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To cite this article: Luis A Martínez-Martínez and Joel Yuen-Zhou 2018 *New J. Phys.* **20** 018002

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Deutsche Physikalische Gesellschaft DPG
IOP Institute of Physics

Published in partnership with: Deutsche Physikalische Gesellschaft and the Institute of Physics

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OPEN ACCESS

RECEIVED
23 August 2017

ACCEPTED FOR PUBLICATION
12 January 2018

PUBLISHED
31 January 2018



COMMENT

# Comment on 'Quantum theory of collective strong coupling of molecular vibrations with a microcavity mode'

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## Abstract

We have found missing terms and incorrect signs in the secular master equations reported by Del Pino *et al* (2015 *New J. Phys.* **17** 053040) for vibrational polariton relaxation. Inclusion of these terms and signs are essential to yield correct (vanishing) pure dephasing rates between polariton states, as well as coherence transfer pathways between polaritons and dark states. We provide corrected expressions for the master equations as well as comparisons with the results reported by the authors. Even though the main conclusions of the article are not significantly altered, the corrections are important to provide a proper description of all possible polariton relaxation mechanisms within the invoked approximations, especially when applying the theory to model nonlinear spectroscopy of vibrational polaritons.

The model introduced in [1] by Del Pino *et al* (hereafter referred to as DP) consists of an ensemble of $N$ molecular harmonic vibrational modes with frequency $\omega_0$ strongly coupled to a single microcavity photonic mode at the same frequency (i.e. at resonance with the vibrational modes). The former are in turn coupled to a rovibrational environment whose spatial extent features two limiting situations: in one case, the $N$ modes are coupled to a common bath; in the other case, each vibrational mode interacts with its own independent but statistically identical bath. Owing to resonance between the vibrational modes and the photon, the resulting polariton states are $|\pm\rangle = \frac{1}{\sqrt{2}}(a^\dagger|0\rangle \pm |B\rangle)$, where $|B\rangle = \frac{1}{\sqrt{N}}\sum_{n=1}^{N} c_n^\dagger|0\rangle$ is the totally-symmetry bright state that couples to the microcavity photon. Assuming periodic boundary conditions, the dark states are orthogonal to $|B\rangle$, $|d\rangle = \frac{1}{\sqrt{N}}\sum_{n=1}^{N} e^{i\frac{2\pi}{N}dn} c_n^\dagger|0\rangle$ with nonzero quasimomentum $d = 1, \cdots, N-1$. Since only the matter part of the polaritons interacts with the rovibrational environment, and both polaritons feature the same matter wavefunction $|B\rangle$ (up to a phase), it follows that both $|+\rangle$ and $|-\rangle$ couple equally to the environment, giving rise to no pure-dephasing contributions for $\langle\pm|\rho|\mp\rangle$. However, an evaluation of master equations (12) and (13) in DP yield non-zero pure-dephasing rates for these coherences (see table 1 below). This observation led us to suspect that the aforementioned equations contain mistakes, which we aim to correct in this note.

To begin with, we point out that the bath correlation functions defined right before equation (6) in DP are missing some terms,

$$\phi_{ij}(t-t') = \sum_k \mathrm{Tr}_b\{\lambda_{ik}\lambda_{jk}(\tilde{b}_{ik}(t) + \tilde{b}_{ik}^\dagger(t))(\tilde{b}_{jk}(t') + \tilde{b}_{jk}^\dagger(t'))\}. \quad (1)$$

This corrected definition is important to have consistency with equation (6) in DP. Next, we rewrite the master equation in the interaction picture, equation (7) in DP, but given our boundary conditions, we take care of properly assuming that the eigenstate-site overlap coefficients $u_{ip} = \langle p|i\rangle$ (where $|i\rangle = c_i^\dagger|0\rangle$) can be complex-valued in general,

$$\partial_t\tilde{\rho}(t) = \sum_{ij}\sum_{pqrs}\int_0^\infty u_{ip}u_{qi}u_{jr}u_{sj}e^{i(\omega_{pq}-\omega_{sr})t+i\omega_{sr}\tau}[|r\rangle\langle s|\tilde{\rho}(t), |p\rangle\langle q|]\phi_{ij}(\tau)d\tau + \mathrm{h.c.} \quad (2)$$

Here, $i, j$ and $p, q, r, s$ are site and system-eigenstate indices, respectively.

In the secular approximation, only non-oscillatory terms ($\omega_{pq} - \omega_{sr} = 0$) give non-negligible contributions to the dynamics of $\tilde{\rho}$; this assumption decouples the time evolution for populations and coherences. The possible combinations $\{p, q, r, s\}$ that satisfy the secular condition are enumerated in table 1 in DP; however, they have ommited $\{p, q, r, s\}$ terms of the form $\{\pm, \pm, \mp, \mp\}$, $\{\pm, \pm, d, d'\}$, $\{d, d', \pm, \pm\}$. The latter are

**Table 1.** Comparison of equations of motion in the interaction picture for coherences ($\langle a|\tilde{\rho}|b\rangle = \tilde{\rho}_{ab}$, $a \neq b$) and populations ($\langle a|\tilde{\rho}|a\rangle = \tilde{\rho}_{aa}$) of the reduced vibrational-polariton density matrix calculated by DP and using the corrected equations (3) and (4). Additional expressions can be obtained by simultaneously making the changes $+ \leftrightarrow -$ and $\Gamma_a \leftrightarrow \Gamma_e$ throughout (for instance, we can obtain $\partial_t\tilde{\rho}_{-+}$ and $\partial_t\tilde{\rho}_{-d}$ from $\partial_t\tilde{\rho}_{-+}$ and $\partial_t\tilde{\rho}_{+d}$, respectively). Also, $\partial_t\tilde{\rho}_{\pm d} = \partial_t\tilde{\rho}^*_{d\pm}$ and $d$ labels dark states. Here, the $\delta_{d_1-d_2,d-d'}$ term indicates conservation of vibrational quasimomentum and must be interpreted in terms of mod $N$ arithmetic.

| $\partial_t\tilde{\rho}_{ab}$ | Master equations in [1] | Corrected master equations |
|---|---|---|
| | Delocalized bath case | |
| $\partial_t\tilde{\rho}_{+-}$ | $\left(-\frac{\gamma_\phi}{4} - \frac{\gamma_a}{8} - \frac{\gamma_e}{8}\right)\tilde{\rho}_{+-}$ | $\underbrace{\left(-\frac{\gamma_e}{8} - \frac{\gamma_a}{8}\right)\tilde{\rho}_{+-}}_{T_1 \text{ term}}$ |
| $\partial_t\tilde{\rho}_{+d}$ | $\left(-\frac{\gamma_e}{8} - \frac{5\gamma_\phi}{8}\right)\tilde{\rho}_{+d}$ | $\underbrace{-\frac{\gamma_e}{8}\tilde{\rho}_{+d}}_{T_1 \text{ term}}$ $\underbrace{-\frac{\gamma_\phi}{8}\tilde{\rho}_{+d}}_{T_2^* \text{ term}}$ |
| $\partial_t\tilde{\rho}_{d_1d_2}$ | 0 | 0 |
| $\partial_t\tilde{\rho}_{++}$ | $-\frac{\gamma_e}{4}\tilde{\rho}_{++} + \frac{\gamma_a}{4}\tilde{\rho}_{--}$ | $-\frac{\gamma_e}{4}\tilde{\rho}_{++} + \frac{\gamma_a}{4}\tilde{\rho}_{--}$ |
| $\partial_t\tilde{\rho}_{dd}$ | 0 | 0 |
| | Localized bath case | |
| $\partial_t\tilde{\rho}_{+-}$ | $-\left(\frac{\gamma_a}{8N} + \frac{\gamma_e}{8N} + \frac{\Gamma_a}{4N}(N-1) + \frac{\Gamma_e}{4N}(N-1) + \frac{\gamma_\phi}{8N}\right)\tilde{\rho}_{+-}$ | $\underbrace{-\left(\frac{\gamma_e}{8N} + \frac{\Gamma_e}{4N}(N-1) + \frac{\gamma_a}{8N} + \frac{\Gamma_a}{4N}(N-1)\right)\tilde{\rho}_{+-}}_{T_1 \text{ term}}$ |
| $\partial_t\tilde{\rho}_{+d}$ | $-\left(\frac{\gamma_e}{8N} + \frac{\Gamma_a}{4N} + \frac{\Gamma_e}{4N}(N-1)\right)\tilde{\rho}_{+d}$ $-\left(\frac{\Gamma_e}{4N} + \frac{\gamma_\phi}{8N} + \frac{\gamma_\phi}{2N}(N-1)\right)\tilde{\rho}_{+d}$ | $\underbrace{-\left(\frac{\gamma_e}{8N} + \frac{\Gamma_e}{4N}(N-1) + \frac{\Gamma_a}{4N} + \frac{\Gamma_e}{4N} + \frac{\gamma_\phi}{2N}(N-2)\right)\tilde{\rho}_{+d}}_{T_1 \text{ term}}$ $\underbrace{-\frac{\gamma_\phi}{8N}\tilde{\rho}_{+d}}_{T_2^* \text{ term}}$ $\underbrace{-\frac{\Gamma_a}{2N}\tilde{\rho}_{\bar{d}-}}_{\text{coherence transfer}}$ |
| $\partial_t\tilde{\rho}_{d_1d_2}$ | $-\left(\frac{\Gamma_a}{2N} + \frac{\Gamma_e}{2N} + \frac{\gamma_\phi}{N}(N-2)\right)\tilde{\rho}_{d_1d_2}$ $+\frac{\gamma_\phi}{N}\sum_{d,d'}(\delta_{d_1-d_2,d-d'} - \delta_{d,d_1}\delta_{d',d_2})\tilde{\rho}_{dd'}$ | $\underbrace{-\left(\frac{\Gamma_a}{2N} + \frac{\Gamma_e}{2N} + \frac{\gamma_\phi}{N}(N-2)\right)\tilde{\rho}_{d_1d_2}}_{T_1 \text{ term}}$ $\underbrace{+\frac{\gamma_\phi}{N}\sum_{d,d'}(\delta_{d_1-d_2,d-d'} - \delta_{d,d_1}\delta_{d',d_2})\tilde{\rho}_{dd'}}_{\text{coherence transfer}}$ |
| $\partial_t\tilde{\rho}_{++}$ | $-\frac{\gamma_e}{4N}\tilde{\rho}_{++} + \frac{\gamma_a}{4N}\tilde{\rho}_{--} - \frac{\Gamma_e}{2N}(N-1)\tilde{\rho}_{++}$ $+\sum_d \frac{\Gamma_a}{2N}\tilde{\rho}_{dd}$ | $-\frac{\gamma_e}{4N}\tilde{\rho}_{++} + \frac{\gamma_a}{4N}\tilde{\rho}_{--} - \frac{\Gamma_e}{2N}(N-1)\tilde{\rho}_{++}$ $+\sum_d \frac{\Gamma_a}{2N}\tilde{\rho}_{dd}$ |
| $\partial_t\tilde{\rho}_{dd}$ | $\frac{\Gamma_e}{2N}\tilde{\rho}_{++} - \frac{\Gamma_a}{2N}\tilde{\rho}_{dd} + \frac{\Gamma_a}{2N}\tilde{\rho}_{--}$ $-\frac{\Gamma_e}{2N}\tilde{\rho}_{dd} - \frac{\gamma_\phi}{N}(N-2)\tilde{\rho}_{dd} + \frac{\gamma_\phi}{N}\sum_{d'\neq d}\tilde{\rho}_{d'd'}$ | $\frac{\Gamma_e}{2N}\tilde{\rho}_{++} - \frac{\Gamma_a}{2N}\tilde{\rho}_{dd} + \frac{\Gamma_a}{2N}\tilde{\rho}_{--}$ $-\frac{\Gamma_e}{2N}\tilde{\rho}_{dd} - \frac{\gamma_\phi}{N}(N-2)\tilde{\rho}_{dd} + \frac{\gamma_\phi}{N}\sum_{d'\neq d}\tilde{\rho}_{d'd'}$ |

important to account for the proper evolution of coherences between polaritons and between polaritons and dark states, as will be shown next.

We find that the corrected master equation in the Schrödinger picture for a common bath ($\phi_{ij}(\tau) = \phi(\tau)$) is given by

$$\partial_t\rho = -\mathrm{i}[H_S, \rho] + \frac{\gamma_a}{4}\mathcal{L}_{\sigma_{+-}}[\rho] + \frac{\gamma_e}{4}\mathcal{L}_{\sigma_{-+}}[\rho] + \frac{\gamma_\phi}{4}\sum_{p=+,-}\mathcal{L}_{\sigma_{pp}}[\rho] + \gamma_\phi\mathcal{L}_{\mathcal{D}}[\rho] \tag{3a}$$

$$+\frac{\gamma_\phi}{4}(\sigma_{++}\rho\sigma_{--}^\dagger + \sigma_{--}\rho\sigma_{++}^\dagger) + \frac{\gamma_\phi}{2}\sum_d(\sigma_{++}\rho\sigma_{dd}^\dagger + \sigma_{dd}\rho\sigma_{++}^\dagger) \tag{3b}$$

$$+\frac{\gamma_\phi}{2}\sum_d(\sigma_{--}\rho\sigma_{dd}^\dagger + \sigma_{dd}\rho\sigma_{--}^\dagger), \tag{3c}$$

where $\sigma_{ab} = |a\rangle\langle b|$. Here, the terms in equation (3a) are identical to those in equation (12) in DP; however, the authors have missed the terms in equations (3b) and (3c), which arise from the omitted secular contributions outlined above.

On the other hand the master equation corresponding to localized baths ($\phi_{ij}(\tau) = \delta_{ij}\phi(\tau)$) reads

$$\partial_t\rho = -\mathrm{i}[H_s, \rho] + \frac{\gamma_a}{4N}\mathcal{L}_{\sigma_{+-}}[\rho] + \frac{\gamma_e}{4N}\mathcal{L}_{\sigma_{-+}}[\rho] \tag{4a}$$

$$+\sum_d\frac{\Gamma_a}{2N}(\mathcal{L}_{\sigma_{d-}}[\rho] + \mathcal{L}_{\sigma_{+d}}[\rho]) + \sum_d\frac{\Gamma_e}{2N}(\mathcal{L}_{\sigma_{d+}}[\rho] + \mathcal{L}_{\sigma_{-d}}[\rho]) \tag{4b}$$

$$+\frac{\Gamma_a}{4N}\sum_d(-[\sigma_{\bar{d}-}\rho,\ \sigma_{d+}]-[\sigma_{+\bar{d}}\rho,\ \sigma_{-d}]+\text{h.c.}) \tag{4c}$$

$$+\frac{\Gamma_e}{4N}\sum_d(-[\sigma_{\bar{d}+}\rho,\ \sigma_{d-}]-[\sigma_{-\bar{d}}\rho,\ \sigma_{+d}]+\text{h.c.}) \tag{4d}$$

$$+\frac{\gamma_\phi}{4N}\sum_{p=+,-}\mathcal{L}_{\sigma_{pp}}[\rho]+\gamma_\phi\sum_i\mathcal{L}_{\mathcal{D}c_i^\dagger c_i\mathcal{D}}[\rho] \tag{4e}$$

$$+\frac{\gamma_\phi}{4N}(\sigma_{++}\rho\sigma_{--}^\dagger+\sigma_{--}\rho\sigma_{++}^\dagger)+\frac{\gamma_\phi}{2N}\sum_d(\sigma_{++}\rho\sigma_{dd}^\dagger+\sigma_{dd}\rho\sigma_{++}^\dagger) \tag{4f}$$

$$+\frac{\gamma_\phi}{2N}\sum_d(\sigma_{--}\rho\sigma_{dd}^\dagger+\sigma_{dd}\rho\sigma_{--}^\dagger). \tag{4g}$$

Here, to avoid confusion, we use the notation $|\bar{d}\rangle\equiv|N-d\rangle$ (the dark state with opposite quasimomentum to that of $|d\rangle$). The terms in equations (4*a*), (4*b*), and (4*e*) coincide with those in equations (13*a*), (13*b*), and (13*e*) in DP, respectively. Equations (4*c*) and (4*d*) differ from those in equations (13*c*) and (13*d*) in DP by some signs. Finally, equations (4*f*) and (4*g*) are missing in DP and arise from the omitted secular terms.

For a better appreciation of the missing information in DP, we compare in table 1 the equations of motion for populations and coherences that follow from equations (3) and (4) with those that follow from DP. By reporting the results in the interaction picture, we simply neglect the trivial coherent dynamics generated by $H_s$.

We now briefly elaborate on the physical processes that are incompletely captured by DP. First, in secular Redfield theory, the decay rate $T_2^{-1}$ of a coherence $\rho_{ab}$ has several contributions: one is an average of the population decay rates of the system states $|a\rangle$ and $|b\rangle$, often labeled $T_1^{-1}$; the other one is the pure dephasing rate $T_2^{*-1}$ associated with fluctuations of the system-environment coupling between $|a\rangle$ and $|b\rangle$; finally, there can also be coherence transfers to $\rho_{cd}$ as long as $\omega_{cd}=\omega_{ab}$. These physical processes have been highlighted in table 1, and can be readily calculated using textbook formalism such as that found in [2].

We note that some of the omitted terms by DP are proportional to $\gamma_\phi$, indicating pure-dephasing contributions, in agreement with our original observation that the pure-dephasing rate between $|\pm\rangle$ and $|\mp\rangle$ is zero; see entries corresponding to $\partial\tilde{\rho}_{+-}$, which do not feature $T_2^*$ terms. We also notice that equations (4*c*) and (4*d*) yield non-zero contributions for the decay rate of coherences between polariton and dark states in the independent baths case. This is in contrast with equations (13*c*) and (13*d*) in DP, which vanish identically due to sign errors. For instance, from table 1, the corrected coherence evolution $\partial_t\rho_{+d}$ contains an additional decay due to coherence transfer $-\frac{\Gamma_a}{2N}\langle\bar{d}|\rho|-\rangle$. This term contributes to an additional decay channel of $\rho_{+d}$ and arises due to the fact that both $|+\rangle$ and $|-\rangle$ feature the same molecular state $|B\rangle$, but with opposite signs.

Table 1 shows that our corrections do not change the main conclusions established by DP, since their discussion was mainly focused on population transfer dynamics, while the omissions affect coherences. These omissions, however, will be essential to understand nonlinear spectroscopic signals of polaritons.

## Acknowledgments

LAMM is grateful for the support of the UC-Mexus CONACyT scholarship for doctoral studies. J. Y.-Z. acknowledges funding from the NSF CAREER Award CHE-1654732. Both authors acknowledge support from generous UCSD startup funds.

## Appendix

### A.1. Missing terms for pure dephasing

We begin by invoking the secular approximation in equation (2), setting $\omega_{pq}-\omega_{sr}=0$. We only consider the missing terms in DP, namely, the cases where $\{p,\ q,\ r,\ s\}$ are $\{\pm,\pm,\mp,\mp\}$, $\{\pm,\pm,d,\ d'\}$, and $\{d,\ d',\pm,\pm\}$. The system-eigenstate-site overlaps $|u_{\pm i}|=\frac{1}{\sqrt{2N}}$ and $|u_{di}|=\frac{1}{\sqrt{N}}$ become handy.

To proceed, let us analyze the two bath cases:

1. For the common bath, $\phi_{ij}(\tau)=\phi(\tau)$, so $\sum_{ij}u_{ip}u_{qi}u_{jr}u_{sj}\phi(\tau)=\langle p|\mathbf{P}_{\text{vib}}|q\rangle\langle r|\mathbf{P}_{\text{vib}}|s\rangle\phi(\tau)$, where $\mathbf{P}_{\text{vib}}=\sum_i|i\rangle\langle i|$ is the projector on the vibrational subspace. For the cases of interest above, we have two possibilities:
   (a) $\{p,\ q,\ r,\ s\}=\{\pm,\pm,\mp,\mp\}$, in which case $\langle p|\mathbf{P}_{\text{vib}}|q\rangle\langle r|\mathbf{P}_{\text{vib}}|s\rangle=\frac{1}{4}$.
   (b) $\{p,\ q,\ r,\ s\}=\{\pm,\pm,d,\ d'\}$, $\{d,\ d',\pm,\pm\}$, in which case $\langle p|\mathbf{P}_{\text{vib}}|q\rangle\langle r|\mathbf{P}_{\text{vib}}|s\rangle=\frac{\delta_{dd'}}{2}$.

2. For independent baths, $\phi_{ij}(\tau) = \delta_{ij}\phi(\tau)$, so $\sum_{ij} u_{ip}u_{qi}u_{jr}u_{sj}\delta_{ij}\phi(\tau) = \sum_i \langle p|i\rangle\langle i|q\rangle\langle r|i\rangle\langle i|s\rangle\phi(\tau)$. We analyze the two possibilities again:
   (a) $\{p, q, r, s\} = \{\pm, \pm, \mp, \mp\}$, in which case $\sum_i \langle p|i\rangle\langle i|q\rangle\langle r|i\rangle\langle i|s\rangle = N\frac{1}{4N^2} = \frac{1}{4N}$.
   (b) $\{p, q, r, s\} = \{\pm, \pm, d, d'\}, \{d, d', \pm, \pm\}$, in which case $\sum_i \langle p|i\rangle\langle i|q\rangle\langle r|i\rangle\langle i|s\rangle = N\frac{1}{2N^2}\delta_{dd'} = \frac{\delta_{dd'}}{2N}$.

This exercise allows us to discard the $d \neq d'$ cases. Hence, we only need to develop the $\{p, q, r, s\} = \{p, p, r, r\}$ term (where $p \neq r$) in the right-hand-side of equation (2),

$$\sum_{ij}\int_0^\infty u_{ip}u_{pi}u_{jr}u_{rj}[|r\rangle\langle r|\tilde{\rho}(t), |p\rangle\langle p|]\phi_{ij}(\tau)\mathrm{d}\tau + \text{h.c.} = \gamma_{r,p}^{deph}\sigma_{rr}\rho\sigma_{pp}^\dagger + \text{h.c.}, \tag{5}$$

where

$$\gamma_{r,p}^{\text{deph}} = \gamma_{p,r}^{\text{deph}} = \sum_{ij} u_{ip}u_{pi}u_{jr}u_{rj}\int_{-\infty}^{\infty}\phi_{ij}(\tau)\mathrm{d}\tau. \tag{6}$$

Inclusion of terms of the form of equation of (5) for $\{p, r\} = \{\pm, \mp\}, \{\pm, d\}, \{d, \mp\}$ into the master equation gives rise to the correction terms in equations (3*b*), (3*c*), (4*f*), and (4*g*), where $\gamma_{\mp,\pm}^{\text{deph}} = \frac{\gamma_\phi}{4}$, $\gamma_{d,\pm}^{\text{deph}} = \gamma_{\pm,d}^{\text{deph}} = \frac{\gamma_\phi}{2}$ for the common bath and $\gamma_{\mp,\pm}^{\text{deph}} = \frac{\gamma_\phi}{4N}$, $\gamma_{d,\pm}^{\text{deph}} = \gamma_{\pm,d}^{\text{deph}} = \frac{\gamma_\phi}{2N}$ for independent baths, where $\gamma_\phi = 2S(0)$.

### A.2. Missing coherence transfer pathways

DP miss coherence transfer pathways that arise from combinations $\{p, q, r, s\} = \{+d_1, -d_2\}$ in equation (2),

$$\begin{aligned}
&\sum_{ij}\int_0^\infty u_{i+}u_{d_1 i}u_{j-}u_{d_2 j}\mathrm{e}^{\mathrm{i}\omega_{d-}\tau}\phi_{ij}(\tau)\mathrm{d}\tau[|-\rangle\langle d_2|\tilde{\rho}, |+\rangle\langle d_1|] + \text{h.c.}\\
&+\sum_{ij}\int_0^\infty u_{id_1}u_{-i}u_{jd_2}u_{+j}\mathrm{e}^{\mathrm{i}\omega_{+d}\tau}\phi_{ij}(\tau)\mathrm{d}\tau[|d_2\rangle\langle +|\tilde{\rho}, |d_1\rangle\langle -|] + \text{h.c.}\\
&+\sum_{ij}\int_0^\infty u_{id_1}u_{+i}u_{jd_2}u_{-j}\mathrm{e}^{\mathrm{i}\omega_{-d}\tau}\phi_{ij}(\tau)\mathrm{d}\tau[|d_2\rangle\langle -|\tilde{\rho}, |d_1\rangle\langle +|] + \text{h.c.}\\
&+\sum_{ij}\int_0^\infty u_{i-}u_{d_1 i}u_{j+}u_{d_2 j}\mathrm{e}^{\mathrm{i}\omega_{d+}\tau}\phi_{ij}(\tau)\mathrm{d}\tau[|+\rangle\langle d_2|\tilde{\rho}, |-\rangle\langle d_1|] + \text{h.c.}
\end{aligned} \tag{7}$$

where $\omega_d = \omega_{d_1} = \omega_{d_2}$. Notice that due to orthogonality $\langle \pm|d_{1(2)}\rangle = 0$, each of these four terms is zero for the case of the common bath. For the case of the independent baths, we use the fact that

$$\sum_i u_{i\pm}u_{d_1 i}u_{i\mp}u_{d_2 i} = -\frac{1}{2N}\sum_i u_{d_1 i}u_{d_2 i} = -\frac{1}{2N}\delta_{d_2, N-d_1}, \tag{8}$$

where conservation of quasimomentum in the finite $N$ molecule chain leads to $d_2 + d_1 = 0 (\text{mod } N)$, or $d_2 = N - d_1 \equiv \bar{d}_1$. The expression in equation (7) then becomes,

$$\begin{aligned}
-\Bigg(&\frac{\Gamma_a}{4N}[|\bar{d}\rangle\langle -|\tilde{\rho}, |d\rangle\langle +|] + \frac{\Gamma_a}{4N}[|+\rangle\langle \bar{d}|\tilde{\rho}, |-\rangle\langle d|]\\
&+\frac{\Gamma_e}{4N}[|-\rangle\langle \bar{d}|\tilde{\rho}, |+\rangle\langle d|] + \frac{\Gamma_e}{4N}[|\bar{d}\rangle\langle +|\tilde{\rho}, |d\rangle\langle -|]\Bigg) + \text{h.c.}
\end{aligned} \tag{9}$$

where, as usual, we have taken only the real part of the resulting half-sided Fourier transforms (assuming that the Lamb shift corresponding to the imaginary part can be absorbed into the coherent dynamics), $\Re\int_0^\infty \mathrm{e}^{\mathrm{i}\omega_{qr}\tau}\phi(\tau)\mathrm{d}\tau = S(\omega_{qr})$ and $\Gamma_a = 2S(\omega_{d+}) = 2S(\omega_{-d})$ and $\Gamma_e = 2S(\omega_{+d}) = 2S(\omega_{d-})$. Equation (9) is equal to equations (4*c*) and (4*d*), which were featured with incorrect signs as equations (13*c*) and (13*d*) in DP.

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