Optically Switched Energy Transfer: Twin-Beam Off-Resonance Control

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It is shown that, in the optical near field, the interaction of two polarized laser beams with different frequencies can promote a transfer of energy between suitably placed donor and acceptor particles, through a process that is rigorously forbidden in the absence of the laser light. The mechanism, which operates through stimulated Raman scattering by the donor-acceptor pair, is identified and characterized by quantum electrodynamic calculation. The results suggest efficiency levels comparable to conventional resonance energy transfer. Optical switching applications are envisaged.

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Much of the current interest in the transfer of energy between atoms, or between nanoscale clusters, stems from technical innovations allowing such processes to occur in essentially static local environments—in contrast to primarily collision-induced transfer in atomic gases. Prominent examples of such systems are cold atoms [1–3], emitters in microcavities [4,5], and quantum dots [6,7]. Some of this interest is fuelled by recent advances in processes that additionally engage the transfer of spin

\[ \frac{\varepsilon_0}{\varepsilon_1} = \frac{\varepsilon_1}{\varepsilon_0} \]

overall energy conservation:

\[ E^B_\beta - E^B_\alpha = E^A_\alpha - E^A_0 + h(\omega_1 - \omega_2). \] (1)

Although the process is less amenable to semiclassical interpretation, it can be loosely described in such terms: The difference frequency of the two optical waves supplies a quantum of energy that, compensating for the mismatch between the donor decay and acceptor uptake in energy, enables a transfer of excitation to occur between the two particles.

The energetics of the process are depicted in Fig. 1. In principle, the optical coupling can result in the population of a level \( \beta \) in \( B \) either higher than level \( \alpha \) in \( A \) (as shown) or lower, according to the sign of \( (\omega_1 - \omega_2) \). Here attention first focuses on the case where the annihilation of photon from beam 1 engages with the decay of the donor atom/particle \( A \), stimulated emission of a beam 2 photon simultaneously engaging with excitation of the acceptor \( B \). It is taken that \( \omega_1 > \omega_2 \), the difference exceeding both excited state linewidths; the corresponding shift in energy
between the initial donor level and the excitation-acquiring acceptor level precludes energy transfer in the absence of the two beams. This energetic constraint is reinforced by the difference in optical selection rules, as two-photon conditions apply to both the donor decay and the acceptor excitation transition.

The transfer rate for the process is determined by the Fermi rule [18] \( \Gamma = (2\pi/\hbar)|M|^2\rho(E_\beta) \), where \( M \) is the quantum amplitude and \( \rho(E_\beta) \) the density of acceptor states at energy \( E_\beta \). The result is secured through textbook methods of QED [19], developing the quantum amplitude as a sum of 24 distinct fourth-order perturbation contributions:

\[
M = \frac{\hbar c(n_1 n_2 k_1 k_2)^{1/2}}{2\varepsilon_0 V}\left\{ e_1 \varepsilon_2 \alpha_{ij}^{0a}(k_1) V_{jk} (q + k_1, (R_B - R_A)) \alpha_{kl}^{0a}(k_2) e^{i(k_1 \cdot R_A - k_2 \cdot R_A)} \right. \\
+ e_1 \varepsilon_2 \alpha_{ij}^{0a}(k_1) V_{jk} (k_2 - q, (R_A - R_B)) \alpha_{kl}^{0a}(k_2) e^{i(k_1 \cdot R_A - k_2 \cdot R_A)} \right\]. \tag{2}
\]

Here \( e_m \) designates the \( i \)th component of the polarization vector \( e_m \) (an overbar denoting complex conjugation, for circular polarizations), \( q \equiv (E_\alpha - E_\beta)/\hbar c \), the scalar \( V \) is the quantization volume, and the coupling tensor [15] (representing the retarded interaction of transition dipoles) is given by

\[
V_{ij}(p, R) = \frac{e^{ipR}}{4\pi\varepsilon_0 R^3} \left\{ (1 - ipR)(\delta_{ij} - 3\hat{R}_i \hat{R}_j) \\
- p^2 R^2 (\delta_{ij} - \hat{R}_i \hat{R}_j) \right\}. \tag{3}
\]

In the present context, the coupling can accurately be approximated by its near-field asymptote \( V_{ij}(R) = (4\pi\varepsilon_0 R^3)^{-1}(\delta_{ij} - 3\hat{R}_i \hat{R}_j) \), and, for application in this region, the phase factors in Eq. (2) can also be dropped.

\[
\begin{align*}
|A| & \quad \hbar\omega_1 & \quad |E_0| \\
& \quad \Delta E_A & \quad \Delta E_B \quad \hbar\omega_2 \\
\quad h\omega_1 & \quad E_\alpha \\
\quad E_\beta & \quad \Delta E_A \\
\quad E_A & \quad \hbar\omega_2 & \quad E_B \\
\end{align*}
\]

FIG. 1. Energetics scheme for optically switched transfer of energy from donor A to acceptor B. Solid-head arrows denote four transitions coupling the donor decay \( \alpha \to 0 \) and acceptor excitation \( 0 \to \beta \), laser interactions denoted by the photon energies. Dotted lines denote virtual states, the closest real states \( E_A \) and \( E_B \) offset in energy by \( \Delta E_A \) and \( \Delta E_B \). The dashed line signifies energy transfer.
\[ \Gamma = \frac{\mu_1 I_2}{32 \pi h c^2 e_0 R^6} |e_{1i} e_{2j}|^2 \alpha_{ij}^{(A,B)}(k_1) (\delta_{ij} - 3 \hat{R}_i \hat{R}_j) \times \alpha_{ij}^{(B)}(k_2)^2. \] (6)

Here \( \hat{R} \) is the displacement vector \((\hat{R}_B - \hat{R}_A)\), and the result is cast in terms of the irradiances \( I_1 \) and \( I_2 \) of the two beams. The sharp \( R^{-6} \) dependence of the rate is a motif commonly associated with short-range RET; in both cases, it signifies that near-field energy transfer occurs primarily between closely neighboring particles, a feature that allows exploitation without significant cross talk in array implementations [22]. In the present case, it is also evident that energy transfer can take place only if both beams are present, signifying a simple logic operation.

To exactly determine the efficiency of the process for a specific system requires knowledge of the generalized polarizability components, and these are not available in the current literature. However, an assessment can be based on observing that the result is not dissimilar in form from the standard result for short-range resonance energy transfer, between particles with energetically matched excited states, due to transition dipole coupling [14]. Normal RET and the optically induced process cannot both occur in the same particle pair, because they have different energetics—but the relative magnitude of the corresponding transition dipole coupling [14] will here occur at the same site (either the donor or the acceptor) such that three-photon selection rules are associated with the corresponding transition. One other intriguing possibility is the engagement of spin transfer in optically activated transfer between quantum dots, where circular beam polarizations can be expected to play a significant determining role (see, for example, Ref. [10]). A full analysis of these cases is currently underway.

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