

Coherence, Decoherence and Incoherence in Natural Light Harvesting Processes

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The Interest: Natural Light Processes for Either Biological or Technological Processes

The issues:

1. Highly unexpected timescales were observed for the coherent flow of electronic energy in some light harvesting molecules. How to explain/understand/relate to system properties?
2. Such observations have prompted many enthusiastic arguments saying that quantum coherence effects are, therefore, active and prominent in biology.
But...nature of the light!

Sample (over) enthusiasm:

“It turns out that bacteria have been up to quantum Computation for hundreds of millions of years”

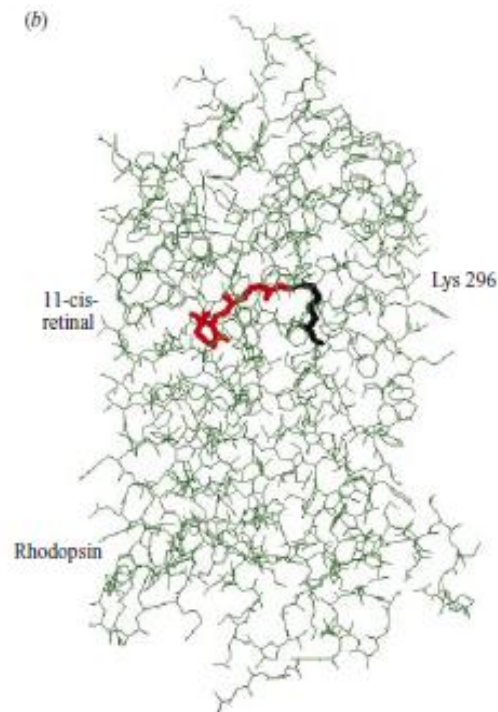
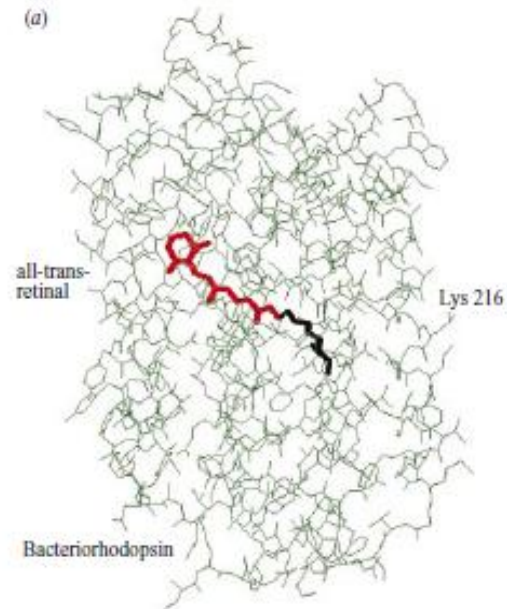
“Now that non-trivial quantum effects have been unambiguously shown in biology---let us make a sensor!”

“We are witnessing the dawn of quantum biology”

Related enthusiasm for dynamics of retinal in vision. One sees quantum coherent dynamics within an apparently very hostile (decohering) environment.

Why hostile? ---

i.e. what do they look like?



Relevant for vertebrate visual transduction: light induced cis \rightarrow trans isomerization

Fig. 14. Crystal structures of bR (a) and vertebrate rhodopsin (b). The retinyl groups are shown in red, and the lysine residues to which they are attached in black. The rhodopsin coordinates are from Palczewski *et al.* (2000); the bR coordinates, from Luecke *et al.* (1999b). The proteins are shown in roughly similar orientations, with the 7-transmembrane α -helices of each protein running vertically, the N-terminus at the top, and the first transmembrane α -helix from this terminus on the right.

Thus for both the quantum information focus and the chemistry focus --- we have

Highly unexpected timescales observed for the coherent dynamics of energy flow in some light induced dynamics.

Unexpected because system is nanoscale system with great potential for decoherence. Yet if the electronic coherence survives → route to sustaining coherence in such systems.

Focus on the photosynthesis case/ comment on retinal

Clarify “Coherence”:

Time dependence through wavefunction superposition of energy eigenstates.

E.g. isolated two level system

$$\Psi(0) = c_1 \phi_1 + c_2 \phi_2$$

$$\Psi(t) = c_1 \phi_1 \exp(-i E_1 t / \hbar) + c_2 \phi_2 \exp(-i E_2 t / \hbar)$$

(Swedish political party)

Open systems:

time dependent off- diagonal matrix elements of the system density matrix in the system energy eigenbasis

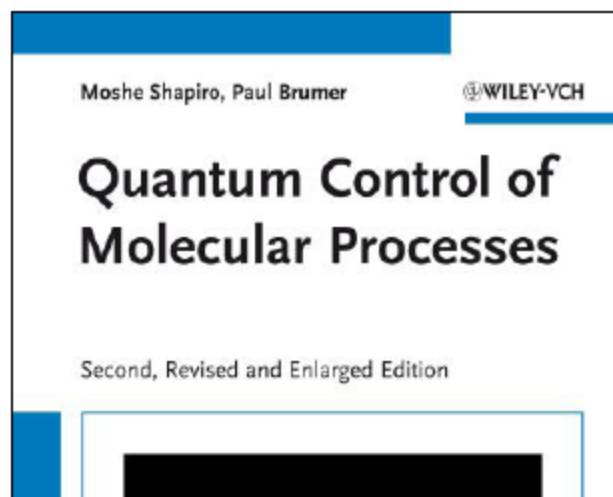
ATOMIC AND MOLECULAR PHYSICS

COMMERCIAL BREAK:
A TERRIFIC INVESTMENT
OPPORTUNITY!!

Moshe Shapiro
(*University of British Columbia, Canada*)
Paul Brumer
(*University of Toronto, Canada*)

Quantum Control of Molecular Processes

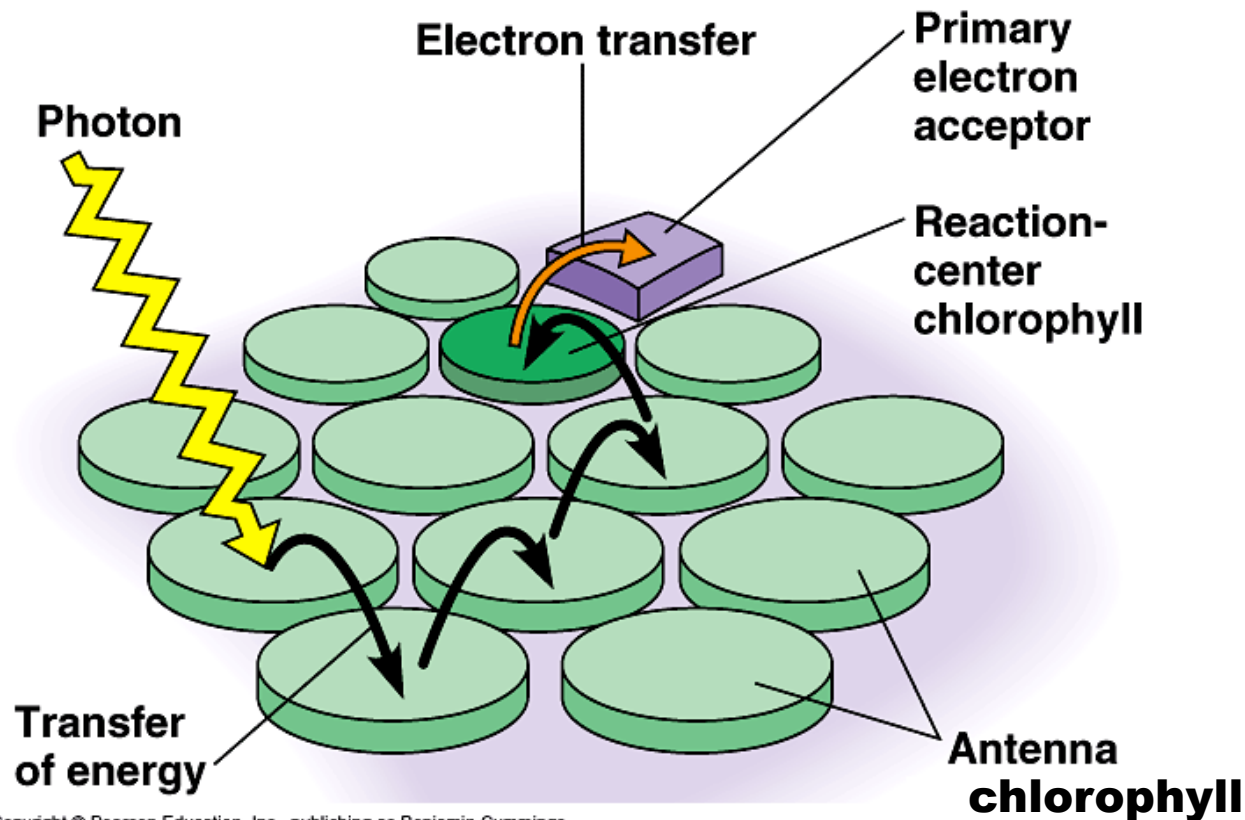
Second, Revised and Enlarged Edition



Written by two of the world's leading researchers in the field, this is a systematic introduction to the fundamental principles of coherent control, and to the underlying physics and chemistry.

This fully updated second edition is enhanced by 80% and covers the latest techniques and applications, including

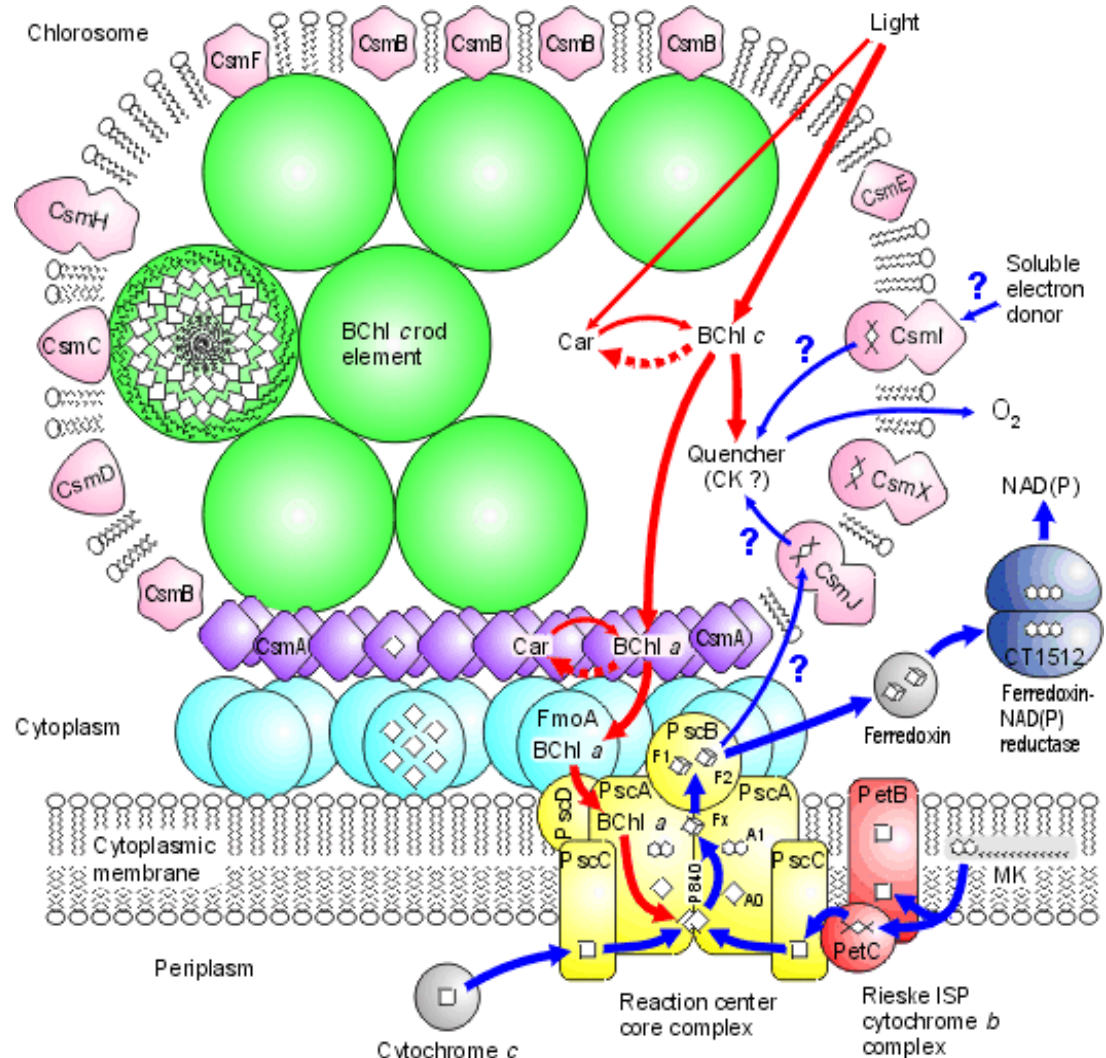
Focus on Energy Transfer in Photosynthesis since Highly Efficient Process



Diffusive Incoherent Hopping??

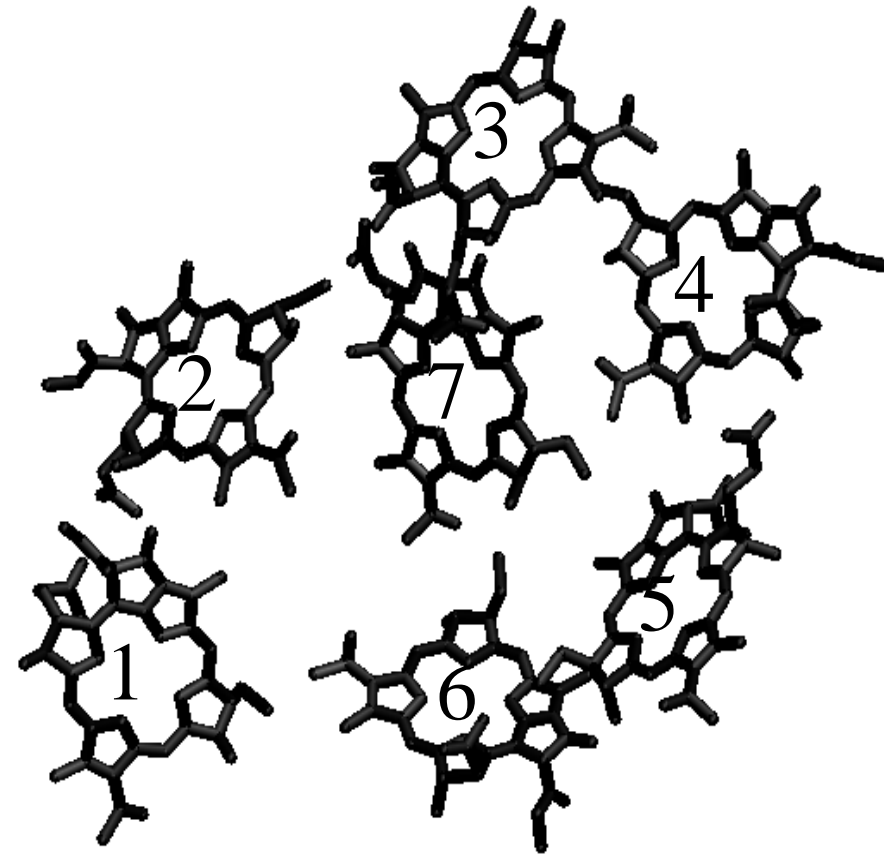
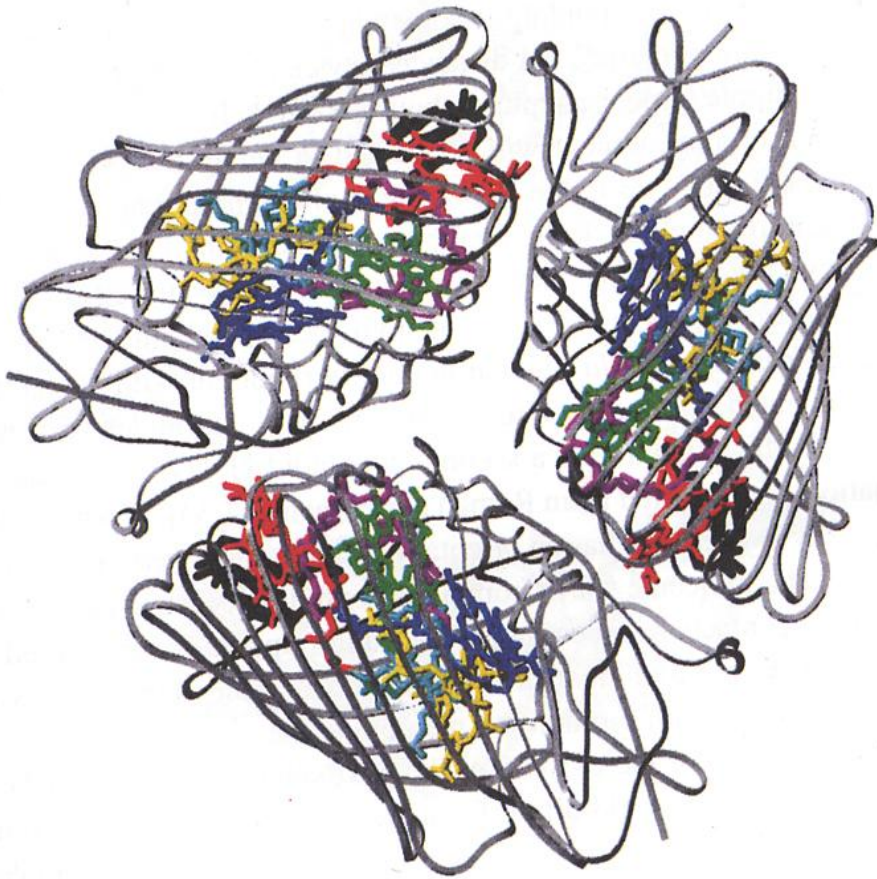
Quantum Coherent Wave-like ?

- FMO's role in light harvesting
 - Trimer structure
 - Monomer contains seven chlorophylls
 - Energetic wire connecting chlorosome to Reaction center



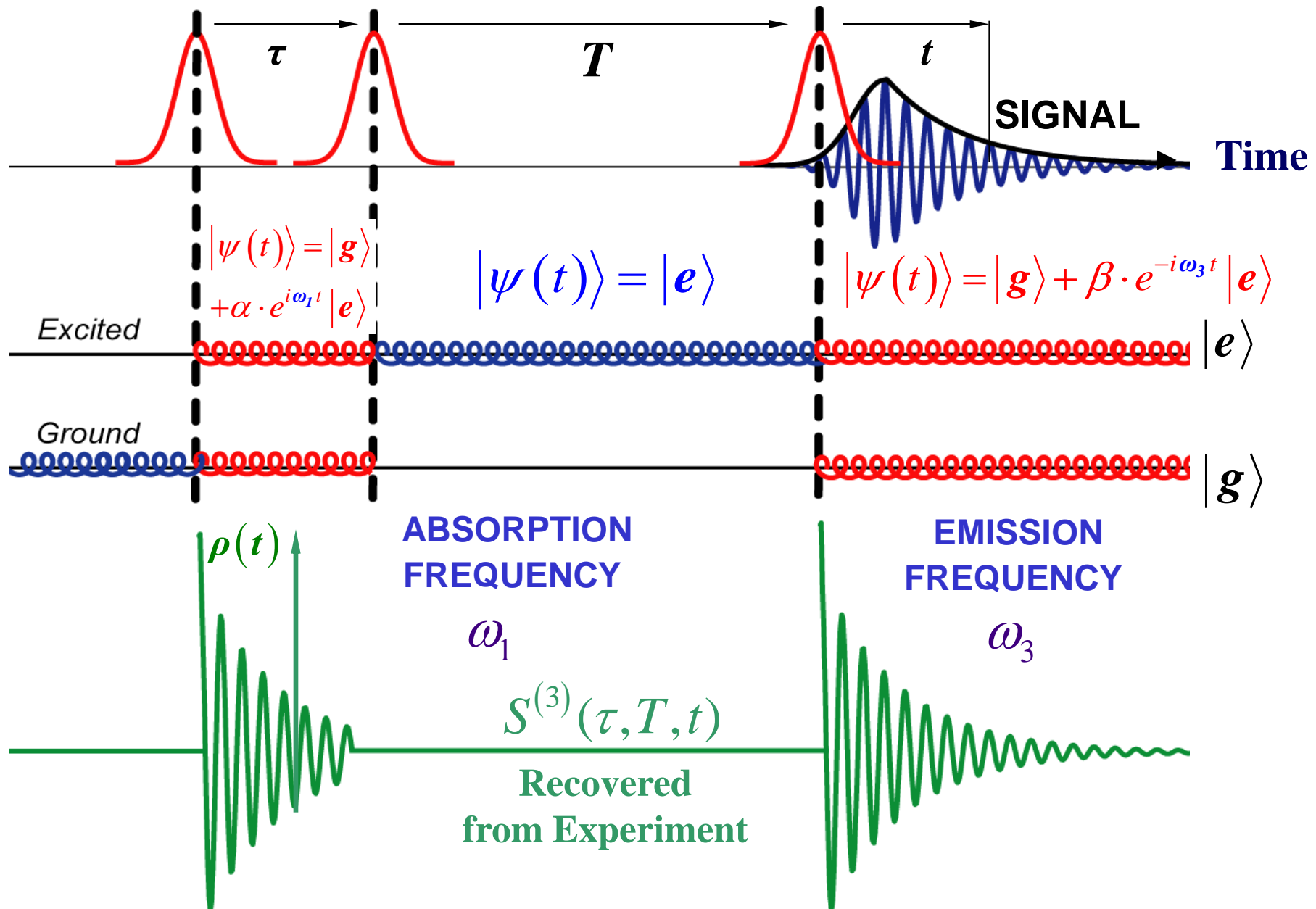
Trimeric FMO complex

- Closely packed BChls in hydrophobic protein environment
- BChls in different subunits are very far apart – only need to consider the 7 BChls in a monomer of FMO



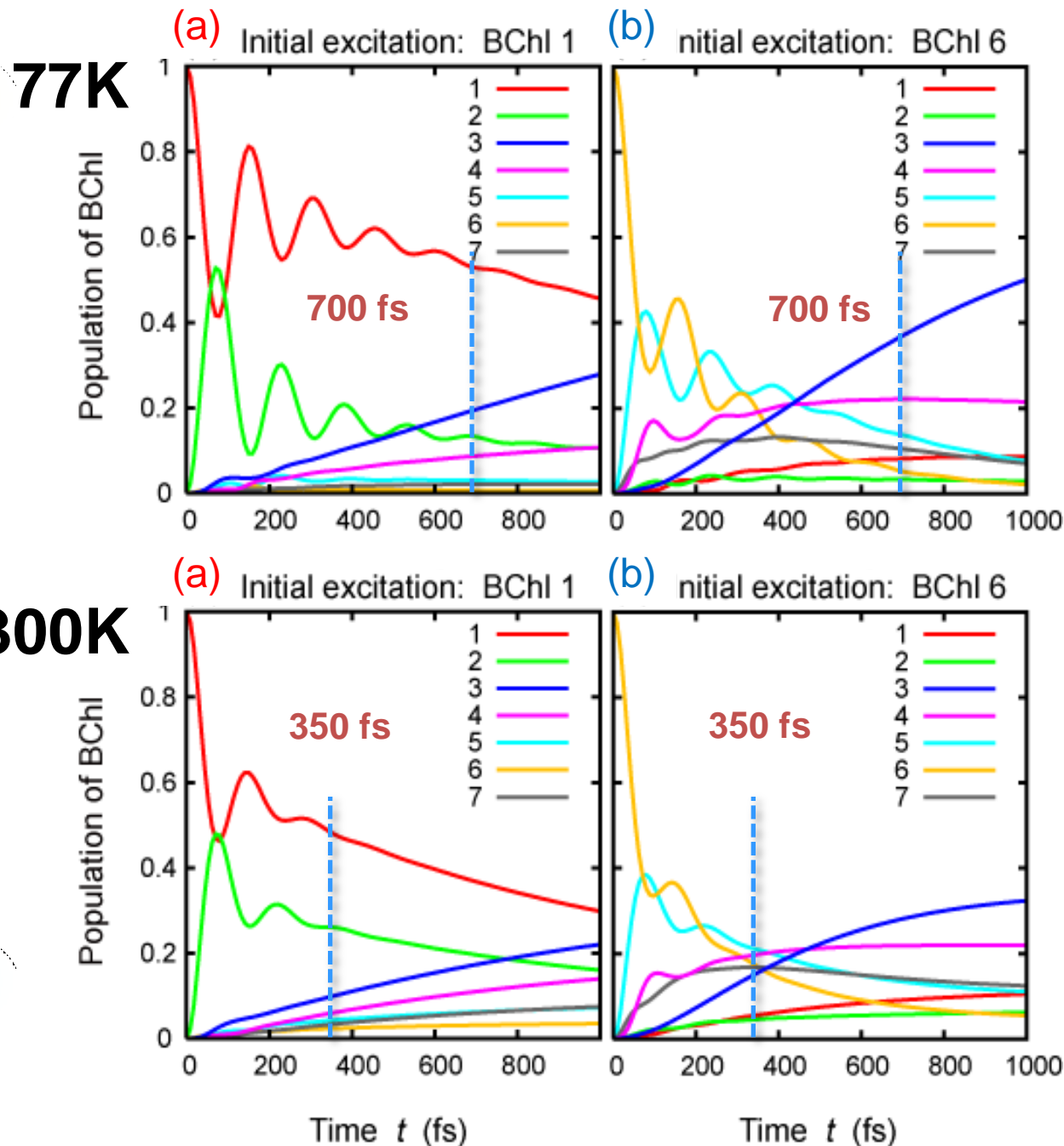
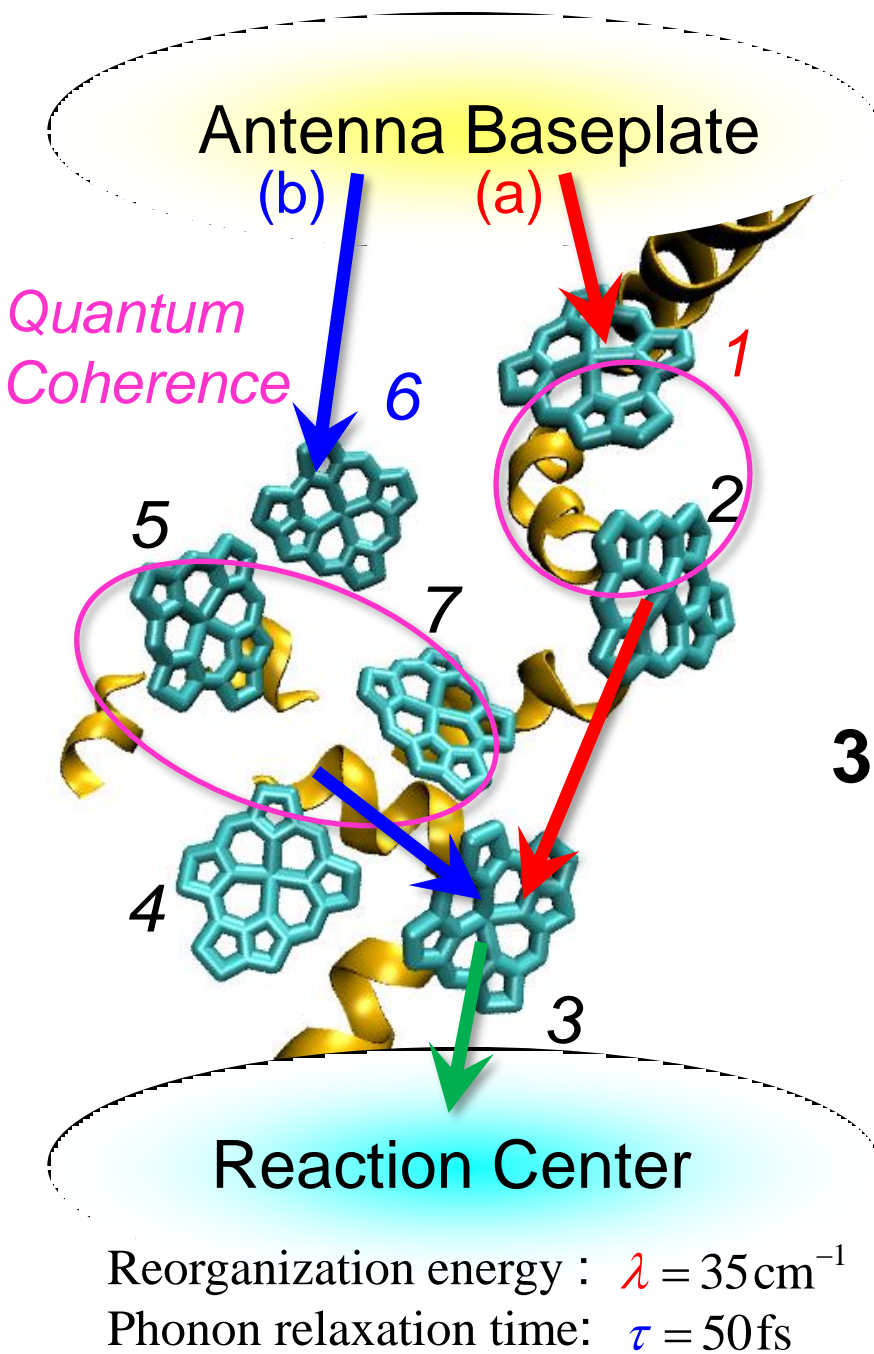
Spatial arrangement of BChls

Along comes 2D Electronic Spectroscopy (Fleming)



- Obtain $S^{(3)}(\omega_1, T, \omega_3)$ by double Fourier Transformations in τ and t
- Retrieves Correlation between Absorption and Emission Frequencies

Coherent Energy Transfer through the FMO Complex (here calc)



Coherently wired light-harvesting in photosynthetic marine algae at ambient temperature

Elisabetta Collini^{1*}†, Cathy Y. Wong^{1*}, Krystyna E. Wilk², Paul M. G. Curmi², Paul Brumer¹ & Gregory D. Scholes¹

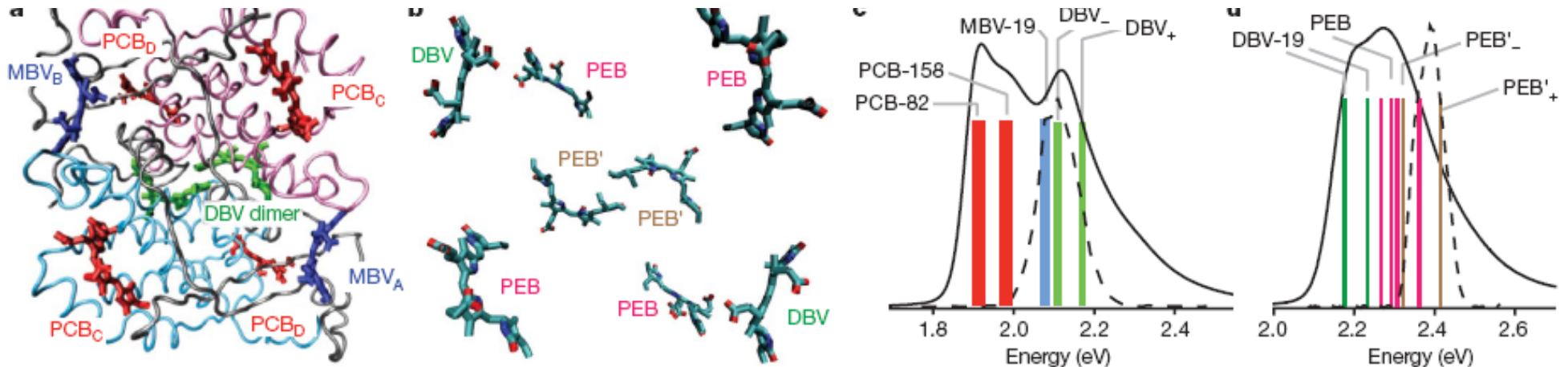


Figure 1 | Structure and spectroscopy of cryptophyte antenna proteins. **a**, Structural model of PC645. The eight light-harvesting bilin molecules are coloured red (PCB), blue (MBV) and green (DBV). **b**, Chromophores from the structural model for PE545 showing the different chromophore incorporation. **c**, Electronic absorption spectrum of isolated PC645 protein

in aqueous buffer (294 K). The approximate absorption energies of the bilin molecules are indicated as coloured bars. **d**, Electronic absorption spectrum of isolated PE545 protein in aqueous buffer (294 K) with approximate absorption band positions indicated by the coloured bars. The spectrum of the ultrafast laser pulse is plotted as a dashed line in **c** and **d**.

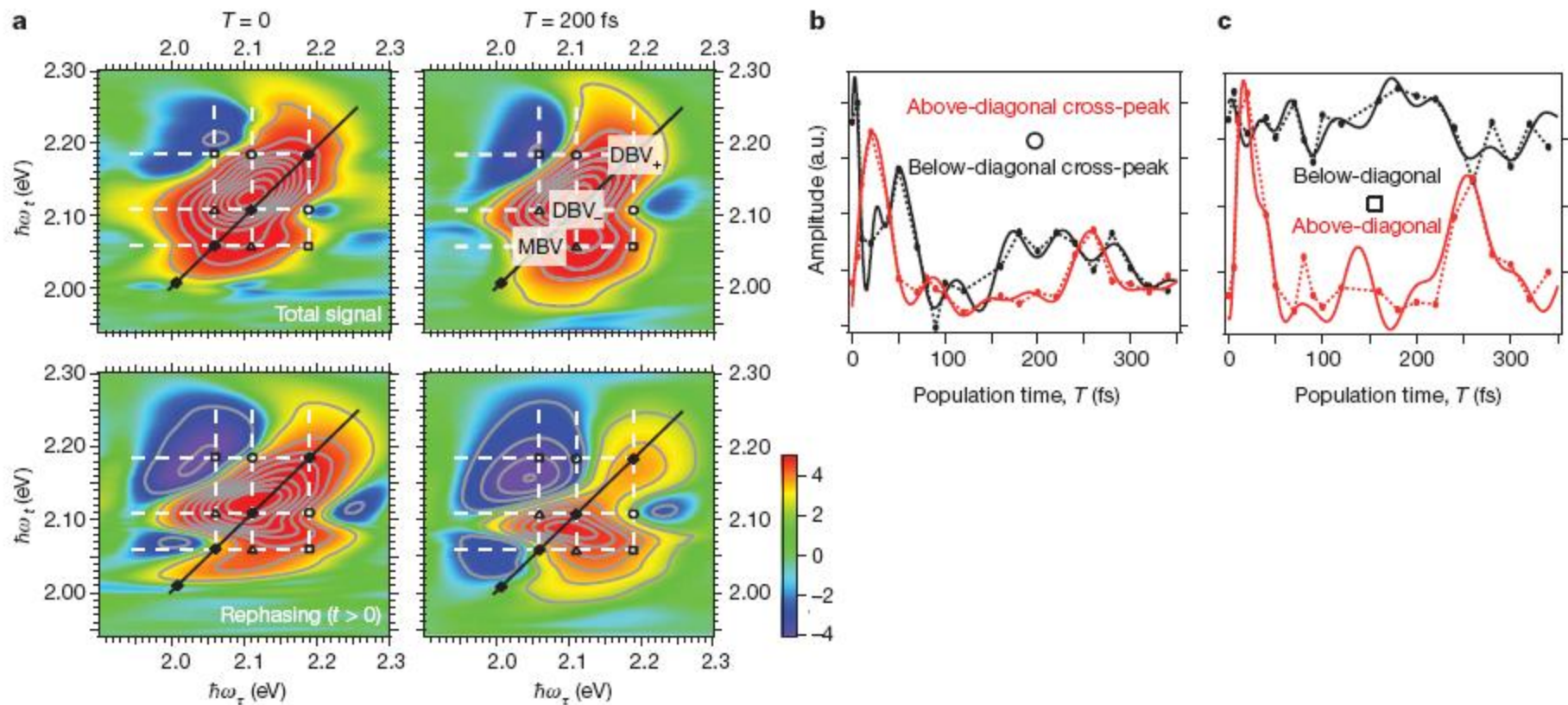


Figure 2 | Two-dimensional photon echo data for PC645. **a**, The left column shows the total real 2DPE spectrum recorded for PC645 at zero waiting time ($T = 0$), together with the rephasing contribution to this signal. The right column shows the data for $T = 200$ fs. The 2DPE spectra show the signal intensity on an arcsinh scale (colour scale, arbitrary units) plotted as a function

of coherence frequency ω_τ and emission frequency ω_r . **b**, Intensity of the DBV dimer cross-peaks (open circle) as a function of time T . **c**, Intensity of the MBV-DBV+ cross-peaks (open square) as a function of time T . The dashed lines interpolate the data points (solid circles). The solid line is a fit to a sum of damped sine functions (Supplementary Information). a.u., arbitrary units.

Hence., in both FMO and in PC645, both display long lived coherences, even at room temperature (even longer times observed since original FMO experiments).

Generated great deal of surprise/computational work--- to explain how electronic coherence survives for > 400 fs when, e.g.

a. Expected coherence time scales of 50 fs, or smaller – e.g.

b. Rossky, Prezhdov, Franco in semiconductors, polyacetylene wires, large molecules, etc. of times on order of 10 fs.

a. Typical approaches --- vast efforts --- e.g.

Theoretical Study of Coherent Excitation Energy Transfer in Cryptophyte Phycocyanin 645 at Physiological Temperature

Pengfei Huo[†] and David F. Coker^{*,†,‡}

acknowledge a grant of supercomputer time from Boston University's Office of Information Technology and Scientific Computing and Visualization as well as an allocation of supercomputing resources from the Irish Center for High End Computing (ICHEC). Thanks Niles.

Update --

Discussion still ongoing as to the nature of these coherences, (e.g. speakers this session) **vibrational or electronic** or mixed- with proposals for various schemes to test origins.

However, enthusiasm for the relevance of such observed coherences to “quantum biology” persist, as does my analysis below.

Hence--

Are These Coherences (and related discussion of entanglement) important in Nature?

Two examples where relevant---

Light Harvesting Complexes

Vision

Focus on the first---

Optics people in audience may be astounded at the confusion (propagated even by reputable physicists)

Issue:

Many photoexcitation experiments done with coherent laser light

Nature uses sunlight /moonlight, which is weak incoherent light.

Question: What is the relationship between these two cases?

**I.e., What, do the coherent light experiments
tell us about the natural case (incoherent light) ?**

**Are these observed coherences evidence for
coherent quantum contributions in Nature?**

**General question applicable to vision,
photosynthesis issues, etc. I.e. natural vs.
laboratory system preparation.**

For those who want their answers up front

What is the relationship between these two cases?

**I.e., What, do the coherent light experiments
tell us about the natural case (incoherent light) ?**

**They tell us about the nature of the Hamiltonian, and the couplings
between system and environment, etc.**

**Are these observed coherences evidence for
coherent quantum contributions in Nature?**

**No, since excitation under natural light and coherent light and
dramatically different. Indeed, in the natural light case the
molecular ensemble show no time evolving coherences!**

The general cases—

Isolated Systems

Open Systems

In either case qualitatively similar conclusions

Consider first the isolated case (i.e. isolated molecule irradiated with light)

Prior studies on the nature of excitation by incoherent and partially coherent light vs. coherent light (all isolated systems):

1. *General theory, with application to molecular excitation:*

Creation and dynamics of molecular states prepared with coherent vs partially coherent pulsed light

Xue-Pei Jiang and Paul Brumer

*Chemical Physics Theory Group, Department of Chemistry, University of Toronto,
Toronto M5S 1A1 Canada*

JCP, 94, 5833 (1991)

2. *Applications to Observations of Quantum Beats:*

Quantum beats induced by partially coherent laser sources

Xue-Pei Jiang and Paul Brumer

Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Toronto, Ontario, Canada M5S 1A1

Chem. Phys. Lett. 180, 222 (1991)

3. *Applications to Coherent Control:*

Pump–dump coherent control with partially coherent laser pulses

Xue-Pei Jiang, Moshe Shapiro,^{a)} and Paul Brumer

*Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Toronto,
M5S 1A1 Canada*

JCP, 104, 607. (1996)

State Preparation: Coherent vs. Incoherent Light

E.g, Laser light === coherent

Natural light (e.g. sunlight, moonlight) === incoherent

$$H = H_M - \mu[\bar{\epsilon}(t) + \bar{\epsilon}^*(t)] ,$$

where μ is the projection of the dipole operator along the field direction.

If initially in a pure matter state $|E_g\rangle$, weak field gives as resultant state:

$$\begin{aligned} |\phi(t)\rangle &= |E_g\rangle e^{-iE_g t/\hbar} \\ &+ \sum_i c_i \int_{-\infty}^t \bar{\epsilon}(t') e^{i(E_i - E_g)t'/\hbar} dt' |E_i\rangle e^{-iE_i t/\hbar}, \\ c_i &= \frac{1}{i\hbar} \langle E_i | \mu | E_g \rangle. \end{aligned}$$

Rewrite as density matrix:

$$\begin{aligned}\rho(t) = |\phi(t)\rangle\langle\phi(t)| &= \sum_{ij} c_i c_j^* \int dt' \bar{\epsilon}(t') e^{i(E_i - E_g)t'/\hbar} \\ &\times \int dt'' \bar{\epsilon}^*(t'') e^{-i(E_j - E_g)t''/\hbar} |E_i\rangle \\ &\times \langle E_j| e^{-i(E_i - E_j)t/\hbar}\end{aligned}$$

Source is not coherent? I.e. described statistically? Then need ensemble average

$$\begin{aligned}\rho(t) &= \sum_{ij} c_i c_j^* \int \int dt' dt'' e^{i\omega_{ig}t'} e^{-i\omega_{ig}t''} \\ &\times \langle \bar{\epsilon}(t') \bar{\epsilon}^*(t'') \rangle |E_i\rangle \langle E_j| e^{-i(E_i - E_j)t/\hbar}\end{aligned}$$

where omega... and braces denote average over ensemble of light

$$\rho(t) = \sum_{ij} c_i c_j^* \langle \epsilon(\omega_{ig}) \epsilon^*(\omega_{jg}) \rangle |E_i\rangle \\ \times \langle E_j | e^{-i(E_i - E_j)t/\hbar}$$

$$\langle \epsilon(\omega_{ig}) \epsilon^*(\omega_{jg}) \rangle = \int \int dt' dt'' e^{i\omega_{ig}t'} e^{-i\omega_{ig}t''} \\ \times \langle \bar{\epsilon}(t') \bar{\epsilon}^*(t'') \rangle ,$$

Coherent Source? --- E.g. pulsed laser---creates pure evolving state

$$\bar{\epsilon}(t) = \frac{1}{2} \epsilon e^{-i(\omega_0 t + \delta)} e^{-(t-t_0)^2/\tau^2} .$$

$$|\phi(t)\rangle = \sum_j \frac{\sqrt{\pi}}{2i\hbar} \epsilon \tau \langle E_j | \mu | E_g \rangle e^{-i(\omega_0 - \omega_{jk})t_0} \times e^{-\tau^2(\omega_0 - \omega_{jk})^2/4} e^{i\delta} |E_j\rangle .$$

$$\rho(t) = \sum_{ij} c_i c_j^* \langle \epsilon(\omega_{ig}) \epsilon^*(\omega_{jg}) \rangle |E_i\rangle \\ \times \langle E_j| e^{-i(E_i - E_j)t/\hbar}$$

$$\langle \epsilon(\omega_{ig}) \epsilon^*(\omega_{jg}) \rangle = \int \int dt' dt'' e^{i\omega_{ig}t'} e^{-i\omega_{ig}t''} \\ \times \langle \bar{\epsilon}(t') \bar{\epsilon}^*(t'') \rangle ,$$

General incoherent source? --- E.g. thermal light- → stationary state.

$$\langle \bar{\epsilon}(t_1) \bar{\epsilon}^*(t_2) \rangle = E_0^2 e^{-i\omega_{ij}(t_1 - t_2)} e^{-|t_1 - t_2|/\tau_d}$$

where τ_d is the mean time between collisions. Fourier transform: Get

$$\langle \epsilon(\omega_{ig}) \epsilon^*(\omega_{jg}) \rangle = 4\pi E_0^2 \delta(\omega_{ig} - \omega_{jg}) \times \frac{(1/\tau_d)}{[(\omega_0 - \omega_{ig})^2 + (1/\tau_d)^2]}$$

Aside: above is for source on forever.

If starts at time zero (“sunrise for a plant”) then there is very short transient coherence, followed by essentially mixture of stationary Hamiltonian eigenstates.

So---

**Laser source creates coherence over pulse time scale
(fs pulse → fs dynamics – (if allowed))**

**Whereas incoherent source creates stationary states
(e.g. sunlight → induces no quantum coherent dynamics)**

**Emphasize: Meaning of “no coherence” = “no coherent
dynamics”. Contrast with time dependent system energy
eigenstate populations**

**But argument ignored --- arguments advanced regarding
Photons, etc. --- so**

New version based on quantized radiation fields

New proof will be clear --- the first reviews?

A. All known --- don't publish

B. Great stuff – publish immediately to resolve issues

C. All wrong – can't publish TWICE!

Reflects the state of confusion in the field

Note, then, whatever your view here -- -you are not alone 😊

Two recent formal/computational approaches:

- 1. Isolated systems redone with quantized field
(allows better discussion of “photons”)**
- 2. Open systems – Exact computation on model system
(non-Markovian, secular approximation)**

I will summarize these results.

Details are:

- 1. Brumer and Shapiro, PNAS 109, 19575 (2012)**
- 2. Pachon and Brumer, Phys. Rev. A 87, 022106 (2013)**

$$|R_i\rangle = \hat{\epsilon} \sum_{N_1, N_2, \dots, N_{max}} c(N_1, N_2, \dots, N_{max}) |N_1\rangle |N_2\rangle \dots |N_{max}\rangle,$$

For computational simplicity we also use the notation

$$|R_i\rangle = \hat{\epsilon} \sum_{\mathbf{N}} c(\mathbf{N}) |\mathbf{N}\rangle.$$

where $\mathbf{N} = (N_1, N_2, \dots, N_{max})$.

Initial State: $|\Psi_i\rangle = |R_i\rangle |E_i\rangle.$

One photon absorption produces excited state:

$$|\Psi_f\rangle = \frac{2\pi i \hat{\epsilon}}{\hbar} \sum_k \sum_{\mathbf{m}} \varepsilon(N_k, \omega_k) |E_k, \mathbf{m}\rangle \langle E_k, \mathbf{m} | \hat{\epsilon} \cdot \mathbf{d} | E_i \rangle \times \\ \sum_{\mathbf{N}} c(\mathbf{N}) |N_1\rangle \dots |N_{k-1}\rangle |N_k - 1\rangle |N_{k+1}\rangle \dots |N_{max}\rangle,$$

With field amplitude:

$$\varepsilon(N_k, \omega_k) = i \left(\frac{\hbar \omega_k N_k}{\epsilon_0 V} \right)^{\frac{1}{2}} \exp(i\omega_k z/c),$$

Create density matrix and trace over the (unobserved) radiation field gives state of the molecule

$$\begin{aligned} \rho_{mol} &= \sum_{\mathbf{N}'} \langle \mathbf{N}' | \Psi_f \rangle \langle \Psi_f | \mathbf{N}' \rangle \\ &= \sum_{\mathbf{N}, \mathbf{m}, \mathbf{m}', k} |c(\mathbf{N})|^2 |A(k, \mathbf{m})\rangle \langle A(k, \mathbf{m}')| + \sum_{\mathbf{N}, \mathbf{m}, \mathbf{m}', k' > k} [d_{k', k} |A(k, \mathbf{m})\rangle \langle A(k', \mathbf{m}')| + cc] \end{aligned}$$

where

$$d_{k', k} = c(N_1, N_2, \dots, N_{k'-1}, N_{k'} - 1, N_{k'+1}, \dots, N_{max}) \quad c^*(N_1, N_2, \dots, N_{k-1}, N_k - 1, N_{k+1}, \dots, N_{max})$$

$$|A(k, \mathbf{m})\rangle = \frac{2\pi i}{\hbar} \varepsilon(N_k, \omega_k) |E_k, \mathbf{m}\rangle \langle E_k, \mathbf{m} | \hat{\epsilon} \cdot \mathbf{d} | E_i \rangle$$

Pulsed laser case (after pulse is over):

$$\begin{aligned} \rho_{mol}(t > t_0) = & \sum_{\mathbf{N}, \mathbf{m}, \mathbf{m}', k} |c(\mathbf{N})|^2 |A(k, \mathbf{m})\rangle \langle A(k, \mathbf{m}')| \\ & + 2 \sum_{\mathbf{N}, \mathbf{m}, \mathbf{m}', k' > k} \text{Re}[d_{k', k} |A(k, \mathbf{m})\rangle \langle A(k', \mathbf{m}')| \exp[-i(E_k - E_{k'})(t - t_0)/\hbar]] \end{aligned} \quad (11)$$

Once again:

Off diagonal elements in the energy eigenbasis

→ coherent time evolution

Consider now absorption of light emitted by an incoherent thermal source, such as sunlight. This source consists of a statistical mixture of number states described by a radiation field density matrix¹⁰:

$$\rho_R = \sum_{\mathbf{N}} p_{\mathbf{N}} |\mathbf{N}\rangle \langle \mathbf{N}|. \quad (12)$$

Here $p_{\mathbf{N}}$ is the probability of finding the number state $|\mathbf{N}\rangle$ in the radiation emitted from the thermal source. If the source is at temperature T this is given by:

$$p_{\mathbf{N}} = \prod_k \frac{(\bar{N}_k)^{N_k}}{(1 + \bar{N}_k)^{1+N_k}} \quad (13)$$

with \bar{N}_k being the mean number of photons at temperature T : $\bar{N}_k = [\exp(\hbar\omega_k/k_B T) - 1]^{-1}$. This radiation field is a statistical mixture of number states. As a consequences, irradiation with this source will yield the result of an uncorrelated collection of states resulting from excitation with the state $|\mathbf{N}\rangle \langle \mathbf{N}|$.

Excitation with the state $|\mathbf{N}\rangle\langle\mathbf{N}|$

$$\rho_{mol} = \sum_{\mathbf{m}, \mathbf{m}'} |A(k, \mathbf{m})\rangle\langle A(k, \mathbf{m}')|$$

$$|A(k, \mathbf{m})\rangle = \frac{2\pi i}{\hbar} \varepsilon(N_k, \omega_k) |E_k, \mathbf{m}\rangle\langle E_k, \mathbf{m}|\hat{\epsilon} \cdot \mathbf{d}|E_i\rangle$$

Lesson again, excitation with incoherent light gives stationary states.

But now can address numerous arguments proposed regarding single photon excitation In weak fields, during various discussions:

“Coherence of the molecule is independent of the source of excitation”

Since focused on time dependent coherences, this is wrong.

“With weak light, the photons arrive at widely separated times. When they do:

Incident photon gives the molecule “a kick” which initiates dynamics.”

The statement that the photons arrive at widely separated times is only meaningful if we are making a measurement of arrival times. We are not making such a measurement.

This particle picture of light is only meaningful if we are making a measurement that detects particle like properties. We are not doing such a measurement.

And:

“With weak light, the photons arrive at widely separated times. When they do:

Incident photon gives the molecule “a kick” which initiates dynamics.”

If you actually want to talk about photons then the following is **not possible** in isolated molecule:

No : Stationary state + fixed energy photon \rightarrow time evolving state

(Although published recently in the literature!)

But incoherent light can be composed of unrelated fs pulses—
hence fs pulsed Initial dynamics is relevant to incoherent case.

Not relevant:

1. The physics is in the ensemble average over these realizations, and the average gives zero coherent dynamics.
2. Any basis for $E(t)$ (of limited frequency width) can be used, e.g. nanosecond lasers, so no central role for fs sources
3. If any basis for $E(t)$ is indeed preferred it is the true random phase CW-like spontaneous emission with Wiener noise.

Careful to note:

1. Any perturbation will cause initial coherent excitation due to the turn-on effect.
2. As shown in the 1991 paper, any pulsed incoherent source will induce dynamics on the time scale of the pulse (due to coherent pulse envelope --- “the molecule is not stupid”)

BUT— here in natural processes the turn-one effect is very short time (20 fs ?) And the envelope induced dynamics is on time scale of the day ---

Essentially we establish steady state with the thermal light source.

But what about the open system case?

(See also related results obtained by Mancal and Ivan Kassal).

Incoherent Excitation of Open Quantum Systems

Leonardo A. Pachón^{*,†,‡} and Paul Brumer^{*,†}

Phys. Rev. A 87, 022106 (2013)

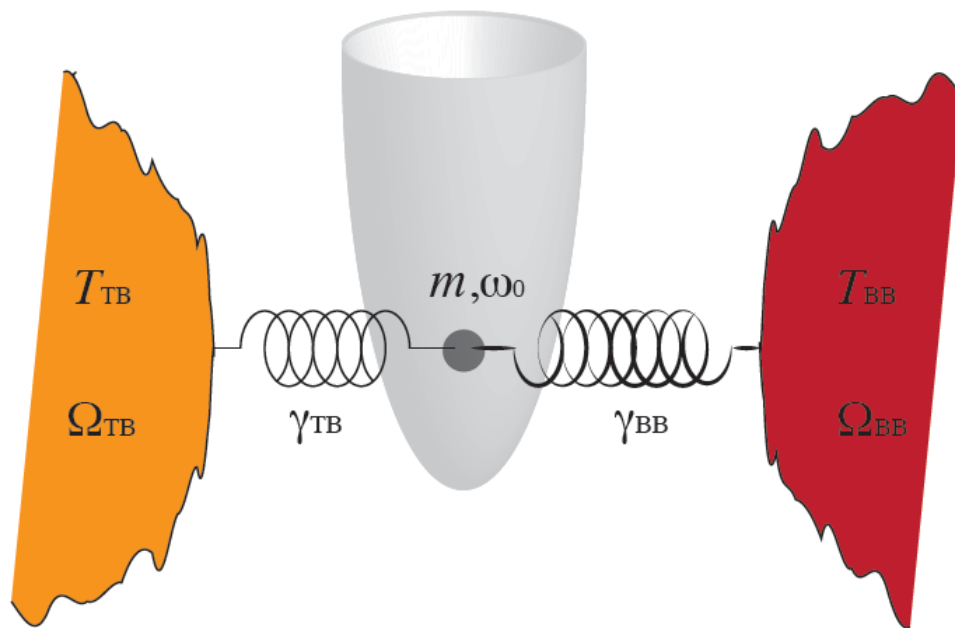


Figure 5: After thermalizing with TB, the system S is put in contact with a second thermal bath TB' (BB in the plot) at different temperature and different coupling constant.

Charged h.o., linear
coupling (p and q)

Note: Is exact, Non-Markovian result

Influence Functional --- Beautiful Result

(reminiscent of Kraus, but not)

$$\begin{aligned}\langle n | \hat{\rho}_S(t) | m \rangle &= \sum_{\nu} J_{nm;\nu\nu}(t) \langle \nu | \hat{\rho}_S(0) | \nu \rangle \\ &+ \sum_{\nu \neq \mu} J_{nm;\nu\mu}(t) \langle \nu | \hat{\rho}_S(0) | \mu \rangle,\end{aligned}$$

Oscillator with Single Bath

$$\hat{H} = \hat{H}_S + \sum_j \frac{\hat{p}_j^2}{2m_j} + \frac{m_j \omega_j^2}{2} \left(\hat{q}_j - \frac{c_j \hat{q}_x}{m_j \omega_j^2} \right)^2,$$

$$\hat{H}_S = \frac{1}{2m} \hat{p}_x^2 + \frac{m \omega_0^2}{2} \hat{q}_x^2,$$

Oscillator with Two Baths (Heat Bath or Thermal Light)

$$\hat{H} = \hat{H}_S + \hat{H}_{TB} + \hat{H}_{TB'} + \hat{H}_{ST} + \hat{H}_{SB'},$$

Result ---

Excite System with Natural Light →

relaxation in the presence of natural light

Relaxation time scales? psec max

Sunlight is on ALL DAY.

I.e. Only see transient initial dynamics. Then stationary.

Hence coherences are not physically relevant (to, e.g. plant)

E.g. with exaggerated parameters:

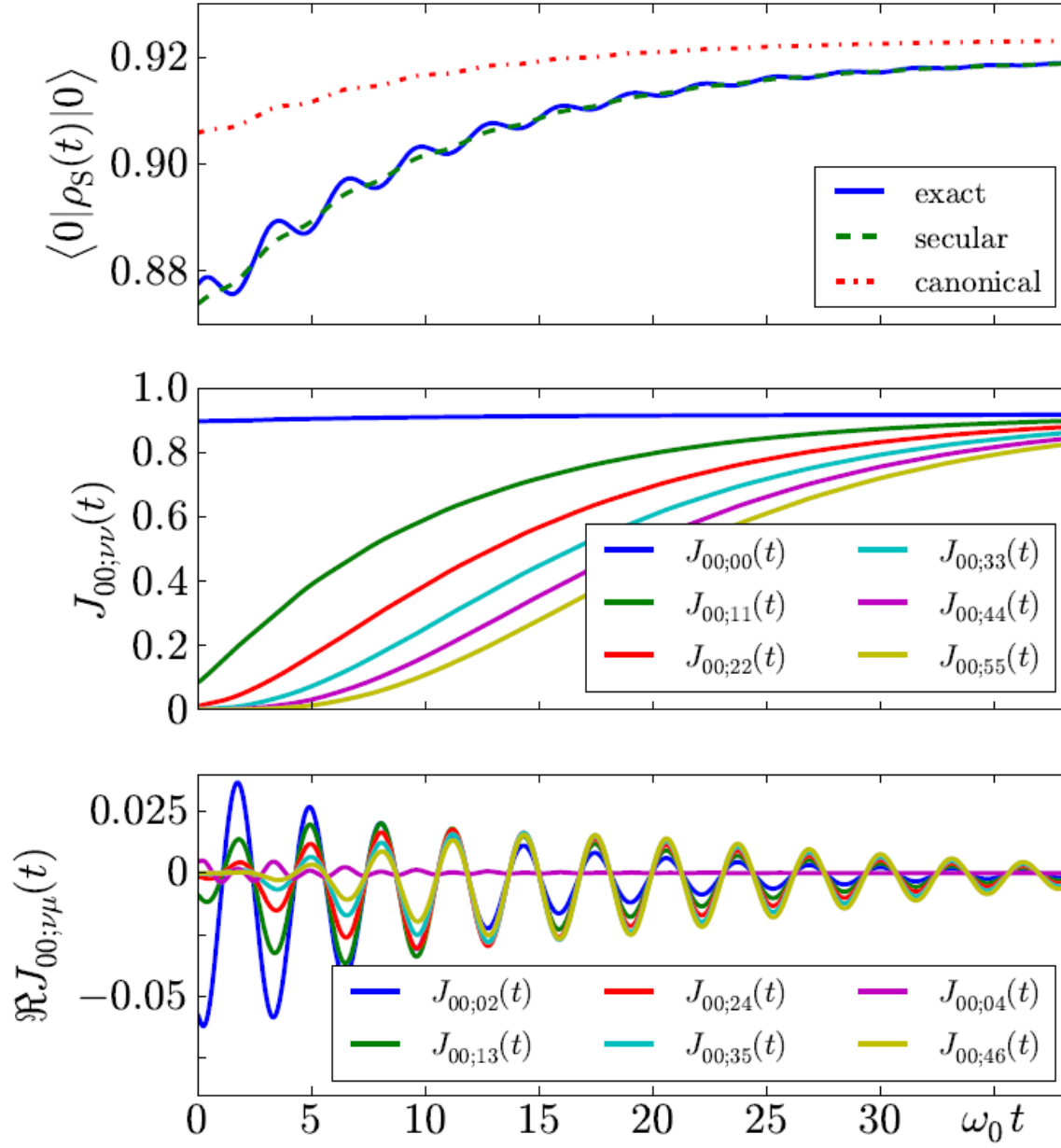


FIG. 8. Parameters for coupling to TB as in 7, with $\hbar\omega_0/k_B T_{\text{BB}} = 0.3884$, and $\Omega_{\text{BB}} = 8.3 \times 10^5 \omega_0$.

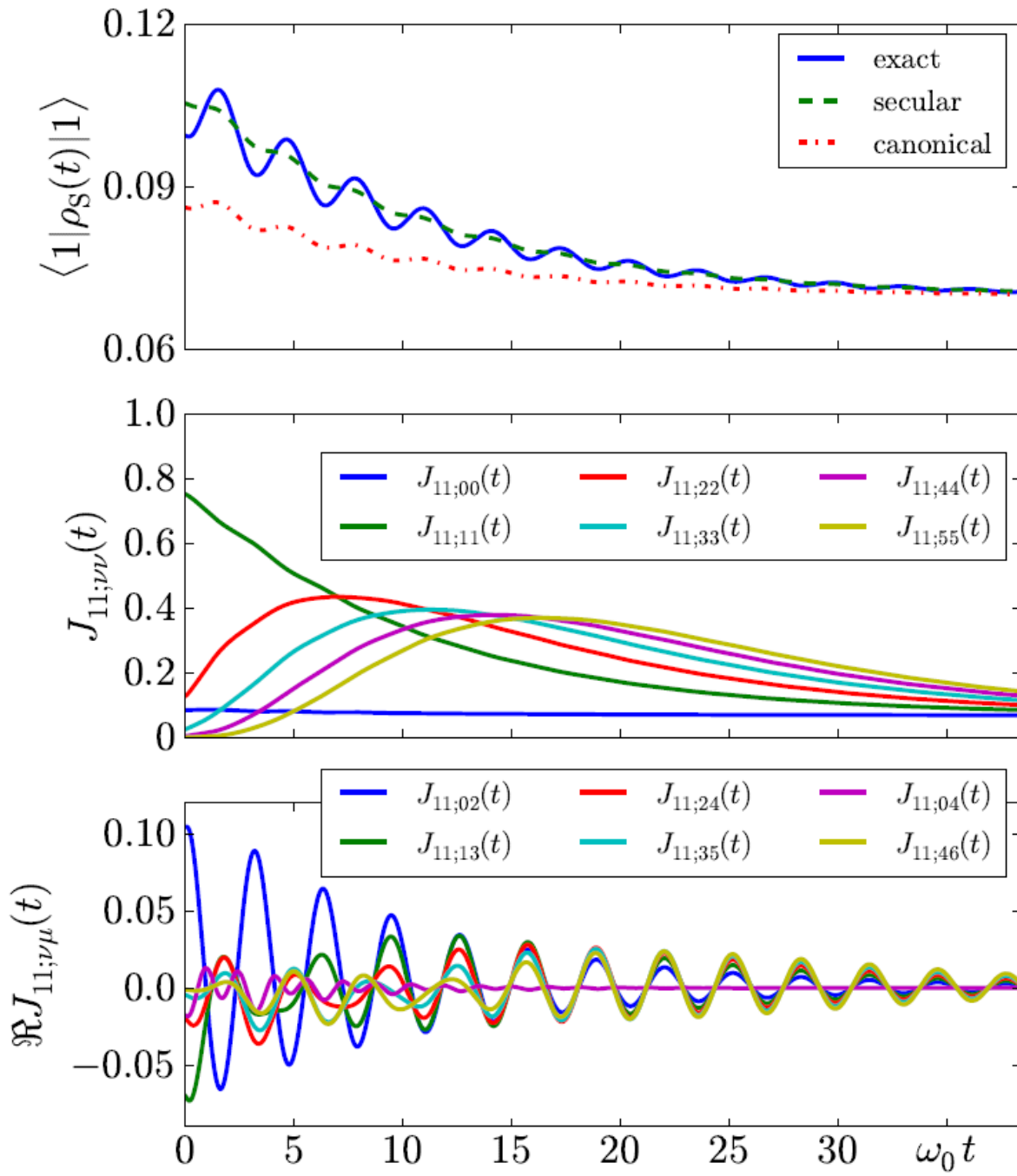
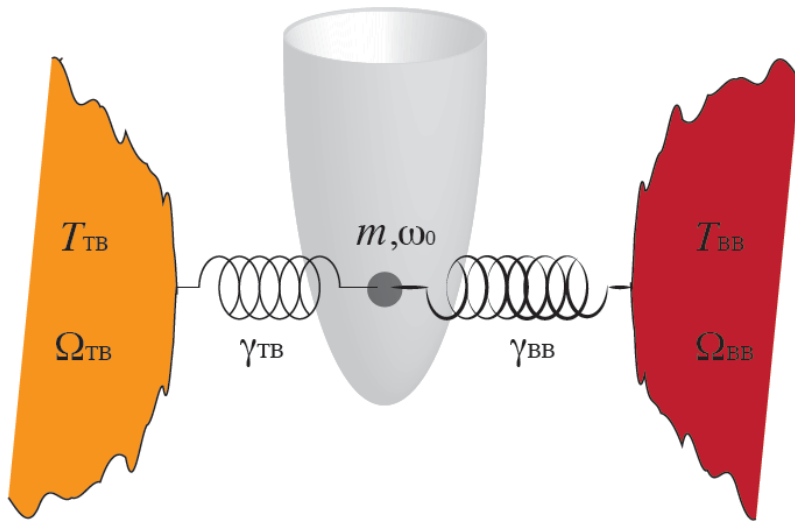


FIG. 9. Parameters as in 8.

Indeed relaxation is primarily to temperature of the surrounding bath---the coupling with the light is too weak



Hence the system ultimately shows steadystate heat transfer, from the hot blackbody source to the cool environment --- but not coherent dynamics.

Some additional comments:

Experiments: molecules are isolated from full environment.
But even in-vivo, entire apparatus Irradiated (20 nm vs 500 nm).
Hence, not localized excitation--- but full energy eigenstates.

Situation similar in retinal re rates --- see K. Hoki and P. B.,
Proc. Chem. 3, 122 (2011); T. Tscherbul and PB (in prep)

Oscillations do not necessarily imply coherent dynamics ---
See I. Franco and P.B. (submitted)

Hit a molecule with incoherent light-- it will ring transiently (studied).
Regeneration scenarios? None found – Z. Sadeq (M.Sc.) and PB

Structure of coherent and incoherently prepared states? See
A. Han, M. Shapiro and PB (submitted and in quant-ph ArXiv) etc...

Summary:

2DPE experiments highly enlightening about the system Hamiltonian, system-bath interactions, persistence of coherence under coherent excitation.

**Are the 2DPE echo coherences relevant in nature?
Doubtful, since excitation is with incoherent light—
either in open or closed system, gives relaxation to
steady state.**

Still many interesting questions relating to entire coupled (living) system.

Thanks to:

Leonardo Pachon, postdoc, University of Toronto

Xupei Jiang, very early postdoc, Univ of Toronto

Moshe Shapiro, on quantized field work

AND

\$\$ NSERC plus AFOSR \$\$