d\tau e^{\varepsilon \tau} K(\tau)\right). \quad (11)$$

Evolution of the spin system can be sufficiently completely described by the density matrix (11) with temporally depending $\beta_z$ and $\beta_d$. To determine the temporal dependence of the inverse temperatures $\beta_z(t)$ and $\beta_d(t)$, let us calculate the average energy flux. This flux is given by averaging expression (10) using non-equilibrium density matrix (11)
\[ \langle K_n (\tau) \rangle = \left\langle \frac{dH_n (\tau)}{d\tau} \right\rangle, \]  
\[ (12) \]

where \( \langle ... \rangle = Tr (\rho_n ...). \) Eqs. (11) and (12) are valid at any temperatures of the sub-systems and can be applied to consider dynamics of the dipolar coupled spin system and generation of the entangled state in the system. The quantities in Eq. (12), averaged with density matrix (11), depend nonlinearly on the inverse temperatures \( \beta_z \) and \( \beta_d \) and their difference \( \beta_z - \beta_d. \) Eq. (12) is solved using different approximations: the high-temperature approximation, \( \frac{|H|}{k_B T} \ll 1; \) the approximation of small values of the temperature difference, \( |\beta_z - \beta_d| \ll \min \{\beta_z, \beta_d\} \) or/and the approximation of small energy flux, \( ||K|| \ll ||H_k|| \) (\( ||...|| \) denotes a norm of an operator)[32].

As it is shown below, the entangled states can be achieved in the cases when a difference \( (\beta_d - \beta_z) \) cannot be regarded as a small parameter. We cannot also use the high-temperature approximation and the expansion of (11) in powers of \( K. \) Hence, the perturbation theory methods cannot be applied effectively to study entanglement in the considered systems.

We will solve numerically Eq. (12) for four-spin systems with various geometry: chain, circle, and rectangle. Examples of such systems are quasi-one-dimensional fluorine chains in calcium fluorapatite \( Ca_5F (PO_4)_3 \) [39], xenon tetrafluoride with chemical formula \( XeF_4 \) (its crystalline square planar structure was determined by both NMR spectroscopy and by neutron diffraction studies [40, 41]), and 1,4-dichlorobenzene \( C_6H_4Cl \) with a rectangle planar structure.

We use dimensionless units where the dipolar coupling constant of the nearest spins is chosen to be \( D = 1. \) The time unit is determined by the DDI energy between these spins and equals \( 100 \mu s. \) The ratio of Zeeman energy splitting to the dipolar energy is chosen to be \( \frac{\omega_0}{\omega_d} = 43 \) for all considered cases. We assume also that the angles \( \theta_{j,k} = \frac{\pi}{2} \) for all pairs of spins in all cases. In our numerical calculation we used: a) for a chain all angles \( \varphi_{j,k} = \frac{\pi}{2} \) and the coupling constants of spins the \( j \)-th and \( k \)-th are \( D_{j,k} = (j - k)^{-3}; \) b) for a circle \( \varphi_{12} = \frac{\pi}{2}, \varphi_{23} = \varphi_{14} = \pi, \varphi_{34} = \frac{3\pi}{2}, \varphi_{13} = \frac{3\pi}{4}, \varphi_{24} = \frac{5\pi}{4} \) and \( D_{j,k} = \left( \frac{\sin \left( \frac{\varphi}{\sin \left( \frac{\pi}{2} (j-k) \right) \right)}{3} \right)^3 \); and c) for a rectangle \( \varphi_{12} = \frac{\pi}{2}, \varphi_{23} = \varphi_{14} = \pi, \varphi_{34} = \frac{3\pi}{2}, \varphi_{13} = \arccos \left( \frac{\sqrt{2}}{2} \right), \varphi_{24} = \arccos \left( \frac{\sqrt{2}}{2} \right) + \pi, \) and \( D_{12} = D_{34} = 1, \ D_{23} = D_{14} = \frac{1}{3\sqrt{2}}, \ D_{13} = D_{24} = \frac{1}{8}. \) Fig. 1 shows evolution of the inverse temperatures \( \beta_z \) and \( \beta_d \) toward the equilibrium value of the four-spin systems in the form of a chain, a circle, and a rectangle.
These systems demonstrate qualitatively similar time dependences of $\beta_z$ and $\beta_d$. At a high ratio $\frac{\omega_0}{\omega_d} = 45$ it is possible to mark out four stages in the $\beta_d$ time dependences. These stages are clearly seen in the $\beta_d$ time dependence for a circle spin system (Fig. 1a, black line). The first stage, $0 < t < 0.5$ is characterized by a fast drop of the inverse temperature about twice. During the second stage, $0.5 < t < 2$, the rate of the $\beta_d$ drop is decreased, and it is increased again at the next stage $2 < t < 2.5$. The dependence is substantivally non-exponential. Only at the last stage when of the temperature difference is small, the time dependence can be fitted by an exponential laws with the characteristic time close to $\tau \sim 1$. Of course, these stages can be marked out in the $\beta_z$ time dependences. However the variations of the Zeeman temperatures are very small because the heat capacity of the Zeeman subsystem is high. The inverse temperature of the Zeeman subsystem $\beta_z$ increases less than by 2.5% (Fig. 1b), while the inverse temperature of the dipolar subsystem decreases 70 times (Fig. 1a).

With decreasing a ratio of the Zeeman energy splitting to the dipolar energy, the behave of the time dependence is changed. Only two stages can be marked out as a ratio $\frac{\omega_0}{\omega_d} = 20$ (Fig. 1a, blue dash-dotted line). Both stages can be fitted by the exponential laws with characteristic times, $\tau$: $\tau = 0.6$ for the first stage $0 < t < 0.5$ and $\tau = 3.3$ the second one by . So, the decrease of the ratio of the Zeeman energy splitting to the dipolar energy leads to the increase of the characteristic time of achieving the equilibrium state.

IV. ENTANGLEMENT AT A LOW ZEEMAN AND DIPOLAR TEMPERATURES

Several parameters have been proposed for characterizing the entangled state of a spin system: the von Neumann entropy, entanglement of formation, log negativity, concurrence of a pair of spins, and etc. [2, 3, 35–38]. We will characterize the entangled states by the concurrence between two, $m$-th and $n$-th, spins which is defined as [35]

$$C_{mn} (\beta_z, \beta_d) = \max \{ q_{mn} (\beta_z, \beta_d), 0 \}, \quad (14)$$

with $q_{mn} (\beta_z, \beta_d) = \lambda^{(1)}_{mn} (\beta_z, \beta_d) - \lambda^{(2)}_{mn} (\beta_z, \beta_d) - \lambda^{(3)}_{mn} (\beta_z, \beta_d) - \lambda^{(4)}_{mn} (\beta_z, \beta_d)$. Here $\lambda^{(k)}_{mn} (\beta_z, \beta_d)$ $(k = 1, 2, 3, 4)$ are the square roots of eigenvalues, in the descending order, of the following non-Hermitian matrix:

$$R_{mn} (\beta_z, \beta_d) = \rho_{mn} (\beta_z, \beta_d) (\sigma_y \otimes \sigma_y) \tilde{\rho}_{mn} (\beta_z, \beta_d) (\sigma_y \otimes \sigma_y), \quad (15)$$
where \( \rho_{mn}(\beta_z, \beta_d) \) is the reduced density matrix. For the \( m \)-th and \( n \)-th spins, the reduced density matrix \( \rho_{mn} \) is defined as \( \rho_{mn} = Tr_{mn}(\rho) \) where \( Tr_{mn}(...) \) denotes the trace over the degrees of freedom for all spins except the \( m \)-th and \( n \)-th spins. In Eq. (15) \( \tilde{\rho}_{mn} \) is the complex conjugation of the reduced density matrix \( \rho_{mn} \) and \( \sigma_y \) is the Pauli matrix 

\[
\sigma_y = \begin{pmatrix}
0 & -i \\
i & 0
\end{pmatrix}.
\]

For maximally entangled states, the concurrence is \( C_{mn} = 1 \) while for separable states \( C_{mn} = 0 \).

Figs. 2 and 3 present the results of numerical calculations of the concurrence \( C_{mn} \) in the four-spin circle (Figs. 2a and 3a), rectangle (Figs. 2b and 3b), and chain (Figs. 2c and 3c). Dependences of concurrences on inverse temperatures \( \beta_z \) and \( \beta_d \) at \( \omega_0 / \omega_d = 45 \) are presented in Figs. 2. Here the dashed red lines present concurrences in the equilibrium states (\( \beta_z = \beta_d \)) and the dotted yellow lines show dynamic concurrency variations at achieving the equilibrium state.

A. Conclusion

We have investigated the entanglement evolution during establishing an equilibrium in four-spin dipolar coupled systems with various structure: circle, rectangle, and chain. The concurrence was considered as a measure of entanglement. Entanglement is observed in non-equilibrium states (\( \beta_z \neq \beta_d \)) and can appear when both temperatures much higher than the temperature of the entanglement appearance in the equilibrium state. This confirms the results of previous works that non-equilibrium conditions are more favorable for the generation of entangled states as equilibrium states [23, 24]. Entangled states appear and are stored at lower dipolar temperatures relative to the Zeeman one \( (\beta_d^{-1} < \beta_z^{-1}) \). We found that the concurrence non-monotonically depends on both temperatures (Fig. 2).

Considering the establishment of the equilibrium state, we showed that the well-known high-temperature approximation is not applicable for a homonuclear spin system at low temperature, and the temporal dependence of the inverse temperatures of the spin subsystems is given by a solution of non-linear equations.

We demonstrated that the concurrence dependences on time is non-monotonic during the equilibrium state establishment process (Fig. 3) and entanglement fades long before equilibrium is established (compare Figs. 1 and 3). The behavior of entanglement depends strongly
on the ratio of the Zeeman energy to the dipolar energy. At high ratio, the concurrence in
the system decreases significantly till the residual very small quantity in time of the order
of $T_2$. At low ratio, the establishment of equilibrium (Fig. 1a and b, blue dash-dotted line)
and the decay of entanglement (Fig. 3a, green dotted line) take time up to 1 ms.

Obtained results open an effective way to the experimental testing of entanglement in
spin systems by using non-equilibrium states to generate the entangled state at higher tem-
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Figure Captions

Fig. 1 Equalization of the inverse spin temperatures of the dipolar (a) and Zeeman (b) subsystems in a four spin circle (solid black line), chain (dashed red line), and rectangle (dotted yellow line) at $\omega_0/\omega_d = 45$. Blue dash-dotted line – the inverse spin temperatures of the dipolar (a) and Zeeman (b) subsystems in a four spin circle at $\omega_0/\omega_d = 20$.

Fig. 2 The concurrence in four spin system as a function of the inverse temperatures $\beta_z$ and $\beta_d$: (a) $C_{12}$ in the circle, (b) $C_{13}$ in the rectangle, and (c) $C_{14}$ in the chain. Dashed red line - equilibrium state with $\beta_z = \beta_d$. Dotted yellow line – evolution of the concurrence under equalization of the Zeeman $\beta_z$ and $\beta_d$ inverse temperatures due the influence of the non-secular term $H_{nd}$.

Fig. 3 Time dependence of concurrence $C_{mn}$ in a four spin system at $\omega_0/\omega_d = 45$: (a) circle, (b) rectangle, (c) chain. Solid black line – $C_{12}$, dashed red line – $C_{13}$, and dotted blue line – $C_{14}$. Green dotted line – $C_{12}$ in a four circle at $\omega_0/\omega_d = 20$. 

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