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# Laser assisted resonance energy transfer

Philip Allcock<sup>1</sup>, Robert D. Jenkins<sup>2</sup>, David L. Andrews<sup>\*</sup>

School of Chemical Sciences, University of East Anglia, Norwich, NR4 7TJ, UK

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#### Abstract

At distances beyond wavefunction overlap, and typically running up to 100 Å, the migration of electronic excitation between donor and acceptor species is generally understood to approximate well to the resonance energy transfer theory developed by Förster. Here we demonstrate that the energy transfer rate can be increased by propagation of an auxiliary laser beam through the sample. Results indicate that, under optimum conditions, auxiliary beam intensities of  $10^{10}$  W cm<sup>-2</sup> can produce an increase in the energy migration rate by as much as 15%. © 1999 Elsevier Science B.V. All rights reserved.

#### 1. Introduction

The migration of energy between an electronically excited atomic or molecular donor species and a neighboring acceptor in its ground electronic state is a well-established phenomenon of ultrafast photochemistry in the condensed phase and it plays, inter alia, an important role in energy storage and migration in photosynthetic systems. It also affords an important tool in the determination of molecular architecture where it is used, mainly within large biological structures, as a so-called spectroscopic ruler [1-4]. Here, for example, the inverse sixth power dependence of the intermolecular separation that features in Förster's theory [5] is utilised in calculating the molecular separation of fluorophores in proteins. This technique affords a means of com-

\* Corresponding author. E-mail: d.l.andrews@uea.ac.uk

paring structures or ascertaining conformational change [6,7] with a resolution of a few ångström.

CHEMICAL PHYSICS

The process of resonance energy transfer (RET) was first discussed in terms of quantum electrodynamics (OED) in pioneering studies by Avery [8] alongside Gomberoff and Power [9]. These studies showed that long-range retardation effects embellish a short-range static coulombic interaction. In the long-range transfer asymptote a photon is first emitted by the donor (A) and then captured by an acceptor (B), the rate of transfer being governed by an inverse square law with respect to donor-acceptor separation R. As such, both short-range (radiationless) and long-range (radiative) transfer are correctly described as limits of a unified theory operating over all R [10]. More recently, research into the QED of condensed matter RET processes [11,12] has led to the formulation of modified radiation field operators which take into account the full effect of an intervening medium [13].

RET plays a major role in systems where donoracceptor separations are generally of the order of

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<sup>&</sup>lt;sup>1</sup> E-mail: p.allcock@uea.ac.uk

<sup>&</sup>lt;sup>2</sup> E-mail: robert.jenkins@uea.ac.uk

tens of ångström. In this regime energy migration is essentially between moieties beyond significant wavefunction overlap. The most important role for RET is illustrated by the harvesting of light by green plants, a key stage in photosynthesis. Energy from sunlight is captured by antenna chlorophyll molecules and migrates with great efficiency to a terminal reaction centre; this affords an example of RET between units separated by distances up to ~ 100 Å. This is only one example of where short-range transfer occurs, but it serves to illustrate the fluorophore separations that offer most efficient transfer. Most RET studies focus on donor–acceptor separations within this distance regime, where long-range behaviour is seldom significant.

Here we discuss the effects on the energy transfer process due directly to a laser beam impinging on a donor-acceptor system. In the light of the above remarks, we shall concentrate exclusively on the dominant short-range regime. This phenomenon, termed laser assisted resonance energy transfer (LARET), is similar to spontaneous RET in that we commence with a prepared donor state, resulting from prior excitation by a primary source which plays no further role in the process. We consider LARET as resonance energy transfer in a system illuminated by an auxiliary laser field. This entails



Fig. 1. Time-ordered diagrams showing applied laser photons absorbed at A and emitted at B in tandem with energy transfer, the short-range limit permitting the transfer to be represented by a horizontal tie-line. Thus, six different time-orderings can be generated by index permutation;  $\alpha$  and  $\beta$  are molecular excited states; 0 a molecular ground state; *s* and *t* the virtual intermediate states.



Fig. 2. Converse process to that shown in Fig. 1, where photons are absorbed from and emitted back into the laser beam at one site.

the coupled absorption and stimulated emission of photons from and into the applied beam, the overall process accomplishing the same net transfer of energy as in RET. Two discernible mechanisms contribute to LARET, necessarily resulting from higher-order perturbational contributions to RET: firstly, as typified by the time-ordered diagram of Fig. 1, processes which embrace laser photon absorption at A (or B) and emission at B (or A), secondly, as in Fig. 2, where the laser photon absorption and emission events occur at one site (A or B).

Although the final state of the impinging radiation field is indistinguishable from its initial state, the latent effect on RET only becomes apparent through interpretation in terms of individual quanta, where spontaneous emission is properly placed in the context of its stimulated counterpart [14]. The ensuing rate equations exhibit new and important correction terms due to the presence the laser field, manifest as LARET. Both linear and quadratic effects are identified, and it is shown that their relative significance is determined by the laser intensity.

### 2. Theory

In the QED formalism short-range RET is represented by the time-ordered diagram given in Fig. 3. The laser-dependent aspect of the LARET phenomenon relates to interactions which can be de-



Fig. 3. Time-ordered diagram for short-range RET.

scribed with the aid of 12 diagrams, time-index permutations of Figs. 1 and 2. In each case the energy transfer is mediated by exchange of a *virtual photon* (deemed 'virtual' since it is not observed), described in terms of a sum over all possible wavevectors (p) and polarisations (l). As a consequence of the uncertainty principle these virtual photons have a high uncertainty in energy, due to their short propagation time.

The full Hamiltonian, *H*, is given by:

$$H = H_{\rm mol}^{\rm A} + H_{\rm mol}^{\rm B} + H_{\rm int}^{\rm A} + H_{\rm int}^{\rm B} + H_{\rm rad} , \qquad (1)$$

where  $H_{\text{mol}}^{\xi}$  is the molecular Hamiltonian for molecule  $\xi$  and  $H_{\text{rad}}$  represents the second-quantised radiation field. Within the electric dipole approximation utilised here, the molecule-field coupling Hamiltonian,  $H_{\text{int}}^{\xi}$ , is explicitly:

$$H_{\text{int}}^{\xi} = -\varepsilon_0^{-1} \boldsymbol{\mu}(\xi) \cdot \boldsymbol{d}^{\perp}(\boldsymbol{R}_{\xi}), \qquad (2)$$

with  $\mu(\xi)$  the electric dipole moment operator and  $R_{\xi}$  the position vector for  $\xi$ . The transverse electric displacement field operator,  $d^{\perp}(R_{\xi})$ , can be expressed in terms of a mode expansion for both vacuum formulation [14] and, incorporating media influences, a host or solvent 'bath' [13]. For presentational simplicity Eq. (3) addresses the vacuum expansion for a photon of wavevector p and polarisation l:

$$d^{\perp}(\boldsymbol{R}_{\xi}) = \sum_{\boldsymbol{p},l} \left(\frac{\hbar c \boldsymbol{p} \boldsymbol{\varepsilon}_{0}}{2V}\right)^{1/2} i \left[\boldsymbol{e}^{(l)}(\boldsymbol{p}) a^{(l)}(\boldsymbol{p}) e^{i\boldsymbol{p}\cdot\boldsymbol{R}_{\xi}} - \bar{\boldsymbol{e}}^{(l)}(\boldsymbol{p}) a^{\dagger(l)}(\boldsymbol{p}) e^{-i\boldsymbol{p}\cdot\boldsymbol{R}_{\xi}}\right], \qquad (3)$$

where e is the electric field unit vector ( $\bar{e}$  being its complex conjugate), a and  $a^{\dagger}$  are annihilation and creation operators, respectively, for that photon, and V is the quantisation volume.

We now introduce the quantum probability amplitude or *matrix element*,  $M_{FI}$ , connecting the initial system state,  $|I\rangle$ , to the final system state,  $|F\rangle$ ,  $M_{FI}$ is expressed through the time-dependent perturbation expansion:

$$M_{FI} = \sum_{m=1}^{\infty} M_{FI}^{(m)}, \qquad (4)$$

with the leading contributions to energy transfer, RET, represented by the second-order (m = 2) term of Eq. (4):

$$M_{Fl}^{(2)} = \sum_{S} \frac{\langle F|H_{\rm int}|S\rangle\langle S|H_{\rm int}|I\rangle}{(E_I - E_S)} \,. \tag{5}$$

Here  $E_N \equiv E_n^{(\text{mol})} + E_n^{(\text{rad})}$  is the energy of the system states  $|N\rangle$ , comprising products of both molecular and radiation states  $|N\rangle = |n_{(\text{mol})}\rangle |n_{(\text{rad})}\rangle$ .

The first corrections offered by the LARET process to the energy transfer matrix element (5), are derived from the fourth-order (m = 4) perturbation summed over three intermediate states *S*, *T* and *U*:

$$M_{FI}^{(4)} = \sum_{S,T,U} \frac{\langle F|H_{\text{int}}|U\rangle\langle U|H_{\text{int}}|T\rangle\langle T|H_{\text{int}}|S\rangle\langle S|H_{\text{int}}|I\rangle}{(E_I - E_S)(E_I - E_T)(E_I - E_U)} .$$
(6)

The rate for the modified energy transfer process can be described using Fermi's Golden rule together with Eq. (4):

$$\Gamma = \frac{2 \pi \rho}{\hbar} \left| M_{FI}^{(2)} + M_{FI}^{(4)} + M_{FI}^{(6)} + \cdots \right|^2,$$
(7)

where  $\rho$  is the density of final molecular states of the acceptor molecule. The second- and fourth-order perturbation are sufficient to reveal the LARET effect; as the series rapidly converges, higher contributions being negligible.

### 3. Results

The consideration of the short-range limit, explicitly for donor-acceptor separations where  $kR \ll 1$ , obviates the retardation effects that naturally emerge using our formalism [10,15,16]. For the pertinent terms in Eq. (7) we explicitly calculate the individual matrix elements, the following expressions being cast in a form utilising the convention of implied summation over repeated indices:

$$M_{FI}^{(2)} = -\mu_i^{0\,\alpha(\mathrm{A})} \frac{\left(\delta_{ij} - 3\,\hat{R}_i\,\hat{R}_j\right)}{4\,\pi\varepsilon_0\,R^3} \mu_j^{\beta\,0(\mathrm{B})}\,,\tag{8}$$

$$M_{FI}^{(4)} = -\frac{n\hbar ck}{2\varepsilon_0 V} \left\{ e_i^{(\lambda)}(\boldsymbol{k}) \alpha_{ij}^{0\alpha(\Lambda)}(\boldsymbol{k}) \frac{\left(\delta_{jk} - 3\hat{R}_j\hat{R}_k\right)}{4\pi\varepsilon_0 R^3} \right. \\ \times \alpha_{kl}^{\beta0(B)}(\boldsymbol{k}) \bar{e}_l^{(\lambda)}(\boldsymbol{k}) + \bar{e}_i^{(\lambda)}(\boldsymbol{k}) \alpha_{ij}^{0\alpha(\Lambda)}(\boldsymbol{k}) \\ \times \frac{\left(\delta_{jk} - 3\hat{R}_j\hat{R}_k\right)}{4\pi\varepsilon_0 R^3} \alpha_{kl}^{\beta0(B)}(\boldsymbol{k}) e_l^{(\lambda)}(\boldsymbol{k}) \\ + e_i^{(\lambda)}(\boldsymbol{k}) \bar{e}_j^{(\lambda)}(\boldsymbol{k}) \beta_{ijk}^{0\alpha(\Lambda)}(\boldsymbol{k}) \frac{\left(\delta_{kl} - 3\hat{R}_k\hat{R}_l\right)}{4\pi\varepsilon_0 R^3} \\ \times \mu_l^{\beta0(B)} + \mu_i^{0\alpha(\Lambda)} \frac{\left(\delta_{ij} - 3\hat{R}_i\hat{R}_j\right)}{4\pi\varepsilon_0 R^3} \\ \times \beta_{jkl}^{\beta0(B)}(\boldsymbol{k}) e_k^{(\lambda)}(\boldsymbol{k}) \bar{e}_l^{(\lambda)}(\boldsymbol{k}) \right\}, \qquad (9)$$

where we define the intermolecular vector  $\mathbf{R} \equiv \mathbf{R}_{\rm B}$  $-\mathbf{R}_{\rm A}$ ,  $\boldsymbol{\mu}^{xy(\xi)} \equiv \langle x | \boldsymbol{\mu}(\xi) | y \rangle$  and  $\boldsymbol{\delta}$  is the Kronecker delta isotropic tensor. In Eq. (9) the generalised polarisabilities  $\boldsymbol{\alpha}^{fi(\xi)}(k)$  and hyperpolarisabilities  $\boldsymbol{\beta}^{fi(\xi)}(k)$  are defined as:

$$\alpha_{ij}^{fi(\xi)}(k) = \sum_{s} \left\{ \frac{\mu_{l}^{fs} \mu_{j}^{si}}{(E_{si} - \hbar ck - i\Gamma_{s})} + \frac{\mu_{j}^{fs} \mu_{i}^{si}}{(E_{si} + \hbar ck - i\Gamma_{s})} \right\}$$
(10)

and

$$\beta_{ijk}^{fi(\xi)}(k) = \sum_{s,t} \left\{ \frac{\mu_{i}^{f} \mu_{j}^{ts} \mu_{k}^{si}}{(E_{si} - \hbar ck - i\Gamma_{s})(E_{ti} - \hbar ck - i\Gamma_{t})} + \frac{\mu_{i}^{ft} \mu_{k}^{ts} \mu_{j}^{si}}{(E_{si} - i\Gamma_{s})(E_{ti} - \hbar ck - i\Gamma_{t})} + \frac{\mu_{k}^{ft} \mu_{i}^{ts} \mu_{j}^{si}}{(E_{si} - \hbar ck - i\Gamma_{s})(E_{ti} - i\Gamma_{t})} + \frac{\mu_{j}^{ft} \mu_{i}^{ts} \mu_{k}^{si}}{(E_{si} + \hbar ck - i\Gamma_{s})(E_{ti} + \hbar ck - i\Gamma_{t})} + \frac{\mu_{j}^{ft} \mu_{k}^{ts} \mu_{k}^{si}}{(E_{si} - i\Gamma_{s})(E_{ti} + \hbar ck - i\Gamma_{t})} + \frac{\mu_{k}^{ft} \mu_{j}^{ts} \mu_{k}^{si}}{(E_{si} - i\Gamma_{s})(E_{ti} + \hbar ck - i\Gamma_{t})} + \frac{\mu_{k}^{ft} \mu_{j}^{ts} \mu_{k}^{si}}{(E_{si} + \hbar ck - i\Gamma_{s})(E_{ti} - i\Gamma_{t})} \right\}, \quad (11)$$

incorporating  $E_{xy} \equiv E_x^{(\text{mol})} - E_y^{(\text{mol})}$ . Proper accommodation of resonance behaviour is achieved by including complex energy addenda in the denominators, where  $\Gamma_s$  for example represents the molecular intermediate state lifetime of virtual state  $|s_{(\text{mol})}\rangle$ , signs consistent with time reversal arguments [17]. Superscript state labels on the molecular tensors are dropped henceforth for brevity.

Describing the laser beam as comprising *n* photons with wave-vector **k** and polarisation  $\lambda$  allows us to define a laser intensity  $I(k) \equiv n\hbar c^2 k/V$ . Substitution into Eq. (9) reveals the linear dependence on the applied laser power in each of its four fourth-order contributions; the first and second terms relate to processes of the kind shown in Fig. 1, and the third and fourth, those associated with Fig. 2. In calculating an overall rate through Eq. (7) we first have a conventional term independent of the laser intensity:

$$\Gamma^{(2-2)} = \frac{2 \pi \rho_f}{\hbar (4 \pi \varepsilon_0 R^3)^2} |\mu^{(A)}|^2 |\mu^{(B)}|^2$$
$$\times \{\cos \theta - 3 \cos \phi \cos \psi\}^2$$
$$= \frac{2 \pi \rho_f}{\hbar (4 \pi \varepsilon_0 R^3)^2} |\mu^{(A)}|^2 |\mu^{(B)}|^2 \kappa^2, \qquad (12)$$

where  $\theta$  is the angle between the donor and acceptor transition moments and  $\phi$  and  $\psi$  the angles between individual donor and acceptor moments and the intermolecular separation vector **R**, respectively. Eq. (12) represents the normal Förster 'coulombic' energy transfer rate,  $\Gamma^{(2-2)}$ , derived from the secondorder matrix element (5), as indicated by the superscript. Here the angular dependence relates to the so-called  $\kappa^2$  factor [18]. Secondly, we have a term dependent on the square of the applied laser intensity as construed from Eq. (7). Explicitly:

$$\Gamma^{(4-4)} = \frac{I^2(k)\rho}{32\hbar c^2 \pi \varepsilon_0^4 R^6} |a' + a'' + b' + b''|^2, \qquad (13)$$

where the parameters a', a'', b' and b'' are defined as:

$$\begin{aligned} d' &\equiv \left\{ e_i \alpha_{ij}^{(A)}(k) \Big( \delta_{jk} - 3\hat{R}_j \hat{R}_k \Big) \alpha_{kl}^{(B)}(k) \bar{e}_l \right\}, \\ a'' &\equiv \left\{ \bar{e}_i \alpha_{ij}^{(A)}(k) \Big( \delta_{jk} - 3\hat{R}_j \hat{R}_k \Big) \alpha_{kl}^{(B)}(k) e_l \right\}, \\ b' &\equiv \left\{ e_i \bar{e}_j \beta_{ijk}^{(A)}(k) \Big( \delta_{kl} - 3\hat{R}_k \hat{R}_l \Big) \mu_l^{(B)} \right\}, \\ b'' &\equiv \left\{ \mu_i^{(A)} \Big( \delta_{ij} - 3\hat{R}_i \hat{R}_j \Big) \beta_{jkl}^{(B)}(k) e_k \bar{e}_l \right\}. \end{aligned}$$

Here the considerably more intricate angle dependence is most clearly expressed by retention of the tensor notation. Finally, Eq. (7) yields a cross term, linearly dependent on laser intensity:

$$\Gamma^{(2-4)} = \frac{I(k)\rho}{16\pi\varepsilon_0^3 \hbar c R^6} |\mu^{(A)}|| \,\mu^{(B)}|\kappa \Big[ 2\,\text{Re}\big\{ \vec{a}' + \vec{a}'' + \vec{b}' + \vec{b}'' \big\} \Big].$$
(14)

The weight-two tensor  $(\delta_{jk} - 3\hat{R}_j\hat{R}_k)$  embedded in Eq. (14), together with the angle-dependent  $\kappa$  factor, will ultimately determine the sign of  $\Gamma^{(2-4)}$ ; in the case of  $\Gamma^{(4-4)}$  a positive (or null) result always ensues.

Assuming the molecules have a fixed orientation the total rate for the newly derived LARET process is given by the sum of the results from Eqs. (12)– (14). Where molecules are randomly oriented as in a molecular fluid then a rotational average has to be taken. Involving high-order isotropic averages, this is an intricate process which does little to illuminate the underlying physics. A fully comprehensive treatment, however, is currently in preparation.

# 4. Discussion

It is instructive to compare each of the contributions (12)–(14) to the overall rate of energy transfer. Determination of the comparative magnitudes represented by (13) and (14) suggests a priori knowledge of the individual components of the molecular tensors. For a particular system of interest this information may be sought through computational molecular modeling. To assess the viability of observing the effect, we shall assume the molecular tensor products  $\alpha^{(A)}\alpha^{(B)}$ ,  $\beta^{(B)}\mu^{(A)}$  and  $\beta^{(A)}\mu^{(B)}$  are similar in magnitude, their precise relative values being determined by the molecular electronic properties.

A conservative estimate of the laser intensities required for the ratios  $\Gamma^{(4-4)}/\Gamma^{(2-2)}$ and  $\Gamma^{(2-4)}/\Gamma^{(2-2)}$  to approach unity is achieved by assigning values of  $\mu^{(A)} \approx \mu^{(B)} \approx 1 \times 10^{-29}$  C m for the transition dipoles moments and estimating the tensor products equally as  $\alpha^{(A)}\alpha^{(B)} \approx \beta^{(A)}\mu^{(B)} \approx$  $\beta^{(B)}\mu^{(A)} \approx 1 \times 10^{-78} \text{ C}^4 \text{ m}^4 \text{ J}^{-2}$ . Assuming optimum conditions such that R is normal to both transition moments and the laser polarisation, calculations reveal that at  $I(k) > 10^{16}$  W m<sup>-2</sup> (1 × 10<sup>12</sup> W cm<sup>-2</sup>) the magnitudes of Eqs. (13) and (14) begin to approach that of Eq. (12). These intensities are readily available from conventional table-top Oswitched or mode-locked laser instrumentation [19] but are likely, however, to induce other nonlinear effects. To avoid such events when monitoring RET, much lower intensities are customarily employed.

Fig. 4 illustrates the relative magnitudes of the individual contributions with increasing laser intensity. Illustration of  $I(k) > 10^{16}$  W m<sup>-2</sup> shows the quadratic and linear nature of  $\Gamma^{(4-4)}$  and  $\Gamma^{(2-4)}$ , respectively. As we might expect, the rate of energy transfer at  $I(k) < 10^{16}$  W m<sup>-2</sup> is dominated by the I(k)-independent Förster-type result. Clearly, unfocused coherent light ( $I(k) \approx 10^7$  W m<sup>-2</sup>). does not perturb RET to any significant extent and the reduction to Förster's theory is valid. However, our estimated numbers have been chosen conservatively and the effects of resonance enhancement largely disregarded. The condition of exact resonance is of little interest since it affords the means for direct laser excitation of the acceptor. Nevertheless, if we consider frequencies in regions of anomalous dispersion where molecular polarisabilities increase by an order



Fig. 4. Graph representing individual contributions to the rate of energy transfer with increasing I(k). The contribution  $\Gamma^{(2-4)}$  is ~ 15% that of the Förster rate at  $I(k) \approx 10^{16}$  W m<sup>-2</sup>. The  $\Gamma^{(4-4)}$  term remains negligible until there is a five-fold increase in I(k).

of magnitude, then at irradiance values of  $\sim 10^{14}$  W m<sup>-2</sup> enhancement of the rate predominantly due to Eq. (14) can be as much as 15%. Translations of the

rate contributions to lower laser intensities under near-resonance conditions are illustrated in the loglog plot of Fig. 5.



Fig. 5. Log–log plot of the Förster rate (normalised to 100) and laser-dependent contributions. Off-resonance rates are depicted together with their resonance-enhanced counterparts. Because  $\Gamma^{(4-4)}$  is quadratically responsive both to resonance enhancement and to the laser intensity, whilst  $\Gamma^{(2-4)}$  varies linearly with each of these factors, the net effect of exploiting resonance is to shift the plots of both rate contributions by equal amounts along the abscissa scale.

# 5. Conclusions

We have calculated that application of an auxiliary laser field modifies the rate of non-radiative energy transfer between two close range species. The transfer rate is calculated to increase by only ~ 2%, at pulsed laser intensities of ~  $10^{15}$  W m<sup>-2</sup>. In consequence RET suffers little modification, whether enhancement or inhibition, at these intensity levels. This reinforces the legitimate use of Förster theory to describe most energy transfer processes. However, if the laser intensity is pulsed and highly focused ( $I(k) > 10^{16}$  W m<sup>-2</sup>) or the applied laser frequency is tuned close to an area of anomalous dispersion, thus enhancing the salient molecular parameters, then we conclude that energy migration can be enhanced to a significant and measurable degree.

It is possible to envisage several methods for the experimental realisation of LARET. Modulation of the auxiliary beam with lock-in detection of the RET signal (e.g. through the fluorescence or saturation of absorption by the acceptor) is one avenue. Another is to arrange for simultaneous irradiation of the sample with a femtosecond pump (to initiate the donor excitation) and a nanosecond probe with a variable delay. In the latter scenario, the measured fluorescence lifetime of the donor will be shortened by competition from an enhanced rate of excitation transfer.

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