

Quantum Simulation of Photoinduced Excitation Dynamics Relevant to Photovoltaic Systems Using Open Quantum Models

Adarsh Devakottai Raghavan¹

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This study asks whether a simplified driven two level model can quantitatively reproduce coherent excitation dynamics of a localized photoactive transition, evaluated through agreement with analytical Rabi oscillations with an error threshold below 1%. A Python simulation using QuTip was developed to reproduce photoinduced excitation dynamics. This was then used to validate the characteristic Rabi oscillations against the analytical solution, demonstrating excellent agreement with a relative error of 0.0174%. To provide a more realistic representation of photovoltaic materials, the model was extended to an open quantum system using the Lindblad master equation. The equation includes decoherence and relaxation effects. Simulations show that an isolated system exhibits sustained coherent oscillations. However, environmental interactions dampen these oscillations and thus reduce the excited state population. A sensitivity analysis quantifies the impact of increasing decoherence on excitation efficiency. The model demonstrates that coherent Rabi oscillations alone do not generate a net photovoltaic current because they represent reversible population exchange rather than irreversible carrier extraction. These results represent the importance of open quantum system modeling for studying quantum excitation dynamics.

Introduction

Solar energy is an integral part of the global transition toward sustainable energy^{1,2}. Photovoltaic devices turn solar radiation into electrical energy by generating and separating charge carriers after optical excitation in semiconductor materials. But, their efficiency is limited, with current photovoltaic devices typically achieving efficiencies around 20–25% for commercial modules and advanced laboratory cells exceeding 30%.

One constraint is the Shockley—Queisser limit, which arises from loss mechanisms such as thermalization and incomplete photo-induced excitation³. Photo-induced excitation in semiconductors is fundamentally a quantum mechanical process^{1,4}. A useful framework for analyzing such transitions is the two level quantum system^{5,6}. Here, electrons can be modelled as occupying either a ground state or an excited state. It is a simple representation, but this model captures the essential physics of semiclassical light matter interaction, such as absorption and emission. Recent advances in quantum optics have been able to demonstrate precise control of light matter interactions of individual quantum transitions^{7–9}.

Quantum descriptions of photovoltaic processes have developed into microscopic descriptions of excitons, and quantum confined systems. In semiconductor structures, optical excitation has been demonstrated through Rabi oscillations of excitons in quantum dots, showing that two level approximations

are able to accurately describe localized transitions under controlled conditions^{8,9}.

Quantum models of photovoltaic systems have been developed to investigate microscopic processes such as exciton formation and charge separations. In quantum dot solar cells, electronic states modify optical absorption and enable the study of confined transitions, providing a system where simplified quantum models can describe localized excitation processes. Realistic photovoltaic materials, though, contain many interacting states, continuous electronic bands, phonon interactions, and carrier transport mechanisms that require significantly more complex approaches. Therefore, simplified two level models remain useful as computational frameworks for isolating fundamental light–matter interactions, provided that their limitations are clearly distinguished from complete photovoltaic device behavior.^{8–10}

In biological light harvesting systems, quantum coherence has also been investigated as a mechanism influencing energy transfer efficiency, although the persistence and functional role of coherence remain debated^{11–14}. Open quantum system approaches based on density matrices and Lindblad dynamics have become widely used to describe the influence of environmental interactions on nanoscale energy transfer^{5,15–17}.

These models provide detailed descriptions of semiconductor processes but often require large many-body simulations. Simplified two level models provide computational accessibility by isolating fundamental optical transitions but cannot reproduce band continua, carrier transport, or device-level be-

¹ Sherwood High School, India

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Existing microscopic quantum models often require computationally intensive many-body calculations. A remaining challenge is developing computationally accessible models that isolate fundamental coherent excitation dynamics while clearly distinguishing them from complete photovoltaic device behaviour.

This paper investigates the behavior of a two level quantum system under excitation as a simplified model for photovoltaic processes. Through the application of principles from quantum mechanics and semiconductor physics, this work aims to evaluate how controlled semiclassical light matter interactions can influence energy conversion efficiency and provide insight into pathways beyond conventional limits^{14,18}.

While two level quantum models have been widely used to investigate light-matter interactions, they often neglect environmental effects which play a significant role in real photovoltaic materials¹⁸. In this study, a validated two level quantum model is extended using the Lindblad master equation to investigate the influence of decoherence and relaxation on photo induced excitation dynamics¹⁹. The resulting simulations provide a framework for examining the transition from ideal coherent dynamics to more realistic open system behavior relevant to photovoltaic materials^{20–22}. The present two level model does not reproduce the Shockley–Queisser absorption threshold because it describes a discrete transition rather than a semiconductor band structure. The model instead isolates the microscopic excitation dynamics occurring after resonance conditions are satisfied.

Previous studies have investigated quantum effects in photovoltaic and light harvesting systems. Engel et al. demonstrated signatures of quantum coherence in photosynthesis complexes, while semiconductor quantum optics studies have explored coherent excitation dynamics in confined systems. However, these microscopic quantum descriptions are hard to translate into computational models. This work addresses this gap by developing and validating a simplified open quantum system model that isolates coherent excitation dynamics before incorporating environmental effects^{11–14}.

Background

Basic Terminology

A photon is a quantum of electromagnetic energy, and its energy is proportional to its frequency. An electron hole pair is a pair formed when a photon excites an electron. It thus promotes it from a lower energy state to a higher energy state and leaves behind a hole in the valence band. A two level system ($|0\rangle, |1\rangle$) is a simplified quantum model containing only a ground state and an excited state which is used to represent electron transitions. A Dipole approximation assumes that the

electromagnetic field across an electron is uniform so that the interaction only depends on the dipole moment⁸. The dynamics of a driven two level system are governed by the Hamiltonian:

$$H = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}\sigma_x,$$

where Δ is the detuning between the photon frequency and the transition frequency, Ω is the Rabi frequency measuring the strength of coupling between the two level system and an oscillating electromagnetic field, and σ_x and σ_z are Pauli matrices^{7,10,23}.

Rotating Wave Approximation and Resonant Excitation

The interaction between a two level quantum system and an oscillating electromagnetic field is described using the rotating wave approximation (RWA). Here, rapidly oscillating terms in the interaction Hamiltonian are neglected as their average contribution over time is negligible when the driving frequency is close to the system's transition frequency. This simplifies the mathematical description while still capturing the dynamics of semiclassical light matter interactions near resonance⁷.

In this study, the resonant condition $\Delta = 0$ was adopted, where the frequency of the incident photon matches the transition frequency of the system. Under resonance, energy transfer between the electromagnetic field and the quantum system is maximized. Although non-zero detuning can modify the oscillation frequency and amplitude, investigating off-resonant excitation is beyond the scope of the present work and is left for future study^{5,6}.

The rotating wave approximation is valid when the coupling strength is much smaller than the transition frequency $\frac{\Omega}{\omega_0} \ll 1$. For typical optical transitions, $\frac{\Omega}{\omega_0}$ is approximately 10^{-6} – 10^{-3} , satisfying the weak coupling condition required for the rotating wave approximation. Therefore, $\frac{\Omega}{\omega_0}$ remains sufficiently small and the approximation is justified^{8,24,25}.

Rabi Oscillations

Under resonant excitation, the probability of finding the electron in the excited state oscillates periodically. These oscillations, known as Rabi oscillations, arise from the coherent exchange of energy between the electromagnetic field and the quantum system. In an ideal isolated system, the excited state probability is given by:

$$P_e(t) = \sin^2\left(\frac{\Omega t}{2}\right),$$

Where Ω is the Rabi frequency. This analytical solution provides an important benchmark for validating numerical

quantum simulations^{8–10}. In this work, Ω denotes the conventional Rabi frequency such that the excited-state probability follows $P_1(t) = \sin^2(\Omega t)$. The Hamiltonian coupling coefficient is therefore $\Omega/2$.

Open Quantum Systems

Real photovoltaic materials are not perfectly isolated. Electrons interact with their environment through the lattice vibrations, and other microscopic processes. These interactions cause decoherence⁴, reducing the coherence of the quantum state. Relaxations, on the other hand, allow excited electrons to return to the ground state.

To account for these effects, the system is described using the Lindblad master equation,

$$-\frac{i}{\hbar}[H, \rho] + \left(\sum_k L_k \rho L_k^\dagger - \frac{1}{2} L_k^\dagger L_k \rho \right)$$

where ρ is the density matrix^{2,3} describing the quantum state and L_k are collapse operators representing environmental interactions. This enables realistic simulations of decoherence and relaxation while being able to preserve the probabilistic interpretation of quantum mechanics^{20–22}.

Quantum Simulation for Photovoltaic Systems

Quantum simulation can provide a framework for studying excitation dynamics that are difficult to observe directly. Modeling coherent excitation together with environmental interactions can reveal how quantum coherence influences energy absorption and transport within photovoltaic materials. Though it is simplified to a two level model, this approach captures the fundamental dynamics of a simplified photoinduced excitation process. It also provides a foundation for more advanced quantum simulations of realistic solar energy materials.

Methods

Simulation Framework

The quantum dynamics of photoinduced excitation were simulated using Python and the Quantum Toolbox in Python (QuTiP)⁶. QuTiP was selected because it provides efficient numerical solvers for both closed and open quantum systems, allowing the simulation of coherent quantum evolution as well as environmental interactions through the Lindblad master equation^{6,16,17,20}. Unlike statevector only approaches, QuTiP allows density matrix simulations required for Lindblad decoherence modelling. All simulations were performed within a Jupyter Notebook environment using a double precision numerical integration.

Two Level Quantum Model

The localized photoactive transition relevant to photovoltaic materials was represented as a two level quantum system consisting of a ground state $|0\rangle$ and an excited state $|1\rangle$. The ground state corresponds to an electron before photo-induced excitation while the excited state represents the electron after excitation by an electromagnetic field^{8–10}. The model does not represent a complete version of valence and conduction bands of a bulk semiconductor. Quantum dots instead provide a physically justified two level approximation because confinement produces well separated energy states. This in turn allows optical manipulation of individual transitions^{8,9}. The model therefore represents a fundamental excitation mechanism rather than the complete operation of a photovoltaic device.

In this study, the two level system represents a localized excitonic transition in a semiconductor quantum dot rather than a bulk photovoltaic absorber. Typical semiconductor quantum dots exhibit optical transition energies in the range of around 1–2 eV, which correspond to visible and near-infrared photon energies, with transition dipole moments on the order of tens of Debye²⁶. Under these conditions, coupling to a resonant electromagnetic field can be approximated by a two-state interaction while neglecting higher-energy states that are not significantly populated during the excitation process.

The system Hamiltonian was defined as

$$H = \frac{\Delta}{2} \sigma_z + \frac{\Omega}{2} \sigma_x,$$

where Δ is the detuning between the photon frequency and the transition frequency, Ω is the Rabi frequency measuring the strength of coupling between the two level system and an oscillating electromagnetic field, and σ_x and σ_z are Pauli matrices. Resonant excitation was modeled by setting the detuning parameter to zero. The factor of 1/2 in the Hamiltonian ensures consistency between the Hamiltonian coupling term and the analytical Rabi oscillation frequency.

The electron was initialized in the ground state. The time evolution of the system was computed over a uniform time interval.

Validity of the Closed Quantum System

The simulated excited state probability was compared with the analytical solution for Rabi oscillations to verify the accuracy of the numerical implementation:

$$P_e(t) = \sin^2\left(\frac{\Omega t}{2}\right),$$

Model accuracy was calculated using the Root Mean Square Error (RMSE), Mean Absolute Error (MAE), maximum absolute error, and the relative difference between the theoretical

and simulated Rabi frequencies. The agreement between the numerical and analytical solutions demonstrated that the implementation reproduced the expected dynamics of the driven two level model before introducing environmental effects.

Open Quantum System Modelling

To represent realistic photovoltaic environments, the simulation was extended to an open quantum system using the Lindblad master equation. The density matrix formalism was used to account for interactions between the quantum system and its surroundings.

Two environmental processes were included: decoherence, which represented the loss of quantum coherence caused by interactions with the surrounding environment, and relaxation, which represented the spontaneous decay of excited electrons back to the ground state.

These effects were implemented through Lindblad collapse operators with adjustable decoherence and relaxation rates. The density matrix evolution which resulted was solved numerically using QuTiP's master equation solver.

Decoherence Sensitivity Analysis

To investigate the influence of environmental noise, a parameter sweep was performed by varying the decoherence rate while maintaining a constant relaxation rate. For each simulation, the excited state population was calculated as a function of time. The maximum excitation probability was recorded.

The analysis enabled a quantitative assessment of how increasing decoherence suppresses coherent excitation dynamics and reduces the efficiency of photoinduced excitation.

Data Visualization

Simulation outputs were visualized using Matplotlib. Figures of the validity of the closed system Rabi oscillations, comparison of closed and open quantum system dynamics, dependence of excitation probability on the decoherence rate, and temporal decay of quantum coherence. These figures form the basis of the analysis presented in the results section.

Validation of the Closed Quantum System

The numerical simulation successfully reproduced the analytical solution for Rabi oscillations in a two level quantum system.

Figure 1 demonstrates excellent agreement between the simulated and theoretical excited state populations, confirming the accuracy of the numerical implementation. Quantitative validation yielded a RMSE of 3.531×10^{-6} , a MAE of 2.692×10^{-6} , and a maximum absolute error of 8.196×10^{-6} . The simulated Rabi frequency differed from the theoretical

value by only 0.0174%, which indicates that numerical errors were negligible throughout the simulation. These results therefore validate the implementation of the Hamiltonian and provide confidence in the subsequent open quantum system simulations.

Influence of Environmental Interactions

To investigate the effects of realistic photovoltaic environments, the model was extended to include decoherence and relaxation through the Lindblad master equation.

Figure 2 compares the evolution of the excited state population for both the closed and open quantum systems. In the system, the excited state population exhibited sustained Rabi oscillations. This indicates continuous coherent energy exchange between the electromagnetic field and the quantum system. The open system simulation, though, displayed progressively damped oscillations. This is due to interaction with the surrounding environment. These environmental effects reduced the oscillation amplitude over time and caused the system to approach equilibrium.

The observed damping demonstrated the influence of environmental interactions on quantum excitation dynamics. This shows the necessity of incorporating open system effects to model realistic photovoltaic materials.

Effect of Decoherence on Excitation Dynamics

A sensitivity analysis was performed by varying the decoherence rate while maintaining a constant relaxation rate and all other model parameters^{5,15}.

Figure 3 illustrates the relationship between decoherence strength and maximum excited state population.

As the decoherence rate increases, the maximum excitation probability decreases. Stronger environmental interactions suppressed coherent quantum evolution, which limits the probability of maintaining electrons in the excited state. This reduction in efficiency reflects the degradation of quantum coherence caused by noise. Relaxation reduces the excited state population. Decoherence suppresses phase coherence and changes the oscillatory dynamics

These results indicate that quantum coherence plays an important role in sustaining photoinduced excitation. Controlling decoherence is essential for understanding the balance between coherent excitation dynamics and dissipative processes.

Decay of Quantum Coherence

The temporal evolution of the off-diagonal density matrix elements was analyzed to quantify coherence loss during the simulation.

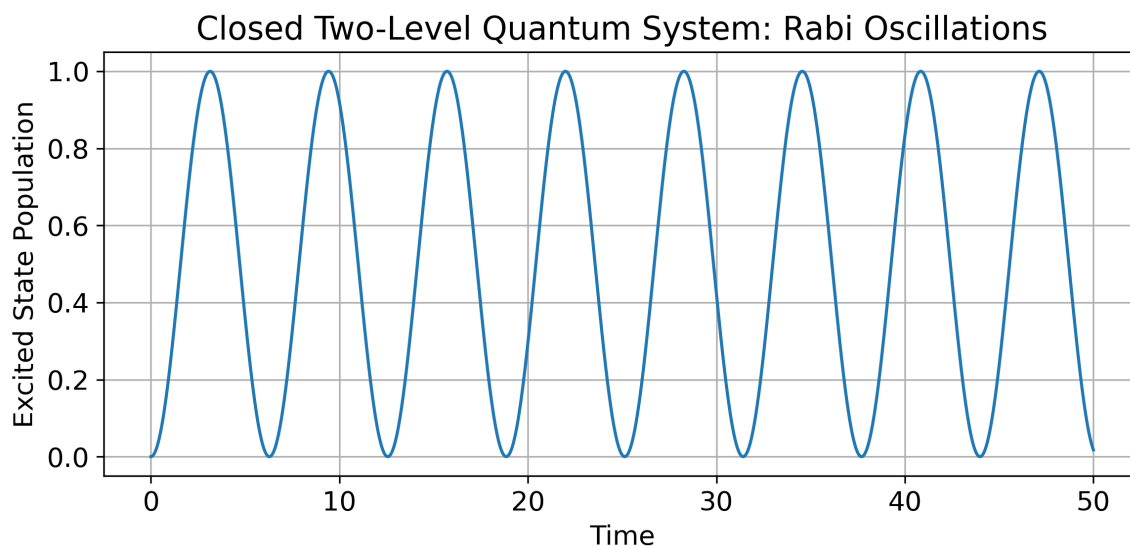


Figure. 1 Comparison of Numerical And Analytical Solutions for Rabi Oscillations

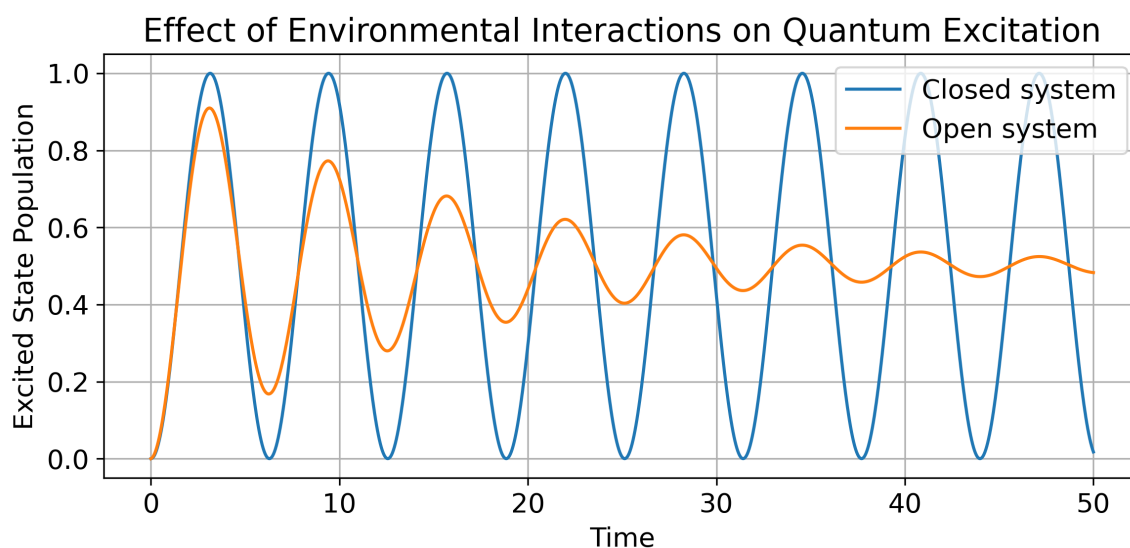


Figure. 2 Evolution of Closed Quantum System and Open Quantum System

Figure 4 shows the magnitude of the coherence term as a function of time. In the absence of environmental interactions, quantum coherence is preserved and leads to a sustenance of oscillatory dynamics. Decoherence is introduced and the coherence term decreases steadily, which indicates the progressive destruction of phase relationships between quantum states. The decay corresponds to the damping observed in the excited state population.

The population dynamics and coherence measurements show that environmental interactions alter the behaviour of

quantum systems by reducing coherent excitation and driving the system towards equilibrium.

Discussion

Physical Interpretation

The oscillatory behaviour observed in the simulations corresponds to Rabi oscillations, a fundamental quantum mechanical phenomenon describing the coherent transfer of population

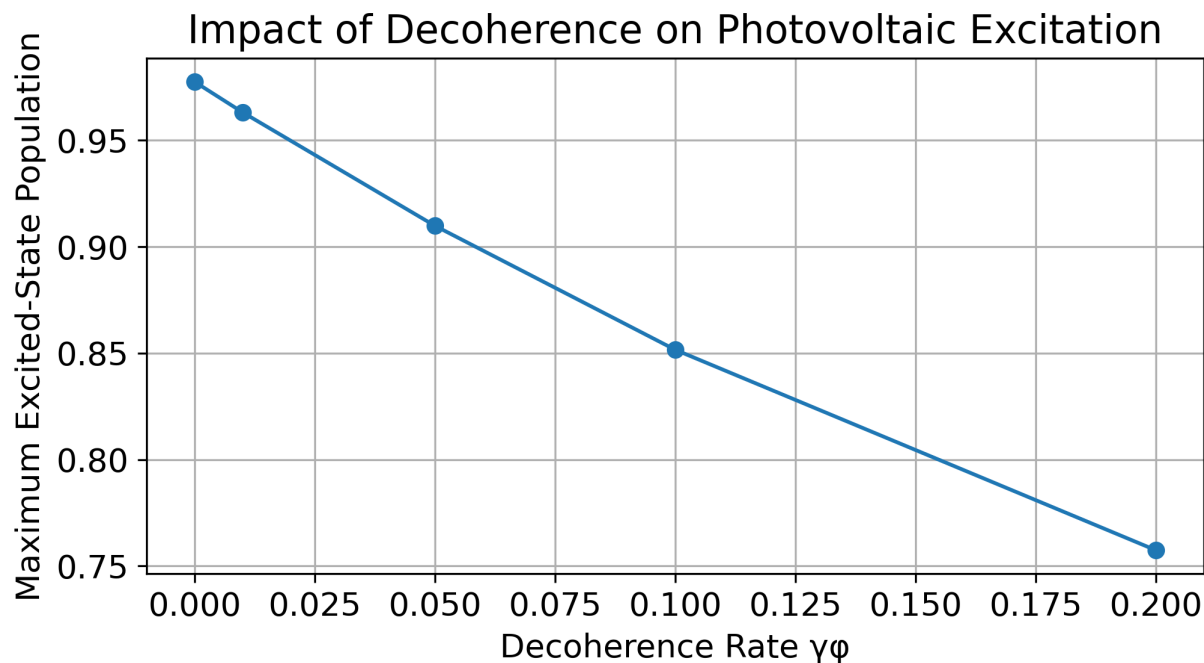


Figure. 3 Impact of Decoherence on Photo-Induced Excitation

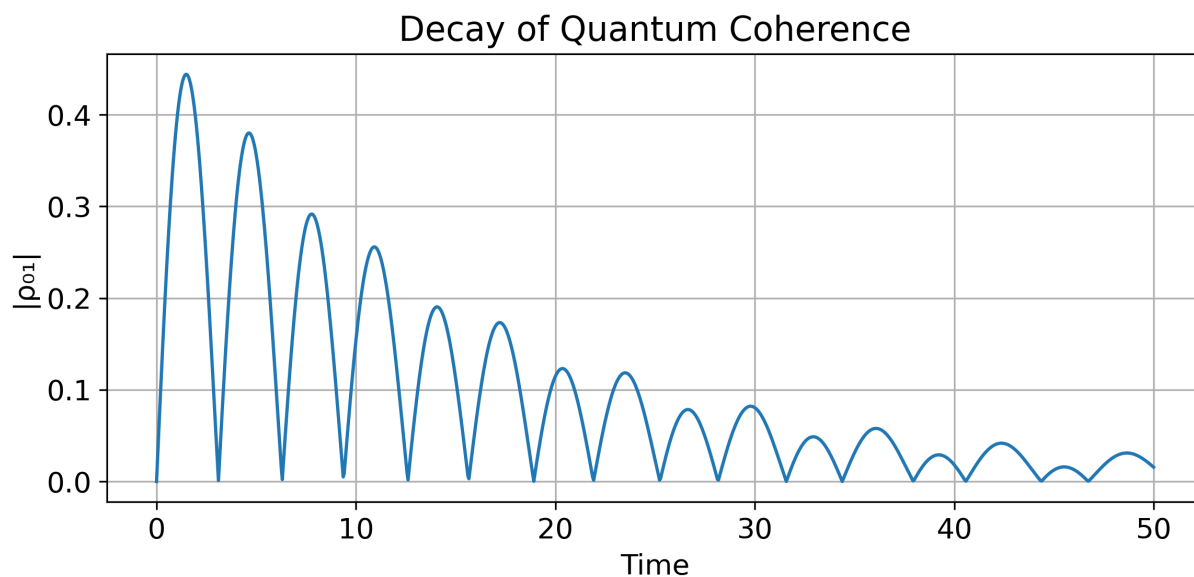


Figure. 4 Decay of Quantum Coherence

between two energy states^{8,10}. As the quantum system interacts with the incident electromagnetic field, the probability of finding the electron in the excited state oscillates as a function of time and coupling strength. The close agreement between the analytical and numerical solutions demonstrates that the

simulation accurately reproduces the expected dynamics of a driven two level quantum system.

Extending the model to an open quantum system demonstrates the significant influence of environmental interactions on these dynamics^{5,15,16}. The closed system exhibits sus-

tained coherent oscillations. However, the introduction of decoherence and relaxation causes the oscillation amplitude to decay. This behavior shows the gradual loss of quantum coherence as the system interacts with its surroundings. Thus it provides a more realistic representation of quantum processes in photovoltaic materials.

Comparison with Experimental Studies

The coherent excitation dynamics observed in this study are consistent with experimental studies of localized semiconductor systems. Stievater et al. and Kamada et al. demonstrated exciton Rabi oscillations in single quantum dots, showing that two level models can accurately describe discrete optical transitions. Zrenner et al. and Ramsay et al. further demonstrated coherent optical control of semiconductor quantum dots, while Press et al. showed precise optical manipulation of quantum states^{26–28}.

These studies support the use of two level models for localized transitions under controlled conditions. However, unlike these experiments, the present model does not include material-specific effects such as phonon interactions. Therefore, it describes instead fundamental coherent excitation dynamics rather than complete photovoltaic device behavior.

Relevance to Solar Energy Conversion

The excitation process represented by the model corresponds to the microscopic optical excitation of an electron into a higher-energy state. This is analogous to photoinduced excitation in localized semiconductor systems. This transition generates charge carriers responsible for electrical current in a solar cell^{1,4}.

Although the two level model is very simplified, it captures the essential quantum mechanics governing photoinduced excitation. In real semiconductor materials, however, coherence times are typically on the femtosecond to picosecond scale depending on material and excitation conditions. This is much shorter than many device operation timescales. Therefore, the coherent dynamics studied here represent the initial excitation regime rather than sustained photovoltaic operations. These interactions produce decoherence and relaxation, reducing the excited state population and thus limits the persistence of coherent quantum dynamics^{5,12,15}.

The open quantum system simulations reproduce this behaviour by using environmental interactions through the Lindblad master equation. The damping of the excited state population and the decay of quantum coherence show the way in which environmental effects influence excitation dynamics in photovoltaic materials. Realistic quantum models of solar energy conversion need to account for both coherent semiclassical light matter interactions and the inevitable loss of coher-

ence caused by the surrounding environment.

Connection to Photovoltaic Devices

The excited state probability calculated in this study represents the likelihood of occupation of the excited quantum state following electromagnetic excitation. In semiconductor materials, this microscopic quantum excitation actually is the first step in conversion of sunlight into electrical energy. After excitation, electrons and the corresponding holes can be separated by the built in electric field of a junction, producing the charge carriers that actually contribute to electrical current^{1,4}.

The simulations show how coherent quantum dynamics governs the initial excitation process. Environmental interactions, represented through the Lindblad master equation, suppress coherence and modify the excited state population.

It is important to note that the model does not directly predict macroscopic photovoltaic quantities power output. These quantities depend on additional processes, including charge transport, and device architecture, which are beyond the scope of the present study. The model presented provides a microscopic description of the processes that precede the phenomena at a larger scale. This model thus establishes a foundation for more comprehensive quantum models of photovoltaic materials and devices^{3,18,19}.

Several assumptions were made to maintain efficiency and emphasize the fundamental physics of photoinduced excitation. The system was represented as a two level model. However, real photovoltaic materials contain multiple electronic bands, many body interactions, excitonic effects, and complex carrier transport mechanisms. Decoherence and relaxation rates are treated as fixed parameters as well.

Future work could extend this model by incorporating multi level electronic structures, experimentally calibrated material parameters, temperature dependent environmental interactions and charge transport processes. Additional investigations using quantum algorithms and emerging quantum computing platforms may also enable the simulation of more complex photovoltaic systems that are challenging to model using classical computational methods^{21,22}.

Conclusion

This study investigated how a two level quantum model can describe the fundamental quantum processes underlying photovoltaic energy conversion. The simulation was successful in reproducing field driven excitation dynamics through Rabi oscillations. This demonstrates that population transfer between quantum states can be represented accurately within this framework. However, these coherent oscillations represent reversible energy exchange between the driving field and the quantum system instead of irreversible carrier extraction.

Therefore, the model does not directly predict steady photovoltaic current or device-level energy conversion, which additionally require processes extraction through electrical contacts.

By extending the model to an open quantum system using the Lindblad master equation, the effects of environmental interactions were incorporated. The results showed that decoherence and relaxation reduce quantum coherence and suppress sustained excitation. This highlights their importance in realistic photovoltaic systems.

However, the two level approximation provides only a microscopic description of photo-induced excitation and cannot predict complete device behavior. Therefore, this model serves as a foundation for the understanding of quantum excitation processes while motivating future studies using more complex material and device level simulations.

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