Erratum: "Ultrafast exciton-exciton coherence transfer in molecular aggregates and its application to light harvesting systems" [J. Chem. Phys. 127, 075101 (2007)]

Kim Hyeon-Deuk,[1,a] Yoshitaka Tanimura,[1] and Minhaeng Cho[2,3]
[1]*Department of Chemistry, Kyoto University, Kyoto 606-8502, Japan*
[2]*Department of Chemistry and Center for Multidimensional Spectroscopy, Korea University, Seoul 136-701, Republic of Korea*
[3]*Multidimensional Spectroscopy Laboratory, Korea Basic Science Institute, Seoul 136-701, Republic of Korea*

(Received 24 January 2008; accepted 30 January 2008; published online 28 March 2008)

[DOI: 10.1063/1.2888969]

A few equations in our recent paper,[1] which will be referred to as Paper I, were found to be inaccurate. First of all, $q^{(c)}_{\mu\mu}$ in Eq. (6) should be replaced by $q^{(c)}_{\mu\nu}$. Equations (14) and (15) should also be corrected as

$$\mathsf{P}_\mu \rho \equiv \bar{\rho}_{\mu\mu}(\{q_j\})\mathrm{Tr}_{\{q_j\}}[\rho_{\mu\mu}] = \hat{\rho}_{\mu\mu}, \tag{14}$$

where $\rho=\rho(\{q_j\})$ is the density matrix, and the equilibrium density matrix in the μth exciton is

$$\bar{\rho}_{\mu\mu}(\{q_j\}) = \frac{\exp[-\beta H_\mu(\{q_j\})]}{Z}. \tag{15}$$

Similarly, Eq. (16) should be corrected. The definition of the Green function for the exciton-exciton coherence transfer (EECT) in Eq. (32) of Paper I should be corrected as

$$\tilde{G}_{\lambda\lambda'}(t_2) \equiv \mathrm{Tr}[\lambda^\dagger \hat{G}_{\lambda\lambda'}(t_2)\hat{\rho}_{\lambda'}]. \tag{32}$$

Accordingly, $\hat{\rho}_{\lambda'}$ in Eq. (31) should be removed. We took the trace of Eq. (39) over bath modes, and the system and bath are assumed to be initially uncorrelated, i.e., $\hat{\rho}_{\lambda'}=\mathbf{Q}_{\lambda'}\rho(0)=\bar{\rho}_{\lambda'}(\{q_j\})\mathrm{Tr}_{\{q_j\}}[\rho_{\lambda'}(0)]$. Note that $\hat{\rho}_{\lambda'}$ should be added to the ends of both sides of Eq. (40). Thus, Eq. (41) is independent of any bath modes and only describes the time-evolution of system's coherence. Appendix B of Paper I should be similarly corrected. We emphasize, however, that the time-evolution equation of the Green function for EECT, Eq. (43) of Paper I, is correct, although there is one typographical error: γ in the integrand should be changed to λ.

Second, $\hat{G}_{\mathsf{PQ}}$ and $\hat{G}_{\mathsf{QP}}$ in Appendix A should be corrected as

$$\hat{\mathrm{G}}_{\mathsf{PQ}}(t_2) = -i\int_0^{t_2} dt \mathsf{P} e^{-i\hat{L}_0(t_2-t)}(\mathsf{P}+\mathsf{Q})\hat{L}_1(\mathsf{P}+\mathsf{Q})e^{-i\hat{L}t}\mathsf{Q} \tag{A2}$$

$$= -i\int_0^{t_2} dt \mathsf{P} e^{-i\hat{L}_0(t_2-t)}\mathsf{P}\hat{L}_1\mathsf{Q}e^{-i\hat{L}t}\mathsf{Q}$$

$$\sim \int_0^{t_2} dt O(\hat{L}_1\hat{\mathrm{G}}_{\mathsf{QQ}}(t)) \tag{A3}$$

and

$$\hat{\mathrm{G}}_{\mathsf{QP}}(t_2) = -i\int_0^{t_2} dt \mathsf{Q} e^{-i\hat{L}_0(t_2-t)}(\mathsf{P}+\mathsf{Q})\hat{L}_1(\mathsf{P}+\mathsf{Q})e^{-i\hat{L}t}\mathsf{P} \tag{A4}$$

$$= -i\int_0^{t_2} dt \mathsf{Q} e^{-i\hat{L}_0(t_2-t)}\mathsf{Q}\hat{L}_1\hat{\mathrm{G}}_{\mathsf{QP}}(t)$$

$$-i\int_0^{t_2} dt \mathsf{Q} e^{-i\hat{L}_0(t_2-t)}\mathsf{Q}\hat{L}_1\mathsf{P}e^{-i\hat{L}t}\mathsf{P}$$

$$\sim \int_0^{t_2} dt O(\hat{L}_1\hat{\mathrm{G}}_{\mathsf{PP}}(t)). \tag{A5}$$

Therefore, we do not have to consider the above higher-order contributions in our calculation. We also note that Eqs. (A6) and (A7) can be removed, since we took $\hat{\mathrm{G}}_{\mathsf{PP}}(t_2)$ into consideration as the exciton population transfer contribution.

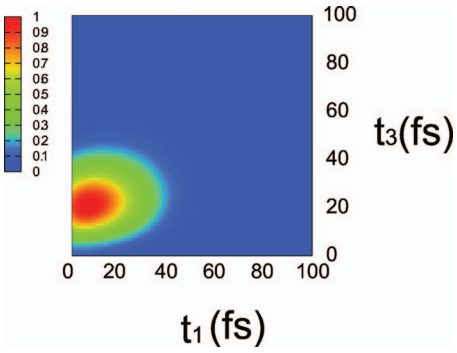

FIG. 3. (Color) (a) The time-resolved echo signals $|R^{(3)}(t_3,t_2,t_1)|^2$ vs t_1 and t_3 with $t_2=100$ fs. The main peak is smaller than that in Fig. 3(a) of Paper I.

[a]Electronic mail: kim@kuchem.kyoto-u.ac.jp.

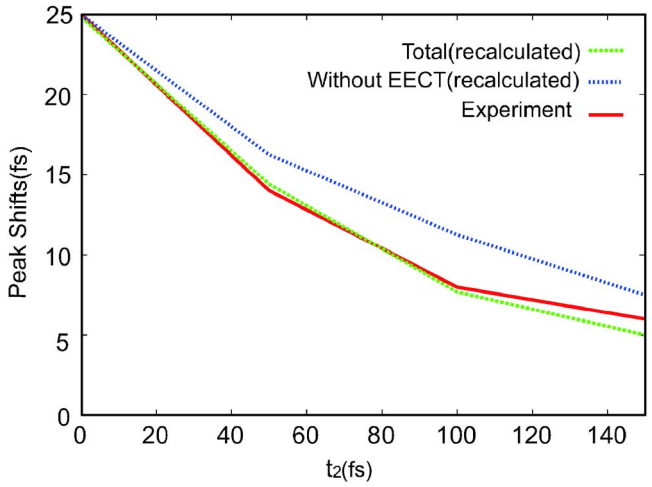

FIG. 4. (Color online) Photon echo peak shifts (PEPS) with respect to t_2. The experimental data (solid line) are measured in Ref. 2, which is the same as the dotted line in Fig. 4 of Paper I. The direct comparison shows that the recalculated PEPS (dashed line) agrees well with the experimental data in the ultrafast time region. The small deviation in the longer time region might be attributed to the instability of coherence in the density matrix.

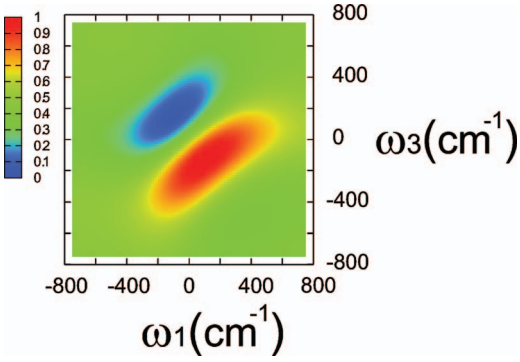

FIG. 5. (Color) (a) Real part of the 2D photon echo spectrum $\int_0^\infty\int_0^\infty \exp(i\Omega_1 t_1)\exp(-i\Omega_3 t_3)R^{(3)}(t_3,t_2,t_1)dt_1dt_3$ at $t_2=100$ fs. The horizontal and vertical axes are $\Omega_1-11\,790$ cm^{-1} and $\Omega_3-11\,790$ cm^{-1}, respectively. The width along the anti-diagonal axis is broader than that in Fig. 5(a) of Paper I, which shows the faster memory loss.

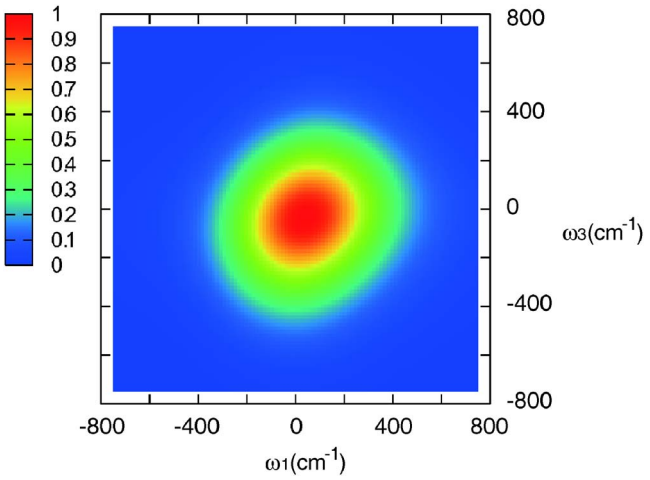

FIG. 6. (Color online) (a) The same as Fig. 5(a), but the absolute magnitude. The recalculated spectrum is rounder than that in Fig. 6(a) of Paper I, which demonstrates the faster decoherence.

Lastly, we recalculated the spectroscopic results in Sec. IX of Paper I with the new parameters; the eigen-energy of B850 $\tilde{\Omega}_m=11\,280$ cm^{-1}, the full width at half maximum $\sigma=2\sqrt{\log 2}\bar{\Omega}_m=225$ cm^{-1}, and the homogeneous parameter $\kappa=1200$ cm^{-1}. In the numerical calculations of the nonlinear optical signals and spectra shown in Paper I, we incorrectly excluded the term $-R_{\mathrm{EECT}}(t_3,0,t_1)$. We found that the recalculated absorption spectrum of B850 with the above parameters is in agreement with the experimental result in Fig. 8 of Ref. 2. Figures 3(a), 4, 5(a), and 6(a) are the recalculated 2D time-resolved photon echo signal, photon echo peak shift, and 2D photon echo, respectively. All the results have been averaged over 1000 realizations of the static disorder.

[1] K. Hyeon-Deuk, Y. Tanimura, and M. Cho, J. Chem. Phys. **127**, 075101 (2007).
[2] R. Jimenez, F. van Mourik, J. Y. Yu, and G. R. Fleming, J. Phys. Chem. B **101**, 7350 (1997).