Vibrationally assisted quantum energy pumps

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Abstract

We show that directed energy transport in a linear array of coupled quantum dots can be achieved by a coherent coupling of each dot to a single coherently driven mechanical mode. Recent work on light harvesting molecules have implicated the role of discrete mechanical modes in enhancing the energy transport through dipole arrays but say less about directed transport. The study of quantum ratchets indicates how directed energy transport is possible in quantum dot arrays. Inspired by these two apparently unrelated models we show how directed energy transport may be implemented in an engineered quantum systems using a single mechanical degree of freedom. This may have implications for nano-engineered artificial energy harvesting systems.

Introduction

The study of quantum energy transport is attracting significant interest particularly in the debate surrounding the role of quantum phenomena in photosynthesis where a Frekel exciton created in a pigment-protein complex (PPC) is transported in picoseconds to a reaction centre with near unit efficiency. Enhancing nanoscopic energy transport is also attracting significant attention in man-made energy harvesting systems and nanodevices, such as in solar cells [1–3], and quantum dot arrays [4]. Recent experimental 2D pump–probe femto-second spectroscopy in biological systems has provided evidence for long lived oscillations which points to a potential description of the transport via a delocalized quantum wave function over the PPC [5]. Theoretical quantum models for the observed near unit efficiency transport have mostly focused on enhancing the transport via the inclusion of environmental noise on the exciton sites [6, 7]. Although such models enhance the exciton transport they provide no directionality of the transport towards a reaction site. The study of quantum ratchets examines how one can achieve directional probability currents in time dependent systems even in the absence of a global bias force [8]. We bring these two disparate fields together to design a coherent quantum energy pump that produces enhanced energy transport in a unidirectional fashion with particular applications man-made synthetic nanosystems.

Achieving directed enhanced quantum transport, or energy pumping, towards the reaction centre irrespective of the exciton’s initial location permits far superior collection of the deposited energy. Achieving directed currents in coherent quantum systems is itself a topic of intense study and recent work on edge currents in topological insulators [9], and in parametrically modulated atomic ratchets [10], illustrate two systems exhibiting directed quantum currents. Models focusing solely on the exciton degrees of freedom in light-harvesting systems may not be the complete explanation. Recent vibrational spectroscopy of light-harvesting complexes like the Fenna–Matthew–Olson (FMO) complex have clearly displayed discrete intramolecular nuclear vibrational modes and these are thought to play an important role in the enhanced exciton transport. While recent experiments indicate an important role for vibrations in exciton transport [11–13], most theoretical investigations consider the effects of weakly coupled vibrational dynamics through the environment for the exciton dynamics [14–22]. Others consider the case when a single vibrational mode is globally coupled to the excitons in an exciton–vibronic Hamiltonian [23–26]. However, investigation into directed charge transport...
such as the recent models of [22, 27], make the assumption of an overall 'downhill' spatial bias in the molecular energies or require noise to achieve transport directionality. Our scheme for a quantum energy pump requires no overall spatial bias but rather we use a spatially periodic molecular energy profile—a profile naturally present in many types of nano-systems, including FMO.

We are guided by the experimental observation that spectroscopic evidence has indicated that discrete intramolecular nuclear vibrational modes are experimentally observed above a background of more weakly coupled vibrational dynamics. We therefore consider the case where such discrete vibrational modes are strongly coupled to the PPC exciton chain. Weaker coupled vibrational modes give rise to vibrational dephasing and decoherence, which can also assist in increased transport [6], but we instead focus on whether the discrete strongly coupled vibrational modes can give rise to enhanced directed transport.

We consider a system where the modelled PPC exciton chain is collectively coupled to a vibrational (phonon) mode. The individual exciton site energies vary periodically along the chain and their energy is collectively modulated in time by the phonon via this collective coupling. The coupled exciton–vibrational system behaves as a type of linear motor: the deposition of an exciton on the chain excites the phonon mode to oscillate. These oscillations couple to modulate the entire energy landscape of the PPC chain to effect directed and enhanced quantum pumping of the exciton along the chain. The system requires no overall spatial bias nor external time-dependent driving to operate and the initial energy for the one-way engine comes from the deposition of the initial exciton.

In what follows we first examine the full autonomous excitonic–motional coupled system and find clear numerical evidence for directed enhanced transport. We then study a semiclassical approximate model where the motional dynamics is replaced by coherent state dynamics, replacing the oscillator’s position by the coherent state mean values, which modulate the PPC chain’s electronic potentials. We find excellent agreement between analytic predictions from this semiclassical modulated model and the fully coupled autonomous excitonic–motional system.

Model

We consider the transport of a single excitation along a linear chain of $N$ two level sites that have reversible homogenous nearest neighbour coupling:

$$\hat{H}_2 = \frac{1}{2} \sum_{j=1}^{N} \omega_j \hat{a}^\dagger_j \hat{a}_j + \lambda \sum_{j=1}^{N-1} \left( \hat{a}^\dagger_{j} \hat{a}_{j+1}^\dagger + \hat{a}_{j+1} \hat{a}_j \right),$$  \hspace{1cm} (1)

where $\omega_j$ is the on-site energy for the $j$th site, $\lambda$ is the tunnelling rate between neighbouring sites, which we consider to be uniform along the chain, with $\hat{a} \equiv |e\rangle\langle e| - |g\rangle\langle g|$, $\hat{a}^\dagger_j = |e\rangle\langle g|/\sqrt{\nu}$. The initial state of the exciton chain we take to be $|g_0, g_{N-1}, \ldots, g_{1}, e_0, e_{N-1}, \ldots, e_{N}\rangle$, and we will later specify that the injection site is midway along the chain i.e. $j \sim N/2$. To consider the exciton–vibrational interaction we couple a single vibrational mode to each of the two-level systems with coupling strength $g_0$. This global coupling of a single mode to the exciton chain yields spatially correlated excitonic fluctuations along the chain [28–30]. This is similar to other vibrationally enhanced transport schemes, such as [24, 31], but in our case we do not consider any external driving of the common vibrational mode. Instead we will show that the initial excitation of the $j$th site will excite the coupled vibrational mode and the resulting exciton–vibrational dynamics induces directed exciton transport. We consider the on-site exciton energy to be modulated by the position of the mechanical vibrational mode e.g. $\delta = q_0(\hat{a} + \hat{a}^\dagger)$, where $q_0$ is a convenient scale for mechanical displacement, for example the standard deviation in position in the ground state of the oscillator. The Hamiltonian for the coupled quantum excitonic–vibrational system is

$$\hat{H} = \frac{1}{2} \sum_{j=1}^{N} \omega_j \hat{a}^\dagger_j \hat{a}_j + \lambda \sum_{j=1}^{N-1} \left( \hat{a}^\dagger_{j} \hat{a}_{j+1}^\dagger + \hat{a}_{j+1} \hat{a}_j \right) + \nu \hat{a} \hat{a}^\dagger + q_0(\hat{a} + \hat{a}^\dagger) \hat{\Sigma}_2,$$  \hspace{1cm} (2)

where $\hat{\Sigma}_2 = \sum_{j=0}^{N} g_j \hat{a}^\dagger_j$ and $\nu$ is the mechanical vibrational frequency. In general we assume the collective vibrational mode is damped to a thermal environment with $\hbar$ mean thermal occupancy at a damping rate of $\gamma_\nu$:

$$\frac{d\rho}{dt} = -i[\hat{H}, \rho] + \gamma_\nu (\hat{a}^\dagger + 1) \hat{D}[\hat{a}] \rho + \gamma_\nu \hat{D}[\hat{a}] \rho$$

where $\hat{D}[\hat{A}] \rho = \hat{A} \hat{\rho} \hat{A}^\dagger - \frac{1}{2} \hat{A}^\dagger \hat{A} \hat{\rho} - \frac{1}{2} \hat{\rho} \hat{A}^\dagger \hat{A}$.

For our analyses here we will ignore thermal damping ($\gamma_\nu \rightarrow 0$), and consider only Hamiltonian evolution from equation (2).

To gauge the directionality of transport along the exciton chain we include irreversibly coupled sinks off site 1 and site $N$ by adding the non-Hermitian terms $-i\epsilon |e\rangle\langle e| - i\epsilon |e\rangle\langle e|$ to the Hamiltonian in equation (2):

$$\hat{H} \rightarrow \hat{H} - i\epsilon |e\rangle\langle e|$  \hspace{1cm} (3)

These non-Hermitian terms allow one to calculate the conditioned state of the entire system $|\Psi(t',t')\rangle$ for each sink site to absorb the excitation in the time interval
[t', t' + dt] \quad \text{d} P_k = 2\kappa \langle \psi_k(t') | \hat{\sigma}_k^+ \hat{\sigma}_k^- | \psi_k(t') \rangle \text{d} t^1, \quad \text{with} \quad \{ j \} = \{ 1, N \}, \quad \text{and calculate the efficiency of transport to a particular sink } \mathcal{E}_k(t), \quad \text{by summing up each of these incremental probabilities from time 0 to time } t:

\mathcal{E}_k(t) \equiv \int_0^t 2\kappa \text{ Tr} \{ \rho_k(t') \hat{\sigma}_k^+ \hat{\sigma}_k^- \} \text{d} t^1 = \int_0^t 2\kappa \langle \psi_k(t') | \hat{\sigma}_k^+ \hat{\sigma}_k^- | \psi_k(t') \rangle \text{d} t^1. \quad \text{Directed transport is evident when } \mathcal{E}_k(t) - \mathcal{E}_k(0) \text{ is appreciable over long durations.}

**Vibrationally actuated directed motion**

We now focus on a particular class of on-site exciton energies \( \omega_j \) inspired by the sawtooth-like on-site energies experimentally observed in the FMO complex [33]. Guided by the literature on Hamiltonian rocking ratchets [10, 34–37], we choose on-site exciton energies and vibrational couplings as shown in figure 1. It is known that by temporally modulating ('rocking'), a locally asymmetric spatially periodic potential one can generate directed currents. In all previous studies this rocking has been implemented via external driving. In our pumping engine the role of rocking will be supplied via coherent coupling to the excited vibrational dynamics.

We study an \( N = 7 \) site system and assume the vibrational mode is initially in the ground state \( | 0 \rangle \). To obtain a fair assessment of the directed nature of the excitation transport from the sinks at either end of the chain we assume the middle site is initially excited, yielding \( | \psi_0 \rangle = | g, g, g, g, g, g, g \rangle \otimes | 0 \rangle \).

We find that a sawtooth pattern for the on-site energies \( \omega_j \) and a linear choice for \( g_j \) produces directed transport for a range of mechanical frequencies \( \nu \), shown in figure 2. To uncover the rocking-ratchet mechanism giving this directed enhanced transport we focus our attention to the case when the vibrational frequency \( \nu \approx 1.75 \) and examine the mean vibrational quantum dynamics. In figure 3 we study the fully coupled dynamics of the system (2), and in particular the dynamics of the average position and momentum of the mechanical oscillator. The latter exhibits a decaying orbit in the stable manifold of an attracting fixed point displaced to the left of the origin. This decay corresponds to an effective transfer of energy from the mechanical system to the electronic system during coherent evolution. We therefore consider the dynamics that results from the Hamiltonian with \( \lambda = 0 \) with the oscillator initially in the ground state and a single excitation at the centre of the electronic chain

\[ \hat{H}_{\nu=0} = \sum_{j=1}^{N} \frac{\omega_j}{2} \hat{\sigma}_j^+ \hat{\sigma}_j^- + \nu \hat{a}^\dagger \hat{a} + q_0 (\hat{a} + \hat{a}^\dagger) \Sigma_c. \]
From this we can obtain
\[ |\psi_0\rangle \rightarrow e^{i\nu_0(t)} |\alpha(t)\rangle, \]
where \( \alpha(t) = q_0(e^{-i\nu t} - 1)\gamma_{ij}/\nu, x = i\rho = q_0\alpha, \) is the phase space mean location (complex) of the coherent state. \( \gamma_{ij} = g_j - g_0 - \cdots - g_j \) and we have used the identity: \( e^{\hat{a}^\dagger + \hat{a} + \hat{q}^\dagger \hat{q}} = e^{\frac{e^{C-1}}{2} \hat{e}^{\dagger}(1-e^{-C}) \hat{d} e^{\hat{C}}} \). The orbits of \( \alpha(t) \) are non-decaying circles centred on \( z_a = (x_a, p_\nu) \), in phase space and \( z_a = (-\gamma_{ij}/\nu, 0) \). This approximation establishes a centre for the phase space dynamics of the oscillator and is in good agreement with the stable attracting point of the full dynamics, which, for the system parameters used in figure 3, is \( z_a \approx (-1.43, 0) \). Considering now \( \lambda = 0 \), an initially localized electronic excitation starts to delocalize and quantum entanglement between different exciton locations and the oscillator mode builds. From the oscillator viewpoint the delocalized excitonic dynamics acts as quantum noise and the mean motional dynamics is drawn to a stable attractor in phase space, the point \( z_a \).

**Semiclassical model**

In order to understand better how particular values of \( (\nu, g_j, \omega) \), lead to enhanced directed transport we consider a semiclassical model where we assume the oscillator remains in a coherent state which undergoes displaced circular dynamics in phase space. To do this we replace \( \hat{x} = q_0(\hat{a}^\dagger + \gamma_j) \), by the mean value of the coherent state \( x(t) = q_0(\alpha(t) + \alpha^\dagger(t)) = q_0(\cos(\nu t) - \beta) \), obtaining the time-dependent semiclassical Hamiltonian for the electronic system alone as

\[ H_{\text{classical}} = \frac{1}{2} \hat{\rho}^2 + \frac{1}{2} \gamma_{ij} \delta_{ij} + \frac{1}{2} \gamma_{ij} \hat{q}_j \hat{q}_i. \]
there are two possible cases to achieve resonance in 

\[ \Delta \omega_j = \omega_j + 2g_j\gamma \cos(\nu t) - \beta, \]

represents the effective modulation from a rotation of the mechanics in a circle in phase space with amplitude \( \gamma \) centred on the \( x \)-axis a distance \( -\beta \) from the origin. We now consider a convenient interaction picture to find resonance conditions that correspond to enhanced directed transport. In a similar fashion to [24], we consider the interaction picture generated by the unitary

\[ U = \exp \left( -\frac{i}{\hbar} \sum_{j=0}^{N} \int_0^t dt' \omega_j(t') \hat{\sigma}_j \right), \]

This yields the interaction Hamiltonian:

\[ \hat{H}_I = \lambda \sum_{j=1}^N (e^{i\theta_j(t)\hat{\sigma}_j^x} \hat{\sigma}_j^+ \hat{\sigma}_j^0 + \text{h.c.}), \]

where \( \theta_j(t) = -\left( \Delta \omega_j t - 2\Delta g_j\gamma/\nu - 2i\Delta g_j\gamma \sin(\nu t)/\nu \right), \) and \( \Delta \omega_j = \omega_j - \omega_{j+1} \) and \( \Delta g_j = g_j - g_{j+1}. \) Using the identity

\[ e^{i\phi} \sin(\phi) = \sum_{n=-\infty}^{\infty} J_n(\phi) e^{i\phi}, \]

we rewrite this interaction Hamiltonian as

\[ \hat{H}_I = \lambda \sum_{j=1}^\infty \left( J_0(\chi_j) e^{-i(\phi_j - \pi n)} \hat{\sigma}_j^+ \hat{\sigma}_j^0 + \text{h.c.} \right), \]

where \( \chi_j = -2\Delta g_j\gamma/\nu \) and \( \phi_j = \Delta \omega_j - 2\Delta g_j\gamma \beta. \) One now observes secular behaviour and enhanced transport at the resonance conditions

\[ \phi_j = \Delta \omega_j - 2\Delta g_j\gamma \beta = n\nu, \quad n = 0, \pm 1, \pm 2, \ldots. \]

For a particular choice of \((\nu, \gamma, \omega_j)\) this resonance condition will give corresponding values of \(\beta\) that lead to enhanced directed transport. Taking the parameter values for \((g_j, \omega_j)\) as in figure 2 then \(\Delta g_j = 1/3\) and \(\Delta \omega_j = -1\) or \(1/2.\) Since we have two possible values of \(\Delta \omega_j\), there are two possible cases to achieve resonance in (6), and their associated values of \(\beta.\) By considering \(\nu \approx 1.75\) one can find values of \(\beta,\) which are encompassed within the initial displaced circular motional dynamics due to the deposition of the exciton on the chain. These resonances are summarized in table 1. We now numerically check for enhanced directed transport at the predicted resonances with \(\nu \approx 1.75,\) by numerically scanning over \((\beta, \gamma)\) values in the Hamiltonian (4), computing the resulting efficiencies to find the results shown in figure 4. There is excellent agreement between the predicted values of \(\beta\) in table 1, and the peaks found in figure 4(b). We also find excellent agreement for our predictions of the values of \(\gamma\) which lead to minima in the transport, as observed in figure 4. For example, when \(\beta \approx 1.33\) we have resonances when \(n = 0\) \((\Delta \omega_j = 1/2),\) and \(n = 1\) \((\Delta \omega_j = -1)\) in equation (6). For vanishing transport we must have \(J_0(\chi_j) \sim J_1(\chi_j) \sim 0\), simultaneously and using \(\Delta g_j = 1/\nu, \nu = 1.7575, \gamma = 1/\sqrt{2},\) we find the transport to vanish when \(\gamma = 0, \pm 8.97, \pm 14.29, \ldots,\) in excellent agreement with the numerical results shown in figures 4(a) and (b). Applying the same analysis for each of the average \(\beta\) values in table 1, we find very good correspondence with the predicted values of \(\gamma\) and figure 4 for vanishing transport in table 2.

We now show that the values of \(\beta\) in table 1 which lead to enhanced directed transport in the semiclassical model (4), are closely related to the initial motional dynamics in the full exciton–vibration model (2). In the latter, the initial mean motional dynamics results in displaced circular paths in phase space about the focal point \(z_0 = (-\delta_0/\nu, 0)\) which also intersect the origin which corresponds to \(\gamma \sim \beta.\) Comparing (2) and (4), we find the relation \(\beta = \delta_0\nu \approx 2.02,\) using the focus \(z_0 = (-1.42, 0).\) From the first row in table 1 we observe that since the initial motional dynamics encompasses the resonant \(\beta\)-focus we expect enhanced directed transport. To further explore this in figure 5 we plot the efficiency of transport at site 1 and 7 in the semiclassical model when \(\beta = 1.33.\) Notice that this plot indicates that the maximum attainable efficiency of transport to site 7 should be approximately 0.86, but this occurs for \(\gamma \sim 4,\) which could only be achieved if the initial motional

| \(\Delta \omega_j\) | \(n\) | \(\beta\) | Average \(\beta\) |
|-----------------|-----|-----|----------------|
| \(1/2\)         | 0   | 1.06| 1.33           |
| \(-1\)          | 1   | 1.61|               |
| \(-1\)          | 1   | 2.67| 2.39           |
| \(-1\)          | 0   | 2.12|               |
| \(-1\)          | 1   | 4.79| 5.06           |
| \(-1\)          | 2   | 5.34|               |
| \(-1\)          | 2   | 6.40| 6.12           |
| \(-1\)          | 1   | 5.85|               |
state is not the vacuum state. When the initial motional state is the vacuum and \( \gamma \sim \beta \), figure 5 yields 0.6, in very good agreement with figure 2. The \( \gamma \) parameter in figure 5 represents the amplitude of the oscillation of the mechanics in phase space in the semiclassical model (4). This freedom is not easily present in the full

**Table 2.** The corresponding values for \( \gamma \) that lead to zeroes of transport in figure 4.

| \( \Delta \omega_j \) | Average \( \beta \) | \( \beta \), that must \( \beta = 0 \) | Corresponding \( \gamma \) values |
|------------------|----------------|------------------|-------------------------------|
| 0.237            | 1.33           | \( J_0, J_1 \)   | 0, ±8.97, ±14.29, ...         |
| 0.237            | –2.39          | \( J_{-1}, J_0 \) | 0, ±8.97, ±14.29, ...         |
| 0.237            | 5.06           | \( J_0, J_2 \)   | 0, ±14.29 ± 19.15, ...        |
| 0.237            | –6.12          | \( J_{-2}, J_{-1} \) | 0, ±14.29, ±19.15, ... |

**Figure 5.** The efficiency of transport at sites 1 and 7, \( \mathcal{E}_{1,7} \), as a function oscillation amplitude \( \gamma \) when \( \beta = 1.33 \) and \( \nu = 1.75 \) for the modulated time dependent excitonic system (4).

![Figure 5](image1.jpg)

![Figure 4](image2.jpg)

![Figure 4](image3.jpg)

![Figure 4](image4.jpg)

![Figure 4](image5.jpg)
quantum coupled exciton–mechanical model (2), and this amplitude can only be altered by either depositing the excitation at a different locations on the chain or by considering alternate initial states for the mechanics. For a physical implementation we examine the ratios: $g^* = \max_j |g_j| \sqrt{\lambda}$, $\omega^* = \max_j |\omega_j| \sqrt{\lambda}$ and $\nu^* = \max_j |g_j| \sqrt{\nu}$. Referring to figures 1 and 2 we require (a) $g^* \sim 5$, (b) $\omega^* \sim 10$, and (c) $\nu^* \sim 0.5$. Stationary chains of quantum dots can be easily engineered to achieve condition (a) (figure 1(a) in [38]), while (c) values of $\nu^* \sim 1$ has been demonstrated in a hybrid quantum dot–cantilever system [39]. Thus, implementing our design in engineered quantum motional system using quantum dots is feasible.

We have designed a type of quantum machine—or quantum energy pump—that utilizes both excitonic and vibrational motions to pump energy along an excitonic chain. The pump is initially in its ground state and the deposition of the initial charge on the exciton chain excites the vibrational motion, i.e. provides the fuel for the pump bringing it out of equilibrium. The resulting coherent mechanical dynamics causes directed enhanced energy transport towards one end of the exciton chain. The efficacy of the pump decreases as the exciton delocalizes along the chain. Interesting questions for future work include the effects of decoherence on the efficacy of the pump, the dependence on various alternative choices for the initial state, whether anharmonic vibrational dynamics could lead to similar directed enhanced transport and whether other types of autonomous quantum machines can be devised to route energy and quantum coherence in an improved fashion using alternative exciton geometries e.g. ring like structures.

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