Cavity-Assisted Remote Energy Transfer

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Strong molecule-light interaction enables the control of molecular structures and dynamical processes. A molecular model is proposed to greatly enhance the intermolecular distance of resonant energy transfer, where the molecules are strongly driven by an optical cavity. The optimal intermolecular distance and quantum yield of energy transfer are observed, resulting from the balance between dipole-dipole interaction and molecule-cavity coupling. Our approach is non-perturbative, going much beyond the Förster mechanism of resonant energy transfer. Our work sheds the light on the spectroscopic study of the cooperative energy transfer in molecular polaritons.

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I. INTRODUCTION

Long-range excitation energy transfer in molecules plays a key role in the study of molecular aggregates and proteins, which involve the exciton migration over 10-100s nm. This offers a highly sensitive approach to investigate intra- and inter-molecular distances on the nanometer scale, revealing the dynamical information about biomolecular structures and interactions [1–3]. The long-range energy transfer, however, is difficult because the efficiency dramatically drops as the donor-acceptor separation grows, resulting in the challenge for the fluorescence detection above 10s nm [4–5]. Some prominent mechanisms are responsible for this bottleneck: disorder, local trapping, vibrations and synthetic limitations. Elaborate experiments demonstrate the megamolecular structure incorporating fluorescent proteins as a promising building block for studying the distance-dependent energy transfer in large molecules [6]. New donor-acceptor constructs and several strategies to extend the energy transfer distance have been investigated, using lanthanides [7–9], quantum dots [10–11], gold nanoparticle quenchers [12–14] and metal-induced energy transfer [15–16]. Although these advanced techniques have the drawbacks of further complicating the sample structures and the preparation, the gold nanoparticles still have the advantage in many applications due to the local field enhancement.

From the quantum mechanical treatment, it is well known that the quantum yield of resonant energy transfer (RET) is strongly determined by the overlap between density of the states in the donor and the acceptor, apart from the dependence on the molecular structures [17–18]. This has been extensively explored in the Förster theory of resonant energy transfer (FRET), treating the dipole-dipole interactions as perturbations [19]. Nevertheless, it is worth noting Purcell effect such that the photonic environment can modify the fluorescence emission of molecules, due to the drastic change of the photonic spectral density in confined geometry [20–21]. This leads to the so-called molecular polaritons that opens a new avenue to control the resonant energy transfer [22–25]. Recently significant efforts were devoted to the microcavities [26–28], surface plasmon polaritons [29–30], nano-apertures [31–35], nanoparticles [36–42] and hyperbolic metamaterials [43–44], in attempts to overcome the 10nm barrier in diffraction-limited confocal microscopes so that to enhance the quantum yield of FRET. Polaritons can act as an efficient and ultrafast energy-transfer pathway between exciton states, evident by the photoluminescence spectroscopy of J-aggregated molecular dyes placed in an optical cavity [35]. However, FRET assumes the weak dipole-dipole interaction to make the perturbation applicable, which is no longer exact in molecular polaritons since the joint photon-matter states may result in a large density of states overlap between donor and acceptor. Such drawback also exists in the transition state theory of electron transfer, despite that it does offer a microscopic description for a broad range of reaction processes [46]. On the other hand, considerable enhancement of RET requires a delicate balance between exciton migration and the other donor radiative as well as nonradiative processes [42–44,47]. These are still open issues, which requires the theoretical efforts beyond the perturbative framework.

In this article we propose a long-range resonant energy transfer in molecules driven by an optical cavity. We consider such a model that the donor molecule is placed in the cavity whereas the acceptor molecule is outside. The quantum master equation incorporating the polariton effect is obtained, going beyond the perturbative treatment of dipole-dipole interaction. We observe an optimal intermolecular distance of energy transfer, showing a great enhancement through the balance between dipole-dipole interaction and the Rabi splitting induced by the joint molecule-cavity states. Our model is promising of being...
wave \( E(\mathbf{r}, t) = E_{\ell}(\mathbf{r}) e^{i \mathbf{k} \cdot \mathbf{r} - i \nu t} \) with frequency \( \nu \) and wave vector \( \mathbf{k} \) irradiates the system and drives the ground state to the excited state “a”. The hopping rate between state “a” and “b” is assumed to be \( \Delta \) and the spatial dependence of it will be elucidated later. The total Hamiltonian \( \mathcal{H} \) can be decomposed into the components:

\[
\mathcal{H}_S = \omega_1 |1\rangle \langle 1| + \omega_1 |2\rangle \langle 2| + \Delta (|1\rangle \langle 2| + |2\rangle \langle 1|) + \Omega |1\rangle \langle 0| e^{i \mathbf{k} \cdot \mathbf{r} - i \nu t} + \Omega |0\rangle \langle 1| e^{-i \mathbf{k} \cdot \mathbf{r} - i \nu t},
\]

(1)

\[\mathcal{H}_I = \sum_{j=1}^{2} \sum_{k, \sigma} g^j_k e^{i \nu t} |0\rangle \langle a_{k, \sigma}| + g^{\ast \dagger j}_k |a_{k, \sigma}\rangle \langle 0|,\]

(2)

and

\[\mathcal{H}_R = \sum_{k, \sigma} \omega_k a^\dagger_{k, \sigma} a_{k, \sigma},\]

(3)

where \( \mathcal{H}_S \) represents the free system Hamiltonian, \( \mathcal{H}_R \) is the Hamiltonian for the free reservoirs and \( \mathcal{H}_I \) is the interaction term. We adopt the convention to set \( \hbar = 1 \).

The real parameter \( \Omega = \mathcal{P}_{ac} \mathcal{E} \) in \( \mathcal{H}_S \) is the Rabi frequency of the field while \( \mathcal{P}_{ac} \) represents the dipole moment of the transition from ground state “c” to “a”, and \( \mathcal{E} \) represents the local electric field. \( \omega \) is the excitation energy of each molecule. The creation and annihilation operators \( a^\dagger_{k, \sigma} \) and \( a_{k, \sigma} \) in \( \mathcal{H}_I \) and \( \mathcal{H}_R \) are the environment photon operators, the index “\( \sigma \)” denotes the polarization of the photons, and \( g^j_k \) in \( \mathcal{H}_I \) denotes the coupling between the environment and the system.

Define \( h_s \equiv \nu (|1\rangle \langle 1| + |2\rangle \langle 2|) \) as the new system Hamiltonian and we can rewrite the total Hamiltonian, \( \mathcal{H} = (h_s + \mathcal{H}_R) + (\mathcal{H}_S - h_s + \mathcal{H}_I) \equiv h_0 + \mathcal{H}_{int} \), where \( h_0 = h_s + \mathcal{H}_R \) is treated as the free Hamiltonian and \( \mathcal{H}_{int} = \mathcal{H}_S - h_s + \mathcal{H}_I \) is the interaction term. The interaction picture description used in the following context is defined according to this definition.

With the free Hamiltonian and the interaction component defined above, the interaction Hamiltonian \( \mathcal{H}_{int} \) in the interaction picture takes the form \( \tilde{\mathcal{H}}_{int}(t) = e^{i \theta_0 (\mathcal{H}_S - h_s)} e^{-i \theta_0 t} + e^{i \theta_0 \mathcal{H}_I} e^{-i \theta_0 t} \equiv \tilde{\mathcal{H}}_0(t) + \tilde{\mathcal{H}}_I(t) \), where \( \tilde{\mathcal{H}}_0(t) = e^{i \theta_0 (\mathcal{H}_S - h_s)} e^{-i \theta_0 t} \) and \( \tilde{\mathcal{H}}_I(t) = e^{i \theta_0 \mathcal{H}_I} e^{-i \theta_0 t} \). The purpose of this treatment is that now \( \tilde{\mathcal{H}}_0(t) \) becomes time independent. \( \tilde{\mathcal{H}}_0(t) \) can be calculated explicitly using the Baker-Hausdorff formula,

\[
\tilde{\mathcal{H}}_0 = \sum_{j=1}^{2} \delta_j |j\rangle \langle j| + (\Delta |1\rangle \langle 2| + \Omega |1\rangle \langle 0| + h.c.)
\]

(4)

where \( \delta_j = \omega_j - \nu \) is the energy difference between the laser and the \( j^{th} \) excited state. Similarly, the interaction Hamiltonian is

\[
\tilde{\mathcal{H}}_I(t) = \sum_{j=1}^{2} \sum_{k, \sigma} g^j_k e^{i (\nu - \omega_k) t} |j\rangle \langle j| a_{k, \sigma} + h.c.
\]

(5)

extended to many molecular ensembles incorporating the cooperativity, which is essential for the nonlinear spectroscopic study of cooperative energy transfer in molecular polaritons.

II. MODEL AND EQUATION OF MOTION

The scheme of energy transfer between coupled donors and acceptors is shown in Fig. 1(a). The donor in the cavity is driven by a strong laser of frequency \( \nu \), and the excitation of the donor can be transmitted through a non-radiative process to the acceptor outside the cavity. The excitation of the acceptor can be measured through a detector. The field inside cavity is assumed to be single mode and can be described classically when the driving laser is intense. The donor is then pumped effectively by a classical field, whose intensity varies with the driving laser. The model is simplified ignoring the doubly-excited state due to the mismatch of transition energies caused by exciton-exciton scattering. As illustrated in Fig. 1(b), the system to be considered consists of three states, the ground state and two excited states denoted as “a” and “b”. The schematic illustration of the system is shown in Fig. 1. The excited states can spontaneously emit a photon and decay to the ground state “c”. The coherent
The equation of motion (von-Neumann equation) for the density operator is determined by \( \dot{\rho}(t) = i \left[ \rho(t), \hat{H}_{\text{int}}(t) \right] = i \left[ \rho(t), \hat{H}_0 \right] + i \left[ \rho(t), \hat{H}_{\text{f}}(t) \right] \). Inserting the formal solution \( \dot{\rho}(t) = \rho(0) + i \int_0^t ds [\rho(s), \hat{H}_{\text{int}}(s)] \) only back to the interaction term \( i \left[ \rho(t), \hat{H}_f(t) \right] \) gives the standard form of the equation of motion for the total density operator,

\[
\dot{\rho}(t) = i \left[ \rho(t), \hat{H}_0 \right] + i \int_0^t ds [\rho(s), \hat{H}_{\text{int}}(s)], \quad \hat{H}_f(t) \quad (6)
\]

where \( n_\nu \) is the number of particles at frequency \( \nu \) in the environment which is set to be zero for the consideration of vacuum surroundings, and \( \Gamma_{jj'} \) is the dissipation constant defined as follows,

\[
\Gamma_{jj'}(\nu) = \frac{V}{(2\pi)^2} \sum_{\sigma} \int d^3k \, g_{\omega_{k,\sigma}}^j g_{\omega_{k,\sigma}}^{j'} \delta(\nu - \omega_k). \quad (8)
\]

### III. RESONANT DIPOLE-DIPOLE INTERACTION

Since the molecular size is much smaller than the wavelength of the light, we may invoke the dipole approximation that leads to \( g_{\omega_{k,\sigma}}^j = \sqrt{\omega_k / 2V} \hat{p}_j \cdot \hat{e}_k \exp[i(k \cdot r)] \), where \( r_j \) is the displacement vector of the \( i^{\text{th}} \) dipole, \( \hat{e}_k \) is the polarization vector of the field, and \( \hat{p}_j \) is the dipole moment of the \( i^{\text{th}} \) atom. The individual decay constant of the \( i^{\text{th}} \) dipole following definition \( 8 \) is \[21\]

\[
\Gamma_{ii}(\nu) = \frac{\alpha^2 \nu^3}{3(2\pi)^2}, \quad (9)
\]

where \( \nu \) is the laser frequency, and in more general scenarios it is the frequency of the system. The cross dissipation constant (or collective dissipation constant) \( \Gamma_{jj'} \) for \( j \neq j' \) is given as follows,

\[
\Gamma_{jj'} = \frac{\nu^3}{(2\pi)^2} \int d^3k \sum_{\sigma} (\hat{P}_j \cdot \hat{e}_k)(\hat{P}_{j'} \cdot \hat{e}_k^*) e^{i\nu k \cdot (r_j - r_{j'})}. \quad (10)
\]

Notice that by Cauchy-Schwarz inequality the cross dissipation constant \( \Gamma_{jj'} \leq (\Gamma_{11} \Gamma_{22})^{\frac{1}{2}} \). The collective dissipation constant \( \Gamma_{12} \) decays with the increasing angle differences between the two dipoles and also with the distance between them when \( \nu |P_2 - P_1| \leq 1 \). Following the Eqn. \[10\] the collective dissipation constant is related to the individual dissipation constants by

\[
\Gamma_{jj'} = f(\xi) \sqrt{\Gamma_{11} \Gamma_{22}}, \quad (11)
\]

Under Born-Markov approximation the total density matrix can be decomposed into system and bath components, \( \rho(t) = \rho_s(t) \otimes \rho_B \). Furthermore, we apply the Weisskopf-Wigner approximation, and ignore the Cauchy principal value of the integral \( \int_0^\infty ds e^{is\xi} = \pi \delta(x) + i\pi \delta(1/x) \), then the reduced equation of motion of the system is as follows,

\[
f(x) = \frac{3}{2} \left\{ (\hat{P}_j \cdot \hat{p}_j - (\hat{P}_j \cdot \hat{R})(\hat{p}_j \cdot \hat{R})) \frac{\sin \xi}{\xi} + (3(\hat{P}_j \cdot \hat{R})(\hat{p}_j \cdot \hat{R}) - \hat{P}_j \cdot \hat{P}_j) \left( \frac{\sin \xi}{\xi^3} - \frac{\cos \xi}{\xi^2} \right) \right\}, \quad (12)
\]

where \( f(x) \) is the real function such that \( |f(x)| \leq 1 \), and \( \xi = \nu |r_2 - r_1| \) is the distance between two dipoles multiplied by the laser frequency. Applying \( \sum_\sigma \hat{e}_k \cdot \hat{e}_k = 1 \) in Eqn. \[10\] where \( \hat{kk} \) is the dyadic product, the function \( f(\xi) \) for two parallel dipoles is calculated to be as follows,

\[
f(\xi) = \frac{3}{4} \left\{ -[1 - \cos^2 \theta] \frac{\cos \xi}{\xi} + [1 - 3 \cos^2 \theta] \left( \frac{\sin \xi}{\xi^3} + \frac{\cos \xi}{\xi^2} \right) \right\}, \quad (13)
\]

where \( \theta \) is the angle between the dipoles and the displacement vector joining the two dipoles. This expression is valid as long as the splitting between the energy levels is much less than the emission and absorption frequency, and that the frequency of the laser is near resonance with the system.
IV. PARAMETER REGIMES RELATED TO EXPERIMENTS

The FRET technique is applicable to the nonradiative dipole-dipole coupling where the wavelength of the virtual photon is larger than the distance between the two dipoles. For example, the typical length scale of the donor fluorescence is in the 300-800 nm range, and the FRET in an approximate 1-10 nm range [22]. This corresponds to $\xi \sim 0.005 - 0.2$ and the scaling relation $\Delta \sim \xi^{-3}$ holds within the range according to Eqn. [13].

When the distance is much less than the length scale, the ideal dipole approximation in Förster’s calculation breaks down. On the other hand, if the distance is much larger than the length scale, the FRET process stops and the intermediate and radiation zones which were ignored become relevant. The physics discussed in this paper is within the regimes where the FRET technique is applicable.

In the near zone region, the relevant components are those proportional to $\xi^{-3}$ due to the static electric field. Ignore the parts due to the intermediate and far field radiation, and consider the case $\theta = \frac{\pi}{2}$ and $\Gamma_{11} = \Gamma_{22} = \Gamma$, then $\Delta \approx \frac{3\pi}{\xi^2}$. The coherent interaction $\Delta$ is divergent as $\xi \to 0$ according to the above definition. When $\xi \to 0$, the Förster’s theory is not applicable and several regularization methods can be performed on $\Delta$ to render it finite [23]. The details of the regularization goes beyond the scope of this study. The typical value of dipole moments $|\vec{P}|$ of a strongly dipolar molecule is roughly of order a few debyes. For example, water molecule has the dipole moment of approximately 1.84 debyes, and chloromethane about 8.5 debyes. The approximate range of values of these strongly dipolar molecules gives an estimation of the order of magnitude for the dissipation constant $\Gamma_{ii}/\nu \sim 10^{-11} - 10^{-9}$ in natural unit. Since $f(\xi)$ is very close to identity function in the range of our interest, we set $\Gamma_{ij} = \Gamma_{ii} = \Gamma_h = 10^{-9}$ in our study. The approximate range of $\Delta$ calculated from Eqn. [13] is shown in Fig. 5. In addition, in the quantum master equation for molecules, the inhomogeneous broadening that originates from vibrations and other dense medium often contributes crucially to the electronic dephasing. This effect can lead to an additional path of decay $\Gamma_{inh,ii}$. Then the total decay rate $\Gamma_{ii} = \Gamma_{h,ii} + \Gamma_{inh,ii}$, where $\Gamma_{h,ii}$ is given above without the subscript “h” and $\Gamma_{inh,ii}$ is the additional contribution coming from inhomogeneous broadening. Typically in many dipolar molecules $\Gamma_{inh,ii} \sim 100\text{GHz}$, which is approximately $10^{-4}\nu$.

V. ENERGY TRANSFER OVER LONG DISTANCE

The efficiency of the excitation energy transfer $\eta$ is related to the effective energy transfer rate $\kappa$ by $\eta = \frac{\kappa}{\kappa+\Gamma_{11}\rho_{ac}}$ [54], and $\eta \approx \frac{\kappa}{\Gamma_{11}\rho_{ac}}$ when the rate is low. In the nonequilibrium steady state (NESS), the rate for the excitation energy transfer and that of the acceptor’s decay to the ground state coincide as the result of the conservation of probability, i.e. $\kappa = \Gamma_{12}\rho_{bd}$. With low driving field intensity so that the Rabi frequency is low, the rate of energy transfer can be achieved perturbatively as a power series of the donor-acceptor coupling $\Delta$, and the leading order in the perturbative expansion is $\Delta^2$. Since $\Delta \sim \xi^{-1}$, the result reduces to the Förster regime that the transfer rate against donor-acceptor distance obeys the inverse sixth power scaling[19]. However, a strong drive field can lead to the remote energy transfer over long distance between the donor and acceptor molecules, resulting in the breakdown of the perturbative treatment. The nonperturbative theory predicts new phenomena beyond the Förster mechanism. In what follows, we develop the nonperturbative theory for the molecular energy transfer and demonstrate the possibility of the greatly enhanced long-range energy transfer.

A. The steady state

The reduced density matrix for NESS can be obtained by solving Eqn. 7. The full nonperturbative solution is extremely complicated, but several interesting special cases exist and show the analytical behaviors that are most relevant for our discussion. Our analysis is based on the analysis of the NESS solutions and the dynamics towards the NESS is discussed separately later.

First we consider the regime of strong driving field when the Rabi frequency is close to the donor-acceptor energy splitting $\omega_1 - \omega_2$. For this “near resonance” regime, the solution behaves very differently at small distances from that at large distances [see the navy lines in Fig. 2 (a)]. First we look at the solution at the short distance, or equivalently, when the coupling $\Delta$ is strong. For the sake of succinctness, consider zero detuning $\delta_1 = \omega_1 - \nu = 0$ (then $\delta_2 = \omega_2 - \omega_1$ is the energy splitting) and symmetric total individual decay constants $\Gamma_{11} = \Gamma_{22} = \Gamma \ll \delta_2$. In the limit of zero detuning and the Rabi frequency close to the donor-acceptor splitting, the convoluted exact solution of the effective rate of energy transfer $\kappa$ given in the appendix is simplified to the following expression,

$$\kappa = \Gamma \left( 1 - \frac{3}{50} \gamma - \frac{4}{45} \frac{\Delta}{\delta_2} \right) \tilde{\Omega} + O(\gamma^2) + O(\delta_1^2)$$

(14)

where $\gamma = \frac{\delta_2^2}{\Delta^2} \approx \frac{1}{9} \frac{\delta_2^2}{\xi^6}$ is a small quantity at short distances, and $\tilde{\Omega} = \frac{\Delta^2}{3\xi^2}$ measures the bias between the Rabi frequency and the donor-acceptor level splitting in the scale of the coupling potential. The terms after the first two are all of the second or higher orders and can be ignored. We will refer to it as the “strong coupling expansion” as it is accurate at short distances and strong couplings. It is in contrast to the “weak coupling expansion” to be introduced later which de-
scribes large distance or weak coupling scenarios. Eq. 14 shows the super energy transfer such that the constant term independent of donor-acceptor distance dominates in the vicinity of $\Omega - \delta_2$ where the perturbative regime breaks down. This equation predicts that the optimal transfer rate is approximately $\frac{\xi}{\xi}$, which is in accordance with Fig. 2 (c). The approximate range of the distance-independent transfer rate extends to until $\xi \approx \min(\frac{3\Gamma_h}{4(\Omega - \delta_2)}, \frac{3\Gamma_h}{4\Gamma})$. It subsequently begins to decay, roughly in the manner of $1 - \frac{8\Gamma^2}{3h}\xi^6$ [see Fig. 2 (a, b) navy lines]. This $-\xi^6$ decay will stop once the rate drops to the vicinity of zero since $\xi$ is non-negative.

Fig. 2 (a, b) show the rate and the efficiency of the excitation energy transfer against the distance between the donor and the acceptor. The near resonance cases are the top navy curves and the off-resonance ($\Omega \neq |\omega_1 - \omega_2|$) results are the dotted and orange lines. Physics at short distances (the leveling off and the curving down segment) of the navy curves in Fig. 2 (a, b) is described by the strong coupling formula Eqn. 14 and the segment that drops at $\xi^{-6}$ (the straight slanted lines) can be described by the expansion Eqn. 15 introduced below. The vertical lines tell the distances at which the transfer rate/efficiencies of the two cases (in and out of resonance) drop to a certain same detectable value, and the black horizontal line connecting the in-resonance curve on top and the off-resonance curve in below measures the distance increased by the resonance phenomenon. As is indicated from Fig. 2 (a, b), the distance at which the rate/efficiency of the transfer drops to the same value is increased by approximately one order of magnitude by the strong driving field at the resonant Rabi frequency. Assuming that the Rabi frequency of the laser-system, which is tuned to the energy splitting of the system, has an error $\sigma$, the rate/efficiency of the excitation energy transfer will begin the rapid drop of $\xi^{-6}$ at a shorter distance $\xi \approx \sqrt[3]{3\Gamma h/(4\sigma)}$, as is indicated in the dotted navy lines in Fig. 2 (a, b).

At large distances when $\xi \gg \min(\frac{3\Gamma_h}{4(\Omega - \delta_2)}, \frac{3\Gamma_h}{4\Gamma})$, $\gamma$ and $\tilde{\Omega}$ are of order one or above, the strong coupling expansion Eqn. 14 breaks down and the exact result approaches the weak coupling expansion Eqn. 15. To be more specific, the strong coupling expansion above is accountable for the near-resonance phenomenon ($\tilde{\Omega} \lesssim 1$) at short distances ($\xi \lesssim \sqrt[3]{3\Gamma h/(4\Gamma)}$), while the weak coupling expansion to be introduced later describes the physics in the complementary regimes, i.e. at large distances or off resonance. At short distances, the rate of energy transfer from the off-resonance mechanism [Eqn. 15] is suppressed roughly by a factor of $\tilde{\Delta}^2$ compared to that of in resonance mechanism Eqn. 14. The efficiency of the excitation energy transfer $\eta = \frac{\kappa}{\kappa + \Gamma_{\text{ratio}}}$ $\sim \kappa$ is approximately proportional to the rate as the result of low transfer rates and slowly changing $\rho_{aa}$ [Fig. 2 (b)].

In the regimes where the above approximation becomes invalid, namely at large distances or off resonance, we refer to the following expansion to describe the physics occurred. Under the assumption that $\Omega \gg \Gamma \gg \Gamma_{12}$ and in the vicinity of zero detuning $\nu = \omega_1$, the leading order of $\kappa$ in $\Delta$ and $\Gamma$ is given as follows,

$$\kappa = \Gamma \cdot \frac{2\Omega^2 + \delta_2^2 + O(\Gamma^2)}{2\delta_2^2} \Delta^2 + O(\Delta^4)$$

(15)

where

$$\Delta^2 = \frac{\Delta^2}{(\delta_2^2 - \Omega^2)^2/\delta_2^2 + O(\Gamma^2)}$$

(16)

is the dimensionless coupling. The coupling in the case of nonradiative energy transfer is give by $\Delta = \frac{\sqrt{h}}{\delta_2}$. We notice that in the off-resonance, weak laser regimes, i.e. $\Omega \ll \omega_1 - \omega_2 = |\delta_2|$, the rate of transfer returns to that of the perturbative result

$$\kappa \approx \Gamma \cdot \frac{\Delta^2}{2\delta_2^2} \approx \frac{9\Gamma \Delta^2}{32\delta_2^2 \xi^6} \sim \frac{1}{\xi^6}$$

(17)

Therefore, the Förster’s scaling in the FRET regime can be reproduced when the Rabi frequency is sufficiently below the energy splitting between the two molecules. For extremely large Rabi frequencies $\Omega \gg \delta_2$ this $-\xi^6$ scaling still holds, but the rate and efficiency will be dependent on the strength of the driving field and in fact drop down as the intensity of the driving field is tuned up.

The above “weak coupling” expansion should not be understood as the perturbative result. It is the nonperturbative result that recovers the perturbative result in the low Rabi frequency limit. The approximation is applicable when $\Delta^2 \ll 1$. The condition is satisfied when $\Delta^2 \ll \Gamma^2$ or $\Delta^2 \ll (\delta_2^2 - \Omega^2)^2/\delta_2^2$. The first condition corresponds to $\xi \gg \sqrt[3]{3\Gamma h/(4\Gamma)}$, which is the complementary regime of the strong coupling expansion Eqn. 14. The second condition is equivalent to $|\Omega - \delta_2| \gg \Delta$, which is the off-resonance condition under which the $-\xi^6$ scaling can be reproduced in the FRET regime.

In Fig. 2 (a, b), the orange lines and the overlapped dotted lines are for the off-resonance regimes. The orange lines represent two different cases of the driving field, one is for the low intensity laser ($\Gamma < \Omega < 20(\omega_1 - \omega_2)$) and the other is for the extremely strong laser ($\Omega = 2(\omega_1 - \omega_2)$). For lasers with Rabi frequency $\Omega$ in the range of $10^{-3}$ to $20\%$ of the energy splitting ($\omega_1 - \omega_2$), the differences in the rates of energy transfer are indistinguishable and all represented by the orange line in Fig. 2 (a). This line provides the scale of the transfer rates in the off-resonance regimes, and it coincides with the overly-strong laser case with $\Omega = 2(\omega_1 - \omega_2)$. For the Rabi frequencies between the two limits, the lines for the transfer rates lie between the solid navy line and the orange line, and for regimes beyond the two limits, they are
FIG. 2: (a) The rate of energy transfer vs. donor-acceptor separation $\xi$. The solid navy line is for the resonance case $\Omega = \omega_1 - \omega_2 = -\delta_2$, the dotted navy line is for $(\Omega - |\delta_2|)/|\delta_2| = 0.2\%$. The exact result for the overly high Rabi frequency $\Omega = 2|\delta_2|$ coincides with the low Rabi frequency results $\Omega/|\delta_2| \in [10^{-3}, 20\%]$, and are shown in orange. Results for parameter regimes that lie between the two limits (near resonance), which are not shown in the plot, lie between the navy and orange lines. The dotted black line that overlaps with the orange line is the weak laser limit (still requiring $\Omega \gtrsim \Gamma$) leading order result scaling as $\xi^{-6}$. (b) The same condition as in (a) for the efficiency of excitation energy transfer $\eta$. The efficiency is almost identical to the rate $\kappa$ up to a factor of $\Gamma$ when $\kappa \ll 1$. For (a) and (b), $\delta_2 = -0.1$, $\Gamma = 10^{-4}$. (c) The navy line is for $\delta_2 = -0.01$ and the orange line for $\delta_2 = -0.02$ at $\delta_1 = 0$, $\Delta = 5 \times 10^{-4}$, and $\Gamma = 10^{-4}$. 
below the orange line. The dotted line is the leading order approximation which is given by Eqn. [17]. Besides, for the in-resonance case, there is a \( \frac{5\delta}{2}(1 - \omega^2/\delta^2) \) enhancement in the transfer rate compared to that of the off-resonance regime as can be inferred from Eqn. [15] [see Fig. 2 (c)]. The above nonperturbative result derived by the master equation approach predicts, for the in-resonance regime, a slower rate of decay for the energy transfer than the inverse sixth power, and in addition a boost in the efficiency of the energy transfer when the Rabi frequency is around the energy splitting.

B. Dynamical relaxation towards NESS

![Figure 3](image-url)

**FIG. 3:** The dynamics of the excitations on the acceptor state with the time multiplied by laser frequency \( \nu \). (a) \( \Omega = \delta_2 \), (b) \( \Omega = \delta_2(1 \pm 3\%) \). The color code is given as follows, the navy lines: \( \Delta = 4\Gamma \), the orange lines: \( \Delta = 2\Gamma \), the green lines: \( \Delta = \Gamma \), and the red lines: \( \Delta = 0.5 \Gamma \). For (a) and (b), \( \Gamma = 10^{-3} \), \( \Gamma_h = 10^{-9} \), and \( \omega_1 - \omega_2 = 0.1 \).

The above discussions are based on the NESS solutions. The validity of the NESS solution can be verified by examining the eigenvalues of the Liouville superoperator. Here we look at the dynamics of the system initially prepared to be on the ground state and irradiated by the laser beam at time \( t = 0 \).

For the resonance case \( \Omega = \omega_1 - \omega_2 \), the dynamics can be roughly divided into three regimes, the under-damped regime \( (\Delta > \Gamma) \), the critically-damped regime \( (\Delta = \Gamma) \), and the over-damped regimes \( (\Delta < \Gamma) \). In the under-damped regime, the system oscillates around the value of the NESS and the amplitude of oscillation decays with time [Fig. 3 (a)]. For the parameters used in Fig. 3 (a), the NESS can be approximated by the strong coupling expansion Eqn. [14] where the leading term is dominated by the constant \( \frac{1}{12} \Gamma \) regardless of the values of \( \Delta \) in the range of its applicability plus a negative correction. Therefore, the asymptotes of the four lines, especially the three with larger \( \Delta \)'s, are close to one another and all around or slightly less than \( \frac{1}{12} \Gamma \). In the over-damped case, the oscillation is replaced by a steadily-increasing curve as the excitation in the acceptor accumulates with time. On the other hand, examining the dynamics of system at \( \Omega \neq \omega_1 - \omega_2 \), it can be easily noticed that the asymptotic values for the different couplings are orders-of-magnitude apart, and that the system oscillates at a dramatically higher frequency than the resonance scenario without the occurrence of the critically- and over-damped phases as in the resonant case. The rationale is as follows. Since asymptotic behavior for systems with parameters given in Fig. 3 (b) can be roughly deduced from the weak-coupling Eqn. [13] reducing \( \Delta \) by a factor of two results in a factor four folding in the transfer rate \( \kappa \), which is in accordance with Fig. 3 (b). Meanwhile, the large energy splitting \( \omega_1 - \omega_2 = \delta_2 \gg \Delta \) ensures that even a minuscule percentages of deviation of the Rabi frequency from \( \omega_1 - \omega_2 \) will cause the energy splitting between the dressed state and the acceptor state larger than the coupling potential \( \Delta \) or the decay constant \( \Gamma \), which renders the system highly oscillatory.

C. Long-range energy transfer and Rabi splitting

Now let us revisit the mechanism of the nonperturbative energy transfer studied before. In the weak laser limit, the laser beam has limited impact on the system besides the excitation of the irradiated molecule, and the process of energy transfer can be calculated assuming no change in the energy levels of the system. However, under strong continuous driving waves, the molecule-photon interaction mixes the electron and photon degrees of freedom, resulting in the Rabi splitting or dynamic stark splitting. In the case where the Rabi frequency of the laser-donor system is close to the energy gap between the donor and the acceptor molecules, the resonance effect can happen and this can dramatically change the behavior predicted from the low laser intensity result.

When the donor molecule is irradiated by the driving field with frequency close to its excitation energy, the energy level splits into the dressed states with levels different from the original level [see Fig. 4 (a)]. In other words, the state “a” splits into two states with frequencies \( \omega_1 \pm \Omega \). Naively, as the laser intensity is strength-
However, in the strong laser field regime the population on the site “b” begins to drop with the increasing laser intensity [the orange line in Fig. 4(b)]. At sufficiently large Rabi frequencies, when the laser frequency equals energy of the donor state, the level of the dressed donor state “a” ω1 ± Ω can pass the level of the acceptor and again creates a large energy gap. When Ω = ω1 − ω2, the lower dressed state is in resonance with the acceptor energy level, leading to the maximal population at the acceptor and the energy transfer is optimized. Note that the peak value of excitation on “b” is about 0.2, which can be inferred from the constant term in Eqn. 14. Further tuning up the driving intensity will render the lower dressed state of the donor and acceptor off-resonant, and results in the drop of energy transfer.

On the other hand, when the frequency of the laser beam is slightly tuned away from the system frequency, the splitted energy levels will be shifted to ω ± δω, where δω = δω1 ± δω2 [Ref. [48]]. When one of the energy levels is in resonant with that of the state “b”, i.e. ω1 = δω ± δω2, the population on state “b” will increase dramatically. This happens when δ1 = δ2 = δ1 + 0.1. For example, when ω = 0.1, δ1 = δ1 + 0.1, this resonance happens at zero detuning, δ1 = 0 [see Fig. 4(c) solid lines]. At zero detuning, we do not see a maximal number of excitations of state “a” as that in an isolated two level system. Since zero detuning corresponds to the resonance between the dressed state “a” and state “b”, the excitations on “a” are transported to state “b”, therefore we observe an unusual dip when the laser and “a” are in resonance. A more common scenario is when the Rabi frequency Ω is not exactly equal to the energy splitting between the two states ω1 − ω2, [e.g. Fig. 4(c) dotted lines].

VI. CONCLUSION

We have developed a non-perturbative theory for the energy transfer in molecular systems. The great enhancement of the long-range energy transfer between donor and acceptor has been predicted when the system is strongly driven. Our results show the optimal transfer at a certain driving power instead of increasing co-linearly along with the strengthening of the Rabi frequency, which results from the dynamic Stark effect. Notably, the transfer rate and quantum yield of energy transfer show the new scaling against the donor-acceptor distance, predicting much slower decay than the R−6 law from the Förster mechanism and larger range of effectiveness for the energy transfer. This results in the enhancement of at least one order of magnitude in molecular energy transfer. In the limit of weak driving or large detuning between donor and acceptor molecules, our theory was shown to reproduce the results given by the classical Förster’s theory. The long-range and enhanced energy transfer predicted by our model can be further measured by the time-resolved fluorescence spectroscopy. In light of
the recent advance in ultrafast nonlinear spectroscopy for nano-scaled materials, our work offers insights and per-
spectives to the spectroscopic study of cooperative energy
transfer in molecular polaritons involving a large number
of molecules.

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Appendix

FIG. 5: The typical range of the coupling potential ∆ in a FRET process as a function of inter-molecular distances ξ
in log-log scale. The horizontal line in the middle is the value of Γ = 10^{-4}. Γ_{h} = 10^{-9}.

The steady state solution

In Section V we discussed the small parameter expansions of the nonperturbative solutions of the NESS. In general,
the steady state solution can always be achieved by solving the equation of motion Eqn. 7, \dot{\rho}_s(t) = 0. Solving for the
solution of the most general scenario, nevertheless, can be extremely involved. However, the solution can be simplified
after making the assumption that Γ_{11} = Γ_{22} = Γ. To simply our discussion, we present the solution under the above
assumption which was assumed in all of the calculations in the paper.

We can reformulate the equation of motion in the Liouville space, where the system density matrix can be expressed
in terms of a nine-tuple,

\[ |\rho_s\rangle = (\rho_{11}, \rho_{22}, \rho_{33}, \Re(\rho_{01}), \Im(\rho_{01}), \Re(\rho_{02}), \Im(\rho_{02}), \Re(\rho_{12}), \Im(\rho_{12}))^T \] (18)

and T represents the matrix transpose. In the Liouville space, the equation of motion takes the following form,

\[ |\dot{\rho}_s\rangle = \mathcal{M}|\rho_s\rangle. \] (19)

The matrix representation of the time evolution operator \mathcal{M} in the Liouville space can be derived from the density
matrix equation [Eqn. 7]. Under the assumption of $\Gamma_{11} = \Gamma_{22} = \Gamma$, $\mathcal{M}$ is given as follows,

$$
\mathcal{M} = \begin{pmatrix}
-2n_\nu \Gamma & (n_\nu + 1) \Gamma & (n_\nu + 1) \Gamma & 0 & -2\Omega & 0 & 0 & 2(n_\nu + 1) \Gamma_h & 0 \\
(n_\nu + 1) \Gamma & -n_\nu \Gamma & 0 & 0 & -2\Omega & 0 & 0 & -(n_\nu + 1) \Gamma_h & -2\Delta \\
(n_\nu + 1) \Gamma & 0 & 0 & -2\Omega & 0 & 0 & 0 & -(n_\nu + 1) \Gamma_h & 2\Delta \\
0 & 0 & 0 & 0 & -3n_\nu \Gamma & -3n_\nu \Gamma & -3n_\nu \Gamma & -2n_\nu \Gamma & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\Gamma_h & 0 \\
0 & 0 & -3n_\nu \Gamma & -3n_\nu \Gamma & -3n_\nu \Gamma & -\Delta & 0 & 0 & 2n_\nu \Gamma \\
0 & 0 & 0 & -n_\nu \Gamma & -n_\nu \Gamma & 0 & 0 & 0 & 0 \\
n_\nu \Gamma & -n_\nu \Gamma & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -n_\nu \Gamma & 0 & 0 & 0 & 0 & 0 & 0 \\
\end{pmatrix},
$$

where $\Gamma_h$ is the cross decay constant, and $n_\nu$ is the photon density at frequency $\nu$.

We are interested in the regime of the steady state solution where the ambient temperature is low compared to the energy levels of the molecules, and thus the thermal excitation is rare. This mimics the experimental situation where the ambient photon bath acts like vacuum—only spontaneous decay is considered and the thermal excitation from the photon bath is absent. To simplify the calculation, we divide the system density operator into two parts: $|\rho_s\rangle = (\rho_p, \rho_c)^T$ with population terms $\rho_p$ (diagonal terms) and coherence terms $\rho_c$ (off-diagonal terms). Similarly, the time evolution matrix $\mathcal{M}$ can be expressed in the block form:

$$
\mathcal{M} = \begin{pmatrix}
\mathcal{M}_{pp} & \mathcal{M}_{pc} \\
\mathcal{M}_{cp} & \mathcal{M}_{cc}
\end{pmatrix}
$$

The steady state of the system corresponds to $\mathcal{M}|\rho_{ss}^p\rangle = 0$, where the superscript “ss” stands for the “steady state”. We substitute the coherent term $\rho_c = -\mathcal{M}_{cc}^{-1}\mathcal{M}_{cp}\rho_p$ into the equation, and obtain a linear equation for the population terms in steady state $\mathcal{A}\rho_{ss}\rangle = 0$, where $\mathcal{A}$ is

$$
\mathcal{A} = \mathcal{M}_{pp} - \mathcal{M}_{pc}\mathcal{M}_{cc}^{-1}\mathcal{M}_{cp}.
$$

Once obtaining the steady state solution for $|\rho_{ss}^p\rangle$, the coherence terms can be subsequently deduced from $\rho_c = -\mathcal{M}_{cc}^{-1}\mathcal{M}_{cp}\rho_p$. We denote the $i^{th}$ component of $|\rho_{ss}^p\rangle$ as $\rho_{ss,i}$. $|\rho_{ss}^p\rangle$ is the solution for a set of linear equations, the solutions are the rational functions. Without loss of generality, we write the $\rho_{ss,i}$ in the following form,

$$
\rho_{ss,i} = \frac{N_i}{D_i},
$$

where $N_i$ and $D_i$ are polynomial functions of $\Gamma$, $\Gamma_h$, $\Delta$, $\delta_1$, $\delta_2$ and $\Omega$. Under the assumption of $\Gamma_{11} = \Gamma_{22} = \Gamma$ and the absence of thermal environment, the explicit expression of $N_i$ and $D_i$ are provided below.

$$
D_1 = D_2 = D_3 = \ldots = D_9 =
$$

$$
- (\Gamma^2 + 4\Delta^2) \Gamma_h^6 + (\Gamma^2 \delta_1^2 - 2(5\Gamma^2 + 16\Delta^2) \delta_2 \delta_1 + \Gamma^2 \delta_2^2 + (\Gamma^2 + 4\Delta^2)(3\Gamma^2 - 8\Delta^2 - 4\Omega^2)) \Gamma_h^4
$$

$$
+ 8\Gamma\Delta ((2\Gamma^2 + 8\Delta^2 + \Omega^2) \delta_1 + (2\Gamma^2 + 8\Delta^2 - \Omega^2) \delta_2) \Gamma_h^3 - (3\Gamma^6 + 4(3\Gamma^2 + 2\Omega^2) \Gamma^4
$$

$$
+ 4(4\Delta^4 + 12\Omega^2 \Delta^2 + \Omega^4) \Gamma^2 - 8\delta_2^2 \Gamma_2^2 + 2(3\Gamma^4 + 4(\Delta^2 + 4\Omega^2) \Gamma^2 + 64\Delta^2 \Omega^2) \delta_2
$$

$$
+ 64\Delta^4 (\Delta^2 + \Omega^2) - 4\delta_1 (3\Gamma^4 + 12\Delta^2 \Gamma^2 + 26\delta_2 \Gamma^2 + 32\Delta^4 - 2(3\Gamma^2 + 8\Delta^2) \Omega^2)
$$

$$
+ 2\delta_2^2 (3\Gamma^4 + 4(\Delta + \Omega) (\Delta + \Omega) \Gamma^2 + 16 (\Gamma^2 + 2\Delta^2) \delta_2^2) \Gamma_h^2 - 8\Gamma \Delta (2\Gamma^2 \delta_2^3
$$

$$
- 2(\Gamma^2 + 2\Omega^2) \delta_2 \delta_2^2 + (2\Gamma^4 + (8\Delta^2 + 5\Omega^2) \Gamma^2 - 2(\Gamma^2 - 2\Omega^2) \delta_2^2 + 4(\Delta - \Omega) \Omega^2 (\Delta + \Omega)) \delta_1
$$

$$
+ \delta_2 (2\Gamma^2 + 26\delta_2 \Gamma^2 + (8\Delta^2 - 5\Omega^2) \Gamma^2 - 4\Delta^2 \Omega^2)) \Gamma_h
$$

$$
+ \Gamma^2 (4(\Gamma^2 + 8\Omega^2 + 4\delta_2^2) \delta_2^2 - 8\delta_1 (\Gamma^2 + 4\Delta^2 + 8\Omega^2 + 4\delta_2^2) \delta_2
$$

$$
+ (5\Gamma^4 + 24\Delta^2 \Gamma^2 + 16\Delta^4 - 64\Omega^4 + 32 (\Gamma^2 + 5\Delta^2) \Omega^2 + 8\delta_1^2 (3\Gamma^2 + 16\Delta^2 + 2\delta_2^2)) \delta_2
$$

$$
- 2\delta_1 (\Gamma^4 + 24\Delta^2 \Gamma^2 + 80\Delta^4 - 32\Omega^4 + 4 (\Gamma^2 - 4\Delta^2) \Omega^2 + 4 (\Gamma^2 + 4(\Delta - \Omega)(\Delta + \Omega)) \delta_2
$$

$$
+ (\Gamma^2 + 4\Delta^2 + 2\Omega^2) (16\Omega^4 + 2(5\Gamma^2 + 4\Delta^2) \Omega^2 + (\Gamma^2 + 4\Delta^2)^2)
$$

$$
+ \delta_2^2 (5\Gamma^4 + 4\delta_2^2 \Gamma^2 + 24 (\Delta^2 + \Omega^2) \Gamma^2 + 16 (\Delta^4 + \Omega^4))),
$$

\text{\textnormal{(24)}}.
\[ N_1 = D_1 - N_2 - N_3, \]
\[ N_2 = \\
4\Omega^2 \left( \Gamma^4 \delta_2^2 - 2\Gamma^2 \delta_2 \left( \Gamma^2 - 4\Omega^2 \right) + 4\Gamma^2 \delta_1^2 - 8\Gamma^2 \delta_2^2 + \delta_2^2 \left( \Gamma^2 \left( 5\Gamma^2 + 4\delta_1^2 + 16\Delta^2 - 8\Omega^2 \right) - 4 \left( \Gamma^2 + 4\Delta^2 \right) \Gamma^3_h \right) + \Gamma^2 \left( \Gamma^2 + 4\Delta^2 + 2\Omega^2 \right) \right), \]
\[ N_3 = \\
4\Omega^2 \left( 4\Delta^2 + \Gamma^5_h \right) \left( \Gamma^2 + \left( \delta_1 - \delta_2 \right)^2 + 4\Delta^2 + 2\Omega^2 \right) - \left( \Gamma^2 + 4\Delta^2 \right) \Gamma^7_h \), \]
\[ N_4 = \\
-4\Omega \left( \Gamma^2 \left( -2\delta_2 \delta_1 \left( \Gamma^2 + 4\delta_1^2 + 2\Delta^2 - 4\Omega^2 \right) - 4\delta_2 \Delta^2 \left( \Gamma^2 + \delta_1^2 + 4\Delta^2 - 4\Omega^2 \right) + \delta_1^2 \left( \Gamma^2 + 4\delta_1^2 \right) + \delta_2 \left( - \left( \Gamma^2 + 4\Delta^2 \right) \right) \Gamma^5_h \right) + \Gamma^2 \left( \Gamma^2 + 4\Delta^2 - 4\Omega^2 \right) + 2 \Gamma^2 \left( \Delta^2 + \Omega^2 \right) + 4 \Omega^4 \right) \right) + \delta_2 \left( - \left( \Gamma^2 + 4\Delta^2 \right) \right) \Gamma^5_h \\
+ \Gamma^2 \left( \Gamma^2 + 4\Delta^2 \right) \left( \Gamma^2 + 4\Delta^2 - 4\Omega^2 \right) + \Gamma^3 \delta_1^2 \delta_2^2 + \delta_2^2 \delta_1 \left( \Gamma^4 + 2\delta_2^2 \left( 3\Gamma^2 + 8\Delta^2 \right) + 4\Omega^2 \Delta^2 \right) \right) \\
- 2\Gamma^2 \Delta \Gamma_h \left( \Gamma^4 + \left( \delta_1 - \delta_2 \right) \left( \Gamma^2 - 4\Omega^2 \right) \right) + \Gamma^2 \left( -2\delta_1 \delta_2 \left( \Gamma^2 + 4\delta_1^2 + 4\Delta^2 - 4\Omega^2 \right) + \delta_1^2 \left( \Gamma^2 + 4\delta_1^2 + 4\Delta^2 \right) + \left( \Gamma^2 + 4\Delta^2 + 2\Omega^2 \right)^2 \right) \\
+ 4 \delta_1^2 \right) + \Gamma^2 \left( 4\Gamma^2 \Delta^2 \right) \Gamma^5_h \right) \right) + \Gamma \left( \delta_1 - \delta_2 \right) \left( \Gamma^2 + 4\Delta^2 \right) \Gamma^5_h \right) \\
+ \Delta \Gamma^2 \delta_2 \left( \Gamma^2 + 8\Delta^2 \right) + \Gamma^2 \delta_1^2 + \Gamma \left( \delta_1 \left( \Gamma^4 - 4\Delta^2 \right) + \left( \delta_1 - \delta_2 \right) \left( \delta_2 \left( \Gamma^2 + 4\Omega^2 \right) \right) + \delta_2 \left( \Gamma^4 + \Gamma^2 \delta_2^2 - 4\Omega^2 \left( \Gamma^2 + 2\Delta^2 \right) + 4\Omega^2 \Delta^2 \right) \right) \right) \\
- 8\Gamma^2 \Delta \Gamma_h \left( \delta_2 \left( \Gamma^2 + 4\Delta^2 \right) + \left( \delta_1 \left( \delta_1 - \delta_2 \right) \delta_2 + \Omega^2 \right) + \delta_2 \left( \Gamma^4 + \Gamma^2 \delta_2^2 - 4\Omega^2 \left( \Gamma^2 + 2\Delta^2 \right) + 4\Omega^2 \Delta^2 \right) \right) \right) \\
N_5 = \\
N_6 = \\
N_7 = \\
N_8 =
\[ -4\Omega^2 (4r^2\Delta (\delta_2 (\Gamma^2 + 4\Delta^2) + (\delta_1 - \delta_2) ((\delta_1 - \delta_2) \delta_2 + \Omega^2)) - (\Gamma^3 + 4\Gamma\Delta^2) \Gamma_h^3 \\
- 4\delta_2\Delta (\Gamma^2 + 4\Delta^2) \Gamma_h^3 + \Gamma_h (\Gamma^3 (\delta_1 - \delta_2)^2 + \Gamma (\Gamma^2 + 4\Delta^2) (\Gamma^2 + 2\Omega^2))), \]

\[ \mathcal{N}_0 = 8\Omega^2 (\Gamma^3 \Delta (\Gamma^2 + (\delta_1 - \delta_2)^2 + 4\Delta^2 + 2\Omega^2) - \Gamma^2 \Gamma_h (\delta_2 (\Gamma^2 + 4\Delta^2) + (\delta_1 - \delta_2) ((\delta_1 - \delta_2) \delta_2 + \Omega^2)) \\
+ \delta_2 (\Gamma^2 + 4\Delta^2) \Gamma_h^3 - \Gamma \Delta \Gamma_h^3 (\Gamma^2 + 4\Delta^2 + 2\Omega^2)). \]
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