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I. Exciton-Phonon Interaction in One-Exciton Absorption Spectra

Tiberius O. Cheche

University of Bucharest, Faculty of Physics, Department of Polymers Physics, P.O. Box 5211-Magurele R-76900, Romania, e-mail: cheche@digi.ro

Abstract: A formal expression of the one-exciton absorption coefficient is derived by a nonadiabatic treatment, which takes into account the exciton-phonon interaction. The approach is built in the Fermi Golden Rule and dipole approximation framework. It recovers the well-known results of the adiabatic approximation.

Keywords: Nanostructures; Electron-phonon interaction; Optical properties

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1. Introduction

Theoretically, the intrinsic response of one or several localized Wannier excitons was often invoked to explain the optical spectra of certain crystalline structures. For example, such excitonic pictures can explain the resonances observed experimentally in the quantum dots (QDs) structures; these resonances have been associated with either phonon-assisted transitions or Raman scattering. The adiabatic approach is the widely used approximation when the electron-phonon coupling is taken into account in such models. This approach implies an ineffective electron-phonon coupling, that is, no phonon-induced mixing between electronic levels; for such an ineffective coupling the scattering processes become more present in the optical spectra. When the exciton level spacing becomes comparable to the optical phonon energy (see, e.g., Refs. [1, 2]) the mixing of exciton states becomes important and the phonon-assisted absorption is a more probable process. In such a case, a weak exciton-phonon interaction demands a nonadiabatic approach [3, 4].

Here, we consider the phonon-assisted processes to describe the absorption spectra in the limit of low excitation intensities of laser light. By using a one-exciton picture, a nonadiabatic treatment, and the Fermi Golden Rule approach, an expression of the linear absorption coefficient is derived. In the adiabatic limit, the well-known expression of optical spectrum of a localized defect is recovered.

2. Theory

To describe the confined Wannier exciton, we use an extension of the Huang-Rhys treatment to *F* centers by using the Hamiltonian [5, 6]:

$$H = H_{ex} + H_{ph} + H_{ex-ph} \equiv \sum_i \varepsilon_i B_i^+ B_i + \sum_\alpha \hbar\omega_\alpha b_\alpha^+ b_\alpha + \sum_{\alpha,i,j} M_\alpha^{ij} B_i^+ B_j (b_\alpha^+ + b_\alpha). \quad (1)$$

$H_{ex}|i\rangle = \varepsilon_i |i\rangle$ with $|i\rangle$ the excitonic eigenfunction, ω_α is the frequency of α-th phononic mode, M_α^{ij} the exciton-phonon coupling between $|i\rangle$ and $|j\rangle$ states, B_i^+ (B_i) and b_α^+ (b_α) are the creation (annihilation) operators of exciton and phonon, respectively. Excitons are considered bosons for different states and fermions for the same state [6]. The radiation field is modeled as a single mode of linearly polarized plane wave. In the limit of linear response theory and long wave approximation the semiclassical exciton-field interaction can be written as [6, 7]

$$V_{\text{int}} = \frac{2iE_0}{\omega} \sum_{f \neq 0} \left[\omega_f \left(\boldsymbol{e}\boldsymbol{d}_f\right)\left(B_f^+ - B_f\right)\right] \sin\omega t \equiv W \sin\omega t, \quad (2)$$

where $\boldsymbol{e}$ is the polarization vector, ω the frequency, $2E_0$ the amplitude of light wave, m is the reduced effective excitonic mass, and $\omega_f = (\varepsilon_f - \varepsilon_0)/\hbar$; one assumes that the transition dipole matrix element between the excitonic vacuum state, $|0\rangle$ and state $|f\rangle$, $\boldsymbol{d}_f = e\langle f|\boldsymbol{r}|0\rangle$ is real, with e the electron charge, and $\boldsymbol{r}$ the coordinate of the electron-hole pair. For a final exciton-phonon state $H|n_F; f\rangle = E_{n_F f}|n_F; f\rangle \equiv E_{\mathrm{F}}|\mathrm{F}\rangle$. For the initial state of the system (ground electronic state plus phonons at thermal equilibrium), $H|n;0\rangle = E_n|n\rangle|0\rangle \equiv E_{\mathrm{G}}|\mathrm{G}\rangle$ and its density matrix is $\rho_{\mathrm{G}} = |0\rangle\rho_G\langle 0| \equiv |0\rangle\sum_n \left(|n\rangle\rho_n\langle n|\right)\langle 0|$; $|n\rangle$ are the eigenstates of phononic system at equilibrium, $H_{ph}|n\rangle = E_n|n\rangle$, and $\rho_n = \exp(-\beta E_n)/Tr\left[\exp(-\beta H_{ph})\right]$. For the absorption rate the Fermi Golden Rule applied to the eigenstates of the system gives

$$R_{abs} = \sum_{\mathrm{G,F}} \Omega_{\mathrm{G}\to\mathrm{F}} = \frac{2\pi}{\hbar}\sum_{\mathrm{G,F}}\left[\rho_{\mathrm{G}}|W_{\mathrm{G}\to\mathrm{F}}|^2 \delta(\hbar\omega + E_{\mathrm{G}} - E_{\mathrm{F}})\right] = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt e^{i\omega t}\langle 0|\left\langle \tilde{W}(t) W^+ \right\rangle_0 |0\rangle \quad (3)$$

where, $\Omega_{\mathrm{G}\to\mathrm{F}} = (2\pi/\hbar)\rho_{\mathrm{G}}|W_{\mathrm{G}\to\mathrm{F}}|^2 \delta(\hbar\omega + E_{\mathrm{G}} - E_{\mathrm{F}})$ is the transition rate $|\mathrm{G}\rangle \to |\mathrm{F}\rangle$, $W_{\mathrm{G}\to\mathrm{F}} \equiv \langle \mathrm{G}|W|\mathrm{F}\rangle = \langle 0|\langle n|W|n_F; f\rangle$, and $\left\langle \tilde{W}(t)W^+\right\rangle_0 \equiv Tr_G\left[\rho_G \tilde{W}(t) W^+\right]$ with $\tilde{W}(t) = e^{itH/\hbar} W e^{-itH/\hbar}$. Thus, with $\alpha(\omega) = 2\pi\hbar\omega R_{abs}/\left(ncE_0^2 V\right)$ [8] the linear one-exciton absorption coefficient reads

$$\alpha(\omega) = \frac{2\pi}{nc\hbar\omega V}\int_{-\infty}^{\infty} dt e^{i\omega t} \sum_{l,m\neq 0}\left[D_{lm}\left\langle \tilde{B}_l(t) B_m^+\right\rangle\right] \quad (4)$$

where V is the volume of absorptive system, n the refractive index, c the speed of light in vacuum, $D_{lm} \equiv \omega_l\omega_m(\boldsymbol{ed_l})(\boldsymbol{ed_m})$, $\tilde{B}_l(t) = e^{itH/\hbar} B_l e^{-itH/\hbar}$, and $\left\langle \tilde{B}_l(t) B_m^+ \right\rangle \equiv \langle 0|\left\langle e^{itH_{ph}/\hbar} B_l e^{-itH/\hbar} B_m^+\right\rangle_0 |0\rangle$.

One can easily see that $\left\langle e^{itH_{ph}/\hbar} B_l e^{-itH/\hbar} B_m^+ \right\rangle_0 = B_l e^{-itH_{ex}/\hbar} \left\langle U(t) \right\rangle_0 B_m^+ ;\;\; U(t) = e^{itH_0/\hbar} e^{-itH/\hbar}$, $H_0 = H_{ex} + H_{ph}$, and $\left\langle U(t) \right\rangle_0 = Tr_G \left[\rho_G e^{itH_0/\hbar} e^{-itH/\hbar} \right]$. With $\left\langle 0 \right| B_l e^{-itH_{ex}/\hbar} = e^{-it\omega_l} \left\langle 0 \right| B_l$ and the usual expansion of the evolution operator, $U(t)$, Eq. (4) becomes

$$\alpha(\omega) = \frac{2\pi}{nc\hbar\omega V} \sum_{l,m\neq 0} D_{lm} \int_{-\infty}^{\infty} dt e^{i(\omega-\omega_l)t} \left\langle 0 \right| B_l \left\langle \hat{T} \exp\left[-\frac{i}{\hbar} \int_0^t dt_1 \tilde{V}(t_1) \right] \right\rangle_0 B_m^+ \left| 0 \right\rangle \tag{5}$$

where $\hat{T}$ is the time ordered operator, $\tilde{V}(t) = \exp(itH_0/\hbar) H_{ex-ph} \exp(-itH_0/\hbar)$, and we write the expansion as the sum of different orders, $U(t) = U_0(t) + U_1(t) + U_2(t) + \dots$. The linear coupling in the phononic operators from Hamiltonian (1) gives $\left\langle 0 \right| B_l \left\langle U_{2n+1}(t) \right\rangle_0 B_m^+ \left| 0 \right\rangle = 0$. For the first two terms of $U(t)$, we obtain $\left\langle 0 \right| B_l \left\langle U_0(t) \right\rangle_0 B_m^+ \left| 0 \right\rangle = \delta_{lm}$ and

$$\left\langle U_2(t) \right\rangle_0 = -\frac{1}{2!\hbar^2} \int_0^t dt_1 \int_0^t dt_2 \hat{T} \sum_{\substack{\alpha_1, i_1, j_1 \\ \alpha_2, i_2, j_2}} \left(M_{\alpha_1}^{i_1 j_1} M_{\alpha_2}^{i_2 j_2} B_{i_1^+ j_1}^{t_1} B_{i_2^+ j_2}^{t_2} \left\langle u_{\alpha_1}(t_1) u_{\alpha_2}(t_2) \right\rangle_0 \right),$$

where $B_{i^+ j}^t \equiv B_i^+ B_j \exp(it\omega_{ij}/\hbar)$ and $u_\alpha(t) \equiv \exp(itH_{ph}/\hbar)(b_\alpha^+ + b_\alpha)\exp(-itH_{ph}/\hbar)$. With (see, e.g., Ref. [9])

$$\begin{aligned} \hat{T} \left\langle u_{\alpha_1}(t_1) u_{\alpha_2}(t_2) \right\rangle_0 &= \delta_{\alpha_1\alpha_2} \left[\overline{N}_\alpha \exp\left(i\omega_{\alpha_1} \left| t_1 - t_2 \right| \right) + (\overline{N}_\alpha + 1) \exp\left(-i\omega_{\alpha_1} \left| t_1 - t_2 \right| \right) \right] \\ &\equiv \delta_{\alpha_1\alpha_2} D^0(\alpha_1, \left| t_1 - t_2 \right|) \end{aligned}$$

one obtains

$$\begin{aligned} &\left\langle 0 \right| B_l \left\langle U_2(t) \right\rangle_0 B_m^+ \left| 0 \right\rangle \\ &= \frac{-1}{2!\hbar^2} \sum_{\alpha,i} \left[M_\alpha^{li} M_\alpha^{im} \int_0^t dt_1 \int_0^t dt_2 \exp(it_1\omega_{li}) \exp(it_2\omega_{im}) D^0(\alpha_1, \left| t_1 - t_2 \right|) \right], \end{aligned} \tag{6a}$$

or more explicitly

$$\begin{aligned} \left\langle 0 \right| B_l \left\langle U_2(t) \right\rangle_0 B_m^+ \left| 0 \right\rangle = -\frac{1}{\hbar^2} \sum_{\alpha,i} \Big\{ M_\alpha^{li} M_\alpha^{im} \int_0^t dt_1 \int_0^{t_1} dt_2 \\ \times \Big\{ \left[\exp[(it_1(\omega_{li} + \omega_\alpha)] \exp[(it_2(\omega_{im} - \omega_\alpha)] + \exp[(it_1(\omega_{im} + \omega_\alpha)] \exp[(it_2(\omega_{li} - \omega_\alpha)] \right] \overline{N}_\alpha \\ + \left[\exp[(it_1(\omega_{li} - \omega_\alpha)] \exp[(it_2(\omega_{im} + \omega_\alpha)] + \exp[(it_1(\omega_{im} - \omega_\alpha)] \exp[(it_2(\omega_{li} + \omega_\alpha)] \right] (\overline{N}_\alpha + 1) \Big\} \end{aligned}, \tag{6b}$$

where $\overline{N}_\alpha$ is the phononic occupation number. The second order term (Eqs. 6a, b) involves transition $\left| l \right\rangle \to \left| i \right\rangle \to \left| m \right\rangle$. For the next orders, we apply the Wick's theoreme directly to u_α [10]. The fourth order term, which involves transition $\left| l \right\rangle \to \left| i_2 \right\rangle \to \left| i_3 \right\rangle \to \left| i_4 \right\rangle \to \left| m \right\rangle$, reads

$$\begin{aligned}&\left\langle 0\left|B_l\left\langle U_4(t)\right\rangle_0 B_m^+\right|0\right\rangle \\ &=\frac{1}{\hbar^2 4!}\sum_{i_2,i_3,i_4}\left\{\sum_\alpha M_\alpha^{li_2}M_\alpha^{i_2i_3}\int_0^t dt_1\int_0^t dt_2\exp[i(\omega_{li_2}t_1+\omega_{i_2i_3}t_2)]D^0(\alpha,|t_1-t_2|)\right. \\ &\times\sum_\alpha M_\alpha^{i_3i_4}M_\alpha^{i_4m}\int_0^t dt_1\int_0^t dt_2\exp[i(\omega_{i_3i_4}t_1+\omega_{i_4m}t_2)]D^0(\alpha,|t_1-t_2|) \\ &+\sum_\alpha M_\alpha^{li_2}M_\alpha^{i_3i_4}\int_0^t dt_1\int_0^t dt_2\exp[i(\omega_{li_2}t_1+\omega_{i_3i_4}t_2)]D^0(\alpha,|t_1-t_2|) \\ &\times\sum_\alpha M_\alpha^{i_2i_3}M_\alpha^{i_4i_m}\int_0^t dt_1\int_0^t dt_2\exp[i(\omega_{i_2i_3}t_1+\omega_{i_4m}t_2)]D^0(\alpha,|t_1-t_2|) \\ &+\sum_\alpha M_\alpha^{li_2}M_\alpha^{i_4m}\int_0^t dt_1\int_0^t dt_2\exp[i(\omega_{li_2}t_1+\omega_{i_4m}t_2)]D^0(\alpha,|t_1-t_2|) \\ &\left.\times\sum_\alpha M_\alpha^{i_2i_3}M_\alpha^{i_3i_4}\int_0^t dt_1\int_0^t dt_2\exp[i(\omega_{i_2i_3}t_1+\omega_{i_3i_4}t_2)]D^0(\alpha,|t_1-t_2|)\right\}\end{aligned} \tag{7}$$

In Eq. (7) the Wick's theorem for bosons couples the exponential excitonic terms in $(2\cdot 2-1)(2\cdot 2-3)$ products of double integrals. Generally, the 2*n*-th order term contains sums over products of *n* double integrals of similar form with those from Eqs. (6b) and (7). Thus

$$\begin{aligned}&\left\langle 0\left|B_l\left\langle U_{2n}(t)\right\rangle_0 B_m^+\right|0\right\rangle \\ &=\left(\frac{-i}{\hbar}\right)^{2n}\frac{1}{(2n)!}\sum_{\substack{all\ possible\\ combinations}}{}'\left\{\sum_\alpha D(\alpha,i_1^{(1)},i_2^{(1)},i_3^{(1)},i_4^{(1)},\left|t_1^{(1)}-t_3^{(1)}\right|)\right., \\ &\times\sum_\alpha D(\alpha,i_1^{(m)},i_2^{(m)},i_3^{(m)},i_4^{(m)},\left|t_1^{(m)}-t_3^{(m)}\right|)\end{aligned} \tag{8}$$

where the sum is over all the $(2n-1)(2n-3)...3\cdot 1$ possible combinations in accordance with the Wick's theorem for bosons; the combination $i_1^{(1)},i_2^{(1)},i_3^{(1)},i_4^{(1)},...i_1^{(m)},i_2^{(m)},i_3^{(m)},i_4^{(m)}$ is one of the possible combinations of $l,i_2,i_3,...,i_{2m},m$ (indexes for the excitonic eigenvalues), the prime means the summation excludes *l* and *m*, and

$$\begin{aligned}&D(\alpha,i_1^{(1)},i_2^{(1)},i_3^{(1)},i_4^{(1)},\left|t_1^{(1)}-t_3^{(1)}\right|)\equiv M_\alpha^{i_{k1}i_{k2}}M_\alpha^{i_{k3}i_{k4}} \\ &\times\int_0^t dt_{k1}\int_0^t dt_{k2}\left[\exp[it_{k1}(\omega_{i_{k1}}-\omega_{i_{k2}})]\exp[it_{k3}(\omega_{i_{k3}}-\omega_{i_{k4}})]D^0(\alpha,|t_{k1}-t_{k3}|)\right.\end{aligned}.$$

The absorption spectrum can be computed now for different orders. The zeroth order term, $U_0(t)=1$, gives the excitonic spectrum in the vanishing coupling case.

3. Discussions

When the adiabatic limit is of interest the mixing between excitonic states is absent, that is, $H_{ex-ph} = \sum_{\alpha,i} M_\alpha^{ii} B_i^+ B_i (b_\alpha^+ + b_\alpha)$. The analytical form deduced from our results has similarities with the well-known expressions for optical spectrum of a localized defect with several electronic states in the independent boson approach**.** Thus

$$\langle 0 | B_l \langle U_{2n}(t) \rangle_0 B_m^+ | 0 \rangle = \frac{1}{n!} \left[\sum_\alpha \left(-\Phi_\alpha^l(t) \right) \right]^n \delta_{lm}, \tag{9}$$

where $\Phi_\alpha^l(t) = \left(M_\alpha^{ll} / \hbar\omega_\alpha \right)^2 \left[(\overline{N}_\alpha + 1)(1 - e^{-i\omega_\alpha t}) + \overline{N}_\alpha (1 - e^{-i\omega_\alpha t}) - i\omega_\alpha t \right]$ and the absorption coefficient reads

$$\begin{aligned} \alpha(\omega) &= \frac{2\pi}{nc\hbar\omega V} \sum_{l,m} \omega_l^2 (\boldsymbol{ed}_l)^2 \int_{-\infty}^{\infty} dt e^{i(\omega-\omega_l)t} \left[1 + \sum_{n=1} \frac{1}{n!} \left[\sum_\alpha \left(-\Phi_\alpha^l(t) \right) \right]^n \right] \\ &= \frac{2\pi}{nc\hbar\omega V} \sum_{l,m} \omega_l^2 (\boldsymbol{ed}_l)^2 \int_{-\infty}^{\infty} dt e^{i(\omega-\omega_l)t} \exp\left[\sum_\alpha \left(-\Phi_\alpha^l(t) \right) \right] \end{aligned} \tag{10}$$

Denoting $\left(M_\alpha^{ll} / \hbar\omega_\alpha \right)^2 \equiv g_\alpha^{(l)}$ and following Ref. [9], we have $i\omega_\alpha t + \sum_\alpha \Phi_\alpha^l(t) = i[\omega_\alpha - \Delta_l]t + \Phi_l(t)$; $\Delta_l = \sum_\alpha \left(g_\alpha^{(l)} \omega_\alpha \right)$ is the renormalization induced by phonons and $\Phi_l(t) = \sum_\alpha \left\{ g_\alpha^{(l)} \left[(\overline{N}_\alpha + 1)(1 - e^{-i\omega_\alpha t}) + \overline{N}_\alpha (1 - e^{-i\omega_\alpha t}) \right] \right\}$.

When our program is applied to the Einstein model, where $\omega_\alpha = \omega_0$, and $\Delta_l = \omega_0 \sum_\alpha g_\alpha^{(l)} \equiv \omega_0 g_l$ (with g_l the Huang-Rhys factor), we obtain

$$\begin{aligned} \alpha(\omega) &= \frac{4\pi^2}{nc\hbar\omega V} \sum_{l \neq 0} \left\{ \omega_l^2 (\boldsymbol{ed}_l)^2 \exp[-g_l \coth(\beta\hbar\omega_0/2)] \right\} \\ &\times \sum_{n=-\infty}^{\infty} I_n \left(g_l / \sinh(\beta\hbar\omega_0/2) \right) \exp(n\beta\hbar\omega_0/2) \delta(\omega - \omega_l + \Delta_l - n\omega_0) \end{aligned} \tag{12}$$

where I_n are the Bessel functions**.** Both side bands are present in the spectra and the function $\exp(n\beta\hbar\omega_0/2)$ shifts the maximum of the intensity envelope to positive side. The positive LO phononic bands correspond to photoluminescence excitation (PLE) spectra obtained at low temperatures (see, e,g. Ref.[11]). The relative intensity of lines is given by the coefficients of Dirac delta function, which arise in expression of the absorption coefficient. Comparatively to Ref. [12], where the absorption coefficient is expressed in the adiabatic framework too, Eqs. (10) and (11) include the influence of the dipole orientation on absorption spectra. The transition dipole matrix and exciton-phonon coupling matrix elements are necessary ingredients to compute absorption spectra. For example for different QD models they have been obtained within the effective mass approximation or by using the pseudopotential approach.

Excepting the divergences induced by resonance conditions, the Fourier transform of double integrals with the structure of Eqs. (6c) and (7) yields the intensities of absorption lines. For example, the divergences which appear in Eq. (6c) may be treated as follows: i) For $l = i \neq m$ and $l \neq i = m$ the divergence occurs for exact matching between the exciton level spacing and phonon energy, i.e., $\omega_{im} + \omega_\alpha = 0$ or $\omega_{im} - \omega_\alpha = 0$, and $\omega_{li} + \omega_\alpha = 0$ or $\omega_{li} - \omega_\alpha = 0$, respectively. To avoid it, we consider the definition (width) of the line of frequency ω_0 within the line width Γ. Then, assuming the coupling factors M_α^{ij} constant for this frequency interval centered on ω_0 (Γ is of order of several to tens μeV at low temperatures for QDs), we may write $\alpha(\omega_0) = \Gamma^{-1}\int_{\omega_0-\Gamma/2}^{\omega_0+\Gamma/2} d\omega\alpha(\omega)$, and the Fourier transforms of functions proportional to time give the approximate intensities now. For example, for $l = i \neq m$ and $\omega_{im} + \omega_\alpha = 0$ the second double integral from Eq. (6c) gives a term $-it/\omega_\alpha$, which yields a line intensity proportional to $-i\int_{-\infty}^{\infty} \exp[i(\omega - \omega_l + \omega_0)t]\sin(\Gamma t/2)$. ii) The cases $l = m \neq i$ and $l \neq m \neq i$ may be treated similarly. If $\omega_{li} \pm \omega_\alpha \neq 0$ and $\omega_{li} >> \omega_\alpha$ or $\omega_{li} << \omega_\alpha$ convergence is established (after the time integration the terms that are proportional to *t* approximately vanish). For the situation $\omega_{li} \pm \omega_\alpha = 0$, we take the main contribution to the t_2 integrals coming from large t_2; thus integration over t_2 yields i/Γ and after that, we again apply the approximation proposed at i).

In conclusion, the present theoretical treatment provides a method to calculate the phononic influence on the one-excitonic spectra in limit of low intensity of light. By specifying the exciton-phonon coupling type, the treatment may take into account both acoustic and LO phonons. As a straight application of our treatment, by modeling the defect-free QDs as one-exciton molecules the absorption spectra at low temperatures of such structures can be obtained, and in accordance with Ref. [13] they may be used to approximate their PLE spectra.

II. AUXILIARY NOTES

a) For Eq. (2) :

In the limit of linear response model the interaction between a system of charges q_i (with masses m_i, coordinates $\boldsymbol{r}_i$ and momenta $\boldsymbol{p}_i$) and the field given by $\boldsymbol{E}(\boldsymbol{r},t) = 2\boldsymbol{e}E_0 \cos(\boldsymbol{kr} - \omega t) \equiv \boldsymbol{e}\left[Ee^{-i\omega t} + E^* e^{-i\omega t}\right]$ is

$$V_{\text{int}}(\boldsymbol{r_i},t) = -\sum_i \frac{q_i}{m_i c} \boldsymbol{A}(\boldsymbol{r_i},t)\boldsymbol{p}_i$$

(A1)

with

$$\boldsymbol{A}(\boldsymbol{r_i},t) = \boldsymbol{A}_0 \sin(\boldsymbol{kr_i} - \omega t) \text{ and } \boldsymbol{A}_0 = 2c\boldsymbol{e}E_0/\omega$$

(A2)

the vector potential. Translating the problem into the effective mass picture for the electron (effective mass m_e and coordinate $\boldsymbol{r}_e$) - hole (effective mass m_h and coordinate $\boldsymbol{r}_h$) pair case, introduction of the center of mass $\xi = (m_e\boldsymbol{r}_e + m_h\boldsymbol{r}_h)/(m_e + m_h)$ and relative coordinates of the pair $\rho = \boldsymbol{r}_h - \boldsymbol{r}_e$ gives

$$V_{\text{int}}(\boldsymbol{r_e},\boldsymbol{r_h},t) = -\frac{e}{c}\left\{\left[\boldsymbol{A}(\boldsymbol{r_h},t) - \boldsymbol{A}(\boldsymbol{r_e},t)\right]\frac{\boldsymbol{p}_\xi}{M} + \left[\frac{\boldsymbol{A}(\boldsymbol{r_h},t)}{m_h} + \frac{\boldsymbol{A}(\boldsymbol{r_e},t)}{m_e}\right]\boldsymbol{p}_\rho\right\}$$

(A3)

where $M = m_e + m_h$, and $\boldsymbol{p}_\xi$ and $\boldsymbol{p}_\rho \equiv \boldsymbol{p}$ are the momentum of center of mass and of relative motion, respectively. In the long wave approximation, $\boldsymbol{k}\rho << 1$, and consequently $\boldsymbol{A}(\boldsymbol{r_h},t) \approx \boldsymbol{A}(\boldsymbol{r_e},t) \equiv \boldsymbol{A}(\boldsymbol{r},t)$; from Eqs. (A2) and (A3) with $\boldsymbol{k} \approx 0$ one obtains

$$V_{\text{int}}(t) = \frac{2eE_0}{m\omega}(\boldsymbol{ep})\sin\omega t$$

In the second quantization [6]

$$\boldsymbol{ep} \to \sum_{f\neq 0}\left[\langle 0|\boldsymbol{ep}|f\rangle B_f + \langle f|\boldsymbol{ep}|0\rangle B_f^+\right]$$

and the transformation from $\boldsymbol{p}$ to $\boldsymbol{r}$ dependence yields Eq. (2).

b) For Eq. (3):

$$\begin{aligned}
&Tr_{\mathsf{G}}\left\{\sum_{\mathsf{F}}\left[\rho_{\mathsf{G}}\left|W_{\mathsf{G}\to\mathsf{F}}\right|^2\delta\left(\hbar\omega+E_{\mathsf{G}}-E_{\mathsf{F}}\right)\right]\right\}\\
&=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dte^{i\omega t}Tr_{\mathsf{G}}\left\{\rho_{\mathsf{G}}\sum_{\mathsf{F}}\left[\langle\mathsf{G}|e^{itH/t}We^{-itH/t}|\mathsf{F}\rangle\langle\mathsf{F}|W^+|\mathsf{G}\rangle\right]\right\}\\
&=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dte^{i\omega t}\sum_m\left[(\rho_{\mathsf{G}})_{mm}\left(\langle\mathsf{G}|\tilde{W}(t)W^+|\mathsf{G}\rangle\right)_{mm}\right]\\
&=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dte^{i\omega t}\sum_m\left[\langle m|\langle 0|\left(\sum_n|0\rangle|n\rangle\rho_n\langle n|\langle 0|\right)|0\rangle|m\rangle\langle m|\langle 0|\tilde{W}(t)W^+|0\rangle|m\rangle\right]\\
&=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dte^{i\omega t}\sum_m\left[\rho_m\langle m|\langle 0|\tilde{W}(t)W^+|0\rangle|m\rangle\right]\\
&=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dte^{i\omega t}\langle 0|Tr_G\left[\rho_G\tilde{W}(t)W^+\right]|0\rangle
\end{aligned}$$

(A4)

c) For Eq. (4) we used

i) $\langle\mathsf{G}|e^{itH/\hbar}=\langle\mathsf{G}|e^{itH_{ph}/\hbar}$, which can be seen as follows:

$$e^{-itH/\hbar}|\mathsf{G}\rangle=e^{-it(H_{ex}+H_{ex-ph}+H_{ph})/\hbar}|0\rangle|n\rangle$$

(A5)

and with the identity $e^{A+B}=e^Ae^Be^{-[A,B]/2}$ we may write

$$e^{-it(H_{ex}+H_{ex-ph}+H_{ph})/\hbar} = e^{-it(H_{ex},H_{ex-ph})/\hbar} e^{-itH_{ph}/\hbar} e^{t^2[H_{ex-ph},H_{ph}]/2\hbar^2}$$

(A6)

But

$$\left[H_{ex-ph}, H_{ph}\right] = \left[\sum_{\alpha,i,j} M_{\alpha}^{ij} B_i^+ B_j (b_{\alpha}^+ + b_{\alpha}), \sum_{\beta} \hbar\omega_{\beta} b_{\beta}^+ b_{\beta}\right]$$
$$= \sum_{\alpha,i,j} M_{\alpha}^{ij} (b_{\alpha}^+ + b_{\alpha}) B_i^+ B_j$$

(A7)

and consequently, with a series expansion and $B_j\left|0\right\rangle = 0$, we have

$$e^{t^2[H_{ex-ph},H_{ph}]/2\hbar^2}\left|n\right\rangle\left|0\right\rangle = \left|n\right\rangle\left|0\right\rangle$$

(A8)

With Eqs. (A8) and (A6), Eq. (A5) reads

$$e^{-itH/\hbar}\left|0\right\rangle\left|n\right\rangle = e^{-it(H_{ex}+H_{ex-ph})/\hbar}\left|0\right\rangle e^{-itH_{ph}/\hbar}\left|n\right\rangle$$

(A9)

from which again with $e^{A+B} = e^A e^B e^{-[A,B]/2}$, we have

$$e^{-it(H_{ex}+H_{ex-ph})/\hbar}\left|0\right\rangle = e^{-itH_{ex}/\hbar} e^{-itH_{ex-ph}/\hbar} e^{t^2(H_{ex},H_{ex-ph})/2\hbar^2}\left|0\right\rangle$$

(A10)

Next, we used in accordance with the excitonic commutation relations (see d) too)

$$\left[\sum_l \varepsilon_l B_l^+ B_l, B_i^+ B_j\right] = \left(\varepsilon_i - \varepsilon_j\right) B_i^+ B_j$$

(A11)

Thus

$$\left[H_{ex}, H_{ex-ph}\right] = \left[\sum_l \varepsilon_l B_l^+ B_l, \sum_{\alpha,i,j} M_\alpha^{ij} B_i^+ B_j (b_\alpha^+ + b_\alpha)\right]$$
$$= \sum_{\alpha,l,i,j} \varepsilon_l M_\alpha^{ij} (b_\alpha^+ + b_\alpha)\left[B_l^+ B_l, B_i^+ B_j\right]$$
$$= \sum_{\alpha,i,j} M_\alpha^{ij} (\varepsilon_i - \varepsilon_j)(b_\alpha^+ + b_\alpha) B_i^+ B_j$$

(A12)

and consequently

$$e^{-itH_{ex}/\hbar} e^{-itH_{ex-ph}/\hbar} e^{t^2 (H_{ex}, H_{ex-ph})/2\hbar^2} \left|0\right\rangle = e^{-itH_{ex}/\hbar} e^{-itH_{ex-ph}/\hbar}\left|0\right\rangle = e^{-itH_{ex}/\hbar}\left|0\right\rangle = \left|0\right\rangle$$

(A13)

The equalities in Eq. (A13) are obtained by using a series expansion of the exponential and $B_j\left|0\right\rangle = 0$. Finally, Eqs. (A9) and (A13) give $\left\langle \mathrm{G}\right| e^{itH/\hbar} = \left\langle \mathrm{G}\right| e^{itH_{ph}/\hbar}$.

ii) Relation $\left\langle \tilde{B}_l^+(t) B_m \right\rangle = 0$, which holds because $\left\langle \tilde{B}_l^+(t) B_m \right\rangle = Tr_G\left[\rho_G \left\langle 0\right| \tilde{B}_l^+(t) B_m \left|0\right\rangle\right]$ and $B_m\left|0\right\rangle = 0$. $\left\langle \tilde{B}_l(t) B_m \right\rangle = 0$, similarly. Also, $\left\langle \tilde{B}_l^+(t) B_m \right\rangle = 0$. To prove the last equality, we use $\left\langle \mathrm{G}\right| e^{itH/\hbar} = \left\langle \mathrm{G}\right| e^{itH_{ph}/\hbar}$ and $\left\langle 0\right| B_m^+ = 0$. Thus, we have

$$\left\langle \tilde{B}_l^+(t) B_m^+ \right\rangle = \sum_n \left[\rho_n \left\langle n\right| \left\langle 0\right| e^{itH/\hbar} B_l^+ e^{-itH/\hbar} B_m^+ \left|0\right\rangle \left|n\right\rangle\right]$$
$$= \sum_n \left[\rho_n \left\langle n\right| \left\langle 0\right| e^{itH_{ph}/\hbar} B_l^+ e^{-itH/\hbar} B_m^+ \left|0\right\rangle \left|n\right\rangle\right]$$
$$= \sum_n \left[\rho_n \left\langle n\right| \left\langle 0\right| B_l^+ e^{itH_{ph}/\hbar} e^{-itH/\hbar} B_m^+ \left|0\right\rangle \left|n\right\rangle\right] = 0$$

(A14)

The term $\tilde{B}_l(t) B_m^+ \left|0\right\rangle \left|n\right\rangle$ means that at the initial moment the field, by B_m^+, creates the excitonic state $\left|m\right\rangle = B_m^+\left|0\right\rangle$ and then at moment *t* the field, by $\tilde{B}_l(t)$, destroys an exciton in state $\left|l\right\rangle$. Transition $\left|l\right\rangle \to \left|m\right\rangle$ involves an emission or absorption of phonon between

t_0 and t. Actually, the present treatment refines the calculus of $\left\langle \widetilde{B}_l(t) B_m^+ \right\rangle$ by including intermediates states (moments) between $\left| l \right\rangle$ and $\left| m \right\rangle$ (t_0 and t).

d) Proof of the equality $\left\langle e^{itH_{ph}/\hbar} B_l e^{-itH/\hbar} B_m^+ \right\rangle_0 = B_l e^{-itH_{ex}/\hbar} \left\langle U(t) \right\rangle_0 B_m^+$ from pp. 5, row 7 is as follows

$$\left\langle e^{itH_{ph}/\hbar} B_l e^{-itH/\hbar} B_m^+ \right\rangle_0 = \left\langle B_l e^{-itH_{ex}/\hbar} e^{itH_{ex}/\hbar} e^{itH_{ph}/\hbar} e^{-itH/\hbar} B_m^+ \right\rangle_0$$
$$= B_l e^{-itH_{ex}/\hbar} \left\langle e^{itH_{ex}/\hbar} e^{itH_{ph}/\hbar} e^{-itH/\hbar} \right\rangle_0 B_m^+ \equiv B_l e^{-itH_{ex}/\hbar} \left\langle U(t) \right\rangle_0 B_m^+$$

(A15a)

Proof of the equality $\left\langle 0 \right| B_l e^{-itH_{ex}/\hbar} = e^{-it\omega_l} \left\langle 0 \right| B_l$ from pp. 5, row 8 is as follows

$$\left\langle 0 \right| B_l e^{-itH_{ex}/\hbar} = \left\langle 0 \right| B_l \left[1 - it \sum_j \varepsilon_j B_j^+ B_j + \frac{(-it)^2}{2!} \left(\sum_j \varepsilon_j B_j^+ B_j \right)^2 + \ldots \right]$$

(A15b)

and in accordance with the excitonic commutation relations

$$\left\langle 0 \right| B_l \sum_j \varepsilon_j B_j^+ B_j = \sum_{j \neq l} (\varepsilon_j \left\langle 0 \right| B_l B_j^+ B_j) + \varepsilon_l \left\langle 0 \right| B_l B_l^+ B_l = 0 + \varepsilon_l \left\langle 0 \right| \left(1 - B_l^+ B_l\right) B_l = \left\langle 0 \right| B_l \varepsilon_l$$

hence,

$$\left\langle 0 \right| B_l e^{-itH_{ex}/\hbar} = \left\langle 0 \right| B_l \left[1 - it\omega_l + \frac{(-it\omega_l)^2}{2!} + \ldots \right] = \left\langle 0 \right| B_l e^{-it\omega_l}$$

e) The even term $\left\langle U_2(t)\right\rangle_0$ from pp. 5, last row is obtained as follows

$$\left\langle U_2(t)\right\rangle_0 = -\frac{1}{2!\hbar^2}\int_0^t dt_1\int_0^t dt_2 \hat{T}\left\langle \widetilde{V}(t_1)\widetilde{V}(t_2)\right\rangle_0$$
$$= -\frac{1}{2!\hbar^2}\int_0^t dt_1\int_0^t dt_2 \hat{T}\left\langle e^{it_1H_{ex}/\hbar}e^{it_1H_{ph}/\hbar}\sum_{\alpha_1,i_1,j_1}M_{\alpha_1}^{i_1j_1}B_{i_1}^{+}B_{j_1}(b_{\alpha_1}^{+}+b_{\alpha_1})e^{-it_1H_{ex}/\hbar}e^{-it_1H_{ph}/\hbar}\right.$$
$$\left.\times e^{it_2H_{ex}/\hbar}e^{it_2H_{ph}/\hbar}\sum_{\alpha_2,i_2,j_2}M_{\alpha_2}^{i_2j_2}B_{i_2}^{+}B_{j_2}(b_{\alpha_2}^{+}+b_{\alpha_2})e^{-it_2H_{ex}/\hbar}e^{-it_2H_{ph}/\hbar}\right\rangle_0$$
$$= -\frac{1}{2!\hbar^2}\int_0^t dt_1\int_0^t dt_2 \hat{T}\sum_{\alpha_1,i_1,j_1}M_{\alpha_1}^{i_1j_1}e^{it_1H_{ex}/\hbar}B_{i_1}^{+}B_{j_1}e^{-it_1H_{ex}/\hbar}\sum_{\alpha_2,i_2,j_2}M_{\alpha_2}^{i_2j_2}e^{it_2H_{ex}/\hbar}B_{i_2}^{+}B_{j_2}e^{-it_2H_{ex}/\hbar}$$
$$\times\left\langle e^{it_1H_{ph}/\hbar}(b_{\alpha_1}^{+}+b_{\alpha_1})e^{-it_1H_{ph}/\hbar}e^{it_1H_{ph}/\hbar}(b_{\alpha_2}^{+}+b_{\alpha_2})e^{-it_1H_{ph}/\hbar}\right\rangle_0$$
$$= -\frac{1}{2!\hbar^2}\int_0^t dt_1\int_0^t dt_2 \hat{T}\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2}}\left(M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}B_{i_1^{+}j_1}^{t_1}B_{i_2^{+}j_2}^{t_2}\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right)$$

(A16)

f) The even term $\left\langle 0\middle|B_l\left\langle U_2(t)\right\rangle_0 B_m^{+}\middle|0\right\rangle$ of Eq. (6a) is obtained as follows

$$\begin{aligned}
&\left\langle 0\left|B_l\left\langle U_2(t)\right\rangle_0 B_m^+\right|0\right\rangle \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\int_0^t dt_1\int_0^t dt_2\hat{T}\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2}}\left(M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}\left\langle 0\left|B_l B_{i_1^+j_1}^{t_1}B_{i_2^+j_2}^{t_2}B_m^+\right|0\right\rangle\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\int_0^t dt_1\int_0^t dt_2\hat{T}\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2}}\left(M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}e^{it_1(\varepsilon_{i_1}-\varepsilon_{j_1})}e^{it_2(\varepsilon_{i_2}-\varepsilon_{j_2})}\left\langle 0\left|B_l B_{i_1}^+B_{j_1}B_{i_2}^+B_{j_2}B_m^+\right|0\right\rangle\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\int_0^t dt_1\int_0^t dt_2\hat{T}\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2}}\left(M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}e^{it_1(\varepsilon_{i_1}-\varepsilon_{j_1})}e^{it_2(\varepsilon_{i_2}-\varepsilon_{j_2})}\delta_{li_1}\delta_{j_1i_2}\delta_{j_2m}\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\sum_{\alpha_1,\alpha_2,i}\left(M_{\alpha_1}^{li}M_{\alpha_2}^{im}\int_0^t dt_1\int_0^t dt_2 e^{it_1\omega_{li}}e^{it_2\omega_{im}}T\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&\equiv\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\sum_{\alpha,i}\left(M_{\alpha}^{li}M_{\alpha}^{im}\int_0^t dt_1\int_0^t dt_2 e^{it_1\omega_{li}}e^{it_2\omega_{im}}D^0(\alpha,\left|t_1-t_2\right|)\right)
\end{aligned}$$

(A17)

where the equality $\left\langle 0\left|B_l B_{i_1}^+B_{j_1}B_{i_2}^+B_{j_2}B_m^+\right|0\right\rangle=\delta_{li_1}\delta_{j_1i_2}\delta_{j_2m}$ derived by using the excitonic commutation relations has been used. The integral in (A17) is written as follows

$$\begin{aligned}
&\int_0^t dt_1\int_0^t dt_2 e^{it_1\omega_{ij}}e^{it_2\omega_{lm}}D^0(\alpha,\left|t_1-t_2\right|)=\int_0^t dt_1\int_0^{t_1} dt_2 e^{it_1\omega_{ij}}e^{it_2\omega_{lm}}D^0(\alpha,t_1>t_2) \\
&+\int_0^t dt_2\int_0^{t_2} dt_1 e^{it_1\omega_{ij}}e^{it_2\omega_{lm}}D^0(\alpha,t_1<t_2) \\
&=\int_0^t dt_1\int_0^{t_1} dt_2 e^{it_1\omega_{ij}}e^{it_2\omega_{lm}}\left[\overline{N}_\alpha^{\,i\omega_\alpha(t_1-t_2)}+(\overline{N}_\alpha+1)e^{-i\omega_\alpha(t_1-t_2)}\right] \\
&+\int_0^t dt_1\int_0^{t_1} dt_2 e^{it_1\omega_{ij}}e^{it_2\omega_{lm}}\left[\overline{N}_\alpha e^{-i\omega_\alpha(t_1-t_2)}+(\overline{N}_\alpha+1)e^{i\omega_\alpha(t_1-t_2)}\right]
\end{aligned}$$

(A18)

g) The even term $\left\langle U_4(t)\right\rangle_0$ from Eq. (7) is obtained with Eq. (A19) by using the Wick's theorem as follows

$$\left\langle U_4(t)\right\rangle_0=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\hat{T}\left\langle \tilde{V}(t_1)\tilde{V}(t_2)\tilde{V}(t_3)\tilde{V}(t_4)\right\rangle_0$$
$$=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\hat{T}$$
$$\times\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2\\ \alpha_3,i_3,j_3\\ \alpha_4,i_4,j_4}}\left(M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}M_{\alpha_3}^{i_3j_3}M_{\alpha_4}^{i_4j_4}B_{i_1^+j_1}^{t_1}B_{i_2^+j_2}^{t_2}B_{i_3^+j_3}^{t_3}B_{i_4^+j_4}^{t_4}\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)u_{\alpha_3}(t_3)u_{\alpha_4}(t_4)\right\rangle_0\right)$$
$$=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\hat{T}$$
$$\times\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2\\ \alpha_3,i_3,j_3\\ \alpha_4,i_4,j_4}}\left\{M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}M_{\alpha_3}^{i_3j_3}M_{\alpha_4}^{i_4j_4}B_{i_1}^+B_{j_1}B_{i_2}^+B_{j_2}B_{i_3}^+B_{j_3}B_{i_4}^+B_{j_4}\right.$$
$$\left\langle e^{it_1(\varepsilon_{i_1}-\varepsilon_{j_1})}u_{\alpha_1}(t_1)e^{it_2(\varepsilon_{i_2}-\varepsilon_{j_2})}u_{\alpha_2}(t_2)e^{it_3(\varepsilon_{i_3}-\varepsilon_{j_3})}u_{\alpha_3}(t_3)e^{it_4(\varepsilon_{i_4}-\varepsilon_{j_4})}u_{\alpha_4}(t_4)\right\rangle_0$$

(A19)

h) The even term $\left\langle 0\right|B_l\left\langle U_4(t)\right\rangle_0 B_m^+\left|0\right\rangle$ of Eq. (7) is obtained as follows

$$\left\langle 0\right|B_l\left\langle U_4(t)\right\rangle_0 B_m^+\left|0\right\rangle$$
$$=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\left\langle 0\right|B_l\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\hat{T}\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2\\ \alpha_3,i_3,j_3\\ \alpha_4,i_4,j_4}}\left\{M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}M_{\alpha_3}^{i_3j_3}M_{\alpha_4}^{i_4j_4}B_{i_1}^+B_{j_1}B_{i_2}^+B_{j_2}B_{i_3}^+B_{j_3}B_{i_4}^+B_{j_4}\right.$$
$$\times\left\langle e^{it_1(\varepsilon_{i_1}-\varepsilon_{j_1})}u_{\alpha_1}(t_1)e^{it_2(\varepsilon_{i_2}-\varepsilon_{j_2})}u_{\alpha_2}(t_2)e^{it_3(\varepsilon_{i_3}-\varepsilon_{j_3})}u_{\alpha_3}(t_3)e^{it_4(\varepsilon_{i_4}-\varepsilon_{j_4})}u_{\alpha_4}(t_4)\right\rangle_0 B_m^+\left|0\right\rangle$$
$$=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\hat{T}\sum_{\substack{\alpha_1,i_1,j_1\\ \alpha_2,i_2,j_2\\ \alpha_3,i_3,j_3\\ \alpha_4,i_4,j_4}}\left\{\delta_{li_1}\delta_{j_1i_2}\delta_{j_2i_3}\delta_{j_3i_4}\delta_{j_4m}M_{\alpha_1}^{i_1j_1}M_{\alpha_2}^{i_2j_2}M_{\alpha_3}^{i_3j_3}M_{\alpha_4}^{i_4j_4}\right.$$
$$\times\left\langle e^{it_1(\varepsilon_{i_1}-\varepsilon_{j_1})}u_{\alpha_1}(t_1)e^{it_2(\varepsilon_{i_2}-\varepsilon_{j_2})}u_{\alpha_2}(t_2)e^{it_3(\varepsilon_{i_3}-\varepsilon_{j_3})}u_{\alpha_3}(t_3)e^{it_4(\varepsilon_{i_4}-\varepsilon_{j_4})}u_{\alpha_4}(t_4)\right\rangle_0 B_m^+\left|0\right\rangle$$

$$=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\hat{T}\sum_{\substack{\alpha_1,\alpha_2,\alpha_3,\alpha_4\\ i_2,i_3,i_4}}\left\langle M_{\alpha_1}^{li_2}e^{it_1(\varepsilon_l-\varepsilon_{i_2})}u_{\alpha_1}(t_1)M_{\alpha_2}^{i_2i_3}e^{it_2(\varepsilon_{i_2}-\varepsilon_{i_3})}u_{\alpha_2}(t_2)\right.$$
$$\times\left.M_{\alpha_3}^{i_3i_4}e^{it_3(\varepsilon_{i_3}-\varepsilon_{i_4})}u_{\alpha_3}(t_3)M_{\alpha_4}^{i_4m}e^{it_4(\varepsilon_{i_4}-\varepsilon_m)}u_{\alpha_4}(t_4)\right\rangle_0$$
$$=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4\int_0^t dt_1\int_0^t dt_2\int_0^t dt_3\int_0^t dt_4\sum_{\substack{\alpha_1,\alpha_2,\alpha_3,\alpha_4\\ i_2,i_3,i_4}}\left\{M_{\alpha_1}^{li_2}M_{\alpha_2}^{i_2i_3}e^{it_1(\varepsilon_l-\varepsilon_{i_2})}e^{it_2(\varepsilon_{i_2}-\varepsilon_{i_3})}\hat{T}\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right.$$
$$\times M_{\alpha_3}^{i_3i_4}M_{\alpha_4}^{i_4m}e^{it_3(\varepsilon_{i_3}-\varepsilon_{i_4})}e^{it_4(\varepsilon_{i_4}-\varepsilon_m)}\hat{T}\left\langle u_{\alpha_3}(t_3)u_{\alpha_4}(t_4)\right\rangle_0$$
$$+M_{\alpha_1}^{li_2}M_{\alpha_3}^{i_3i_4}e^{it_1(\varepsilon_l-\varepsilon_{i_2})}e^{it_3(\varepsilon_{i_3}-\varepsilon_{i_4})}\hat{T}\left\langle u_{\alpha_1}(t_1)u_{\alpha_3}(t_3)\right\rangle_0$$
$$\times M_{\alpha_2}^{i_2i_3}M_{\alpha_4}^{i_4m}e^{it_2(\varepsilon_{i_2}-\varepsilon_{i_3})}e^{it_4(\varepsilon_{i_4}-\varepsilon_m)}\hat{T}\left\langle u_{\alpha_2}(t_2)u_{\alpha_4}(t_4)\right\rangle_0$$
$$+M_{\alpha_1}^{li_2}M_{\alpha_4}^{i_4m}e^{it_1(\varepsilon_l-\varepsilon_{i_2})}e^{it_4(\varepsilon_{i_4}-\varepsilon_m)}\hat{T}\left\langle u_{\alpha_1}(t_1)u_{\alpha_4}(t_4)\right\rangle_0$$
$$\times M_{\alpha_2}^{i_2i_3}M_{\alpha_3}^{i_3i_4}e^{it_2(\varepsilon_{i_2}-\varepsilon_{i_3})}e^{it_3(\varepsilon_{i_3}-\varepsilon_{i_4})}\hat{T}\left\langle u_{\alpha_2}(t_2)u_{\alpha_3}(t_3)\right\rangle_0$$

(

A20)

i) For unmixed excitonic states, we have

$$
\begin{aligned}
&\left\langle 0\left|B_l\left\langle U_2(t)\right\rangle_0 B_m^+\right|0\right\rangle \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\int_0^t dt_1\int_0^t dt_2\hat{T}\sum_{\alpha_1,i_1,\alpha_2,i_2}\left(M_{\alpha_1}^{i_1i_1}M_{\alpha_2}^{i_2i_2}\left\langle 0\left|B_l B_{i_1^+i_1}^{t_1}B_{i_2^+i_2}^{t_2}B_m^+\right|0\right\rangle\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\int_0^t dt_1\int_0^t dt_2\hat{T}\sum_{\alpha_1,i_1,\alpha_2,i_2}\left(M_{\alpha_1}^{i_1i_1}M_{\alpha_2}^{i_2i_2}\left\langle 0\left|B_l B_{i_1}^+B_{i_1}B_{i_2}^+B_{i_2}B_m^+\right|0\right\rangle\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\int_0^t dt_1\int_0^t dt_2\hat{T}\sum_{\alpha_1,i_1,\alpha_2,i_2}\left(M_{\alpha_1}^{i_1i_1}M_{\alpha_2}^{i_2i_2}\delta_{li_1}\delta_{i_1i_2}\delta_{i_2m}\left\langle u_{\alpha_1}(t_1)u_{\alpha_2}(t_2)\right\rangle_0\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\sum_{\alpha_1,\alpha_2}\left(M_{\alpha_1}^{ll}M_{\alpha_2}^{ll}\delta_{lm}\int_0^t dt_1\int_0^t dt_2\delta_{\alpha_1\alpha_2}D^0(\alpha_1,|t_1-t_2|)\right) \\
&=\frac{1}{2!}\left(\frac{-i}{\hbar}\right)^2\sum_{\alpha}\left[\left(M_{\alpha}^{ll}\right)^2\delta_{lm}\int_0^t dt_1\int_0^t dt_2 D^0(\alpha,|t_1-t_2|)\right]=\delta_{lm}\sum_{\alpha}\left(-\Phi_{\alpha}^l(t)\right)
\end{aligned}
$$

(A21)

Similarly, by using the Wick's theorem one finds

$$
\begin{aligned}
&\left\langle 0\left|B_l\left\langle U_4(t)\right\rangle_0 B_m^+\right|0\right\rangle \\
&=\frac{1}{4!}\left(\frac{-i}{\hbar}\right)^4 3\left\{\sum_{\alpha}\left[\left(M_{\alpha}^{ll}\right)^2\delta_{lm}\int_0^t dt_1\int_0^t dt_2 D^0(\alpha,|t_1-t_2|)\right]\right\}^2=\frac{1}{2!}\left[\sum_{\alpha}\left(-\Phi_{\alpha}^l(t)\right)\right]^2
\end{aligned}
$$

(A22)

and generally Eq. (9),

$$
\left\langle 0\left|B_l\left\langle U_{2n}(t)\right\rangle_0 B_m^+\right|0\right\rangle=\frac{1}{n!}\left[\sum_{\alpha}\left(-\Phi_{\alpha}^l(t)\right)\right]^n\delta_{lm}
$$

(A23)

j) For (Eq. 11)

At $T=0$, $\overline{N}_{\alpha}=0$, $\Phi_l(t)=g_l[1-\exp(-i\omega_0 t)]$ and the absorption coefficient reads

$$\alpha(\omega) = \frac{4\pi^2}{nc\hbar\omega V}\sum_{l\neq 0}\left\{\omega_l^2(\boldsymbol{ed_l})^2 e^{-g_l}\sum_{n=0}^{\infty}\left[\frac{g_l^n}{n!}\delta\left(\omega-\omega_l+\Delta_l-n\omega_0\right)\right]\right\}$$

(A24)

The intensities are proportional to the Poisson distribution.

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