Nonequilibrium Landau-Zener-Stückelberg spectroscopy in a double quantum dot

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We study theoretically nonequilibrium Landau-Zener-Stückelberg (LZS) dynamics in a driven double quantum dot (DQD) including dephasing and, importantly, energy relaxation due to environmental fluctuations. We derive effective nonequilibrium Bloch equations. These allow us to identify clear signatures for LZS oscillations observed but not recognized as such in experiments [Petersson et al., Phys. Rev. Lett. 105, 246804, 2010] and to identify the full environmental fluctuation spectra acting on a DQD given experimental data as in [Petersson et al., Phys. Rev. Lett. 105, 246804, 2010]. Herein we find that super-Ohmic fluctuations, typically due to phonons, are the main relaxation channel for a detuned DQD whereas Ohmic fluctuations dominate at zero detuning.

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Quantum electronic devices, as qubits realized by double quantum dots (DQD), require coherence times which exceed their quantum operation time during which the DQD is typically strongly driven by external voltage pulses. Tremendous research efforts studied semiconductor-based devices to achieve coherent quantum control\textsuperscript{[4–7]}. Many fluctuation sources of the noisy solid state environment, which act on the electron in the DQD and thus destroy coherence, were revealed but a comprehensive picture is elusive. Furthermore, driving by voltage pulses causes an intrinsic nonequilibrium situation in which relaxation competes with driving\textsuperscript{[8, 9]} which renders a theoretical description of the dissipative nonequilibrium dynamics highly nontrivial.

Here, we theoretically study the dissipative nonequilibrium dynamics of a single electron charge qubit defined in a DQD embedded in a noisy solid state environment driven by voltage pulses. While DQD charge qubits have relatively short coherence times, this disadvantage is compensated by the possibility of fast quantum operations. We model the DQD and its dissipation as a quantum two-level system in an open quantum system approach\textsuperscript{[11]}. We determine the dissipative nonequilibrium real time dynamics (initialized by applying voltage pulses) by deriving effective nonequilibrium Bloch equations (NBEs). These allow fast numerical treatment in contrast to numerical exact methods\textsuperscript{[10]} and thus allow a comprehensive analysis of recent experiments by Petersson et al.\textsuperscript{[1]} and Dovzhenko et al.\textsuperscript{[12]}. In these experiments quantum control of a single electron was achieved by means of applying ultra short voltage pulses to control gates of the laterally defined DQD. In these time ensemble measurements the DQD was cycled (with 40 MHz repetition rate) between two different ground state configurations while its average charge occupation was continuously detected via the electric current through a capacitively coupled quantum point contact (QPC). The applied voltage pulses generated Landau-Zener-Stückelberg (LZS) dynamics\textsuperscript{[13]} and we can identify so far unexplained features in the experimental data as signatures of coherent LZS oscillations.

In the experimental ensemble measurements the dephasing time $T_\varphi^2$ due to slow noise\textsuperscript{[7, 14]} is much shorter than relaxation times $T_1$ and thus dominates the decoherence times $T_2$ since $T_2^{-1} = T_1^{-1} + (2T_3)^{-1}$. Relaxation and dephasing could be caused by thermal phonons, intrinsic or externally triggered charge noise\textsuperscript{[15, 18]}, or detector back-action\textsuperscript{[10, 22]}. Ref.\textsuperscript{[1, 12]} neglected relaxation and employed solely an $1/f$ noise model\textsuperscript{[23–27]} for dephasing which allowed them to describe the detuning dependence of the observed decoherence of the DQD.

The experimentally observed steady state charge occupation of the DQD is, however, heavily influenced by relaxation\textsuperscript{[10]}. Therefore, in this letter, we go beyond this simple dephasing model and include various environmental fluctuation spectra to describe dephasing and relaxation. This allows us to simulate the full nonequilibrium real time dynamics experimentally studied. We find that the observed visibility reduction is caused by relaxation. Moreover, by analyzing the measured LZS dynamics we identify the full environmental fluctuation spectrum as a sum of three processes. In addition to slow noise, already considered in ref.\textsuperscript{[1]}, which causes detuning dependent dephasing, super-Ohmic fluctuations, as typically originating from phonons, are the main relaxation channel for the detuned DQD. Near zero detuning, however, Ohmic fluctuations dominate relaxation. The latter also limit the experimentally observed maximal decoherence time of $T_2 \sim 7$ ns.

\textbf{Modelling of pulse driven DQD} We model the single electron DQD using a two-level system Hamiltonian

\begin{equation}
H = \frac{\hbar}{2}\Delta \sigma_x + \frac{\hbar}{2}\epsilon(t)\sigma_z
\end{equation}

where the eigenstates of $\sigma_z$ correspond to the electron in the left / right dot, $\epsilon(t)$ is the level detuning and $\Delta$ is the interdot tunnel splitting\textsuperscript{[28]}. Experimentally,
gate voltage pulses are applied to change the level detuning as sketched in Fig. 1(left). Initially the detuning is \( \epsilon_0 = \epsilon(0) \gg \Delta \) and the DQD in the according ground state (0,1) [electron is in the right dot]. A voltage pulse then drives the system within a rise time \( t_r \) to a plateau detuning \( \epsilon_p \) close to resonance (\( \epsilon = 0 \)) and keeps it there for a plateau time \( t_p \). Then the DQD is driven back within \( t_r \) to the initial detuning and the probability \( P_{(1,0)} \) of occupation of the excited state (1,0) [electron in the left dot] is studied as a function of the pulse duration \( t_v = 2t_r + t_p \). For a quantum mechanical two-level system without dissipation we expect coherent oscillations in the voltage pulse time \( t_v \).

A voltage change on a single gate [1, 12] not only affects the level detuning but also causes a common energy shift of both states (1,0) and (0,1) which is not relevant in the following and thus neglected. The sharp corners of \( \epsilon(t) \) in Fig. 1(left) are smoother in reality and might even contain oscillatory features (due to a finite bandwidth transfer function). At voltage pulse times \( t_v \gtrsim 2t_r \) deviations between linear voltage ramps and more accurate descriptions are negligible.

Solving the quantum dynamics of the DQD with a single applied voltage pulse (details given below) results in the probability \( P_{(1,0)}(t = t_v) \) after the voltage pulse, plotted in Fig. 2 as a function of \( t_v \) and plateau detuning \( \epsilon_p \). In order to model the experiments [1, 12] we use \( \Delta/h = 4.5 \text{ GHz}, \epsilon_0 = 200\mu\text{eV} \approx 11\Delta \) and \( t_r = 35\text{ ps} \) which corresponds to the fastest experimental achievable rise time [24]. The sweep speed \( v_p = |\epsilon_p - \epsilon_0|/t_r \) can be approximated by \( v_0 = |\epsilon_0|/t_r \) for \( \epsilon_0 \gg \epsilon_p \) [slope in Fig. 1(left)] which provides an indication on the overall adiabaticity of the dynamics [13]. For \( v_0 = 200\mu\text{eV}/35\text{ps} \approx 11\Delta^2/h \), Fig. 2 shows coherent oscillations between states (0,1) and (1,0) with frequency \( E/h \) and eigenenergy \( E = \sqrt{\Delta^2 + \epsilon_p^2} \) of Eq. (1). At zero detuning \( P_{(1,0)}(t_v) \) oscillates between 0 and 1. With increased \( \epsilon_p \) the oscillation frequency grows while its amplitude gradually decreases. Fig. 2 shows a clear asymmetry: the visibility is larger for \( \epsilon_p < 0 \) compared to \( \epsilon_p > 0 \). Remarkably, this asymmetry was observed experimentally, too (but not explained) [1]. The asymmetry decreases with increasing sweep speed as highlighted by the inset of Fig. 2 which shows an almost symmetric \( P_{(1,0)}(t_v) \) for \( \epsilon_0 = 2000\mu\text{eV} \) leading to \( v_0 \gtrsim 110\Delta^2/h \). We conclude that the asymmetry is solely an effect of adiabaticity when the DQD is driven through the avoided crossing at zero detuning. For \( \epsilon_p \leq 0 \), the quantum system accumulates, during the voltage pulse, not only phase due to the coherent oscillation of the electron between the two dots but also due to the superposition state occupied between two successive Landau-Zener transitions [13]. As such, the asymmetry is a clear signature of coherent LZS oscillations.

Periodically Cycled Pulses In an ensemble measurement with continuous charge detection, as in Ref. [1] with \( t_{\text{rep}} = 25\text{ ns} \), it is essential to choose \( t_{\text{rep}} \gg t_v \) to ensure readout of the charge occupation after application of the pulses (of duration \( t_v \lesssim T_2 \)). An interpretation in terms of an ensemble measurement, which simply averages over many independent shots, further requires \( t_{\text{rep}} \gg T_2 \) and, interestingly, \( t_{\text{rep}} \sim T_1 \), where \( T_1 \) is the (thermal) energy relaxation time which depends on the detuning: for \( t_{\text{rep}} \gg T_1 \) initialization into configuration (1,0) is guaranteed, but the visibility of the continuous measurement is close to zero as mostly (1,0) is occupied; for \( t_{\text{rep}} \ll T_1 \) initialization into (1,0) independently of \( P_{(1,0)} \) right after the pulse— is impossible. No matter of the choice of pulse sequence, continuously cycled pulses will result in a steady state which, in principle, contains the information of dephasing and energy relaxation times [10].

In order to model the experimental repeated pulse train sequence we replace our simulation after the first cycle (up to the repetition time \( t_{\text{rep}} \)) with the final statistical operator of the previous cycle as initial state. This procedure is repeated until the population \( P_{(1,0)} \) [mea-
sured as average over a full cycle as plotted in Fig. [left] changes by less than 0.005. This \( \tilde{P}_{(1,0)} \) approximates the experimentally observed steady state population.

**Driven dissipative dynamics** Including dissipation into the driven dynamics of the DQD causes a competition between driving and relaxation [8, 9] which renders all but expansive numerical treatments inadequate. Following the standard approach within open quantum dynamics [11] we couple the driven two-level Hamiltonian of the DQD to environmental fluctuations described by harmonic oscillators. This results in

\[
H_{\text{tot}} = H(t) + \frac{\sigma}{2} \sum_k \lambda_k (b_k + \hat{b}_k^\dagger) + \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k
\]  

(2)

with bosonic annihilation/creation operators \( b_k / \hat{b}_k^\dagger \). The spectrum \( G(\omega) = \sum_k \Delta_k^2 \delta(\omega - \omega_k) \) is typically a smooth function [11] at the energies of interest, i.e. \( G(\omega) = 2 \omega_s \exp(-\omega/\omega_c) \) with spectral exponent \( s \), cut-off frequency \( \omega_c \), and the coupling strength \( \sigma \). Typically [1], \( \Delta \gg \gamma_2 \gg \gamma_1 \) with the relaxation rate \( \gamma_1 = 1/T_1 \) and the decoherence rate \( \gamma_2 = 1/T_2 \), which justifies a weak DQD-environment coupling approach. Due to the time dependence of \( H(t) \) the weak coupling Born-Markov approximation, however, fails. Assuming that the change of energy during the memory time in the environment is small, allows an additional adiabatic rate approximation. For this, we switch to the time dependent rate approximation. For this, we switch to the time dependence of  \( \gamma_1(t) = \frac{\pi}{2 \hbar} \coth[\frac{1}{2} \beta E(t)] u(t) G[E(t)] \) \( \gamma_2(t) = \frac{\pi}{2} \gamma_1(t) + \Gamma_2(t) \)

(4)

(5)

where \( \Gamma_2(t) = 1/T_2^2(t) \) is the decohersing time. Herein, \( \phi(t) = \arctan[\epsilon(t)/\Delta] \), \( u(t) = \cos \phi(t) \), and \( v(t) = \sin \phi(t) \). The presented nonequilibrium Bloch equations with time-dependent rates and momentary equilibrium describe adequately dissipative Landau-Zener dynamics, i.e. the regime of competition between driving and relaxation, for weak DQD-environment coupling. We ensured this by extensive comparison with numerical exact results [8, 9]. Details will be presented elsewhere.

To proceed, we need the spectra of fluctuations acting on the DQD. Estimates can be gained by the observations for fixed DQD parameters of Petersson et al. [1] of a relaxation time \( T_1(\epsilon_0) = 10 \text{ns} \) at the initial detuning \( \epsilon_0 \), a decoherence time of \( T_2 \approx 7 \text{ns} \) at zero detuning and the detuning dependent decohersing times presented in Fig.4c of Ref. [1]; all at the temperature \( T = 80 \text{mK} \).

**Relaxation rates** Charge fluctuations and bulk phonons likewise couple to the DQD. In order to include both, we consider an Ohmic fluctuation spectrum with \( s = 1 \) (charge fluctuations) as well as a super-Ohmic one with \( s = 3 \) (phonons) [11]. The rate [1] reflects a one-boson process and accordingly only yields a substantial relaxation if the spectrum at the eigenenergy of the DQD is finite, i.e. \( E(t) \ll \omega_c \).

At fixed \( T, \Delta \) and \( \epsilon_0 \) we can determine the coupling strength \( \sigma \) (assuming \( E(t) \ll \omega_c \)) using the known energy relaxation time \( T_1(\epsilon_0) \), and estimate \( T_1(\epsilon = 0) \) using Eq. [1] and \( u(t) = \Delta / E(t) \). For Ohmic fluctuations \( T_1(\epsilon = 0) \approx (\Delta / \sqrt{\Delta^2 + \epsilon_0^2}) T_1(\epsilon_0) \) while for super-Ohmic fluctuations \( T_1(\epsilon = 0) \approx (\sqrt{\Delta^2 + \epsilon_0^2}/\Delta) \cdot T_1(\epsilon_0) \). With the reported \( T_1(\epsilon_0) = 10 \text{ns} \) we find \( T_1(\epsilon = 0) \approx 900 \text{ps} \) assuming Ohmic fluctuations compared to \( T_1(\epsilon = 0) \approx 110 \text{ns} \) assuming super-Ohmic fluctuations. According to Eq. [5] energy relaxation causes an upper bound of the decoherence \( T_2 \leq 2T_1 \). The decoherence time of \( T_2 = 7 \text{ns} \), observed in the experiment at \( \epsilon = 0 \), lies well in between our predictions \( T_1^{\text{eq}} = 7 \text{ns} \ll T_1^{\text{fin}} \).
An Ohmic fluctuation spectrum alone – consistent with \( T_1(\epsilon_0) = 10 \text{ ns} \) – would result in much too fast decoherence at \( \epsilon = 0 \) compared to the experimental results. Hence, super-Ohmic fluctuations, typically caused by phonons, are the main relaxation mechanism at \( \epsilon_0 \).

**Decoherence and Dephasing rates** For a super-Ohmic spectrum \( \Gamma_2(t) \equiv 0 \), and the decoherence rate \( \gamma_2 \) [see Eq. (5)] would be solely determined by the energy relaxation mechanism. The spectrum of charge noise, which is well known to cause additional dephasing, strongly depends on the sample and how it has been treated \[1\]. Often, charge noise can be assumed to be slow noise as done by Petersson et al. \[1\]. They use an \( 1/f \) dephasing model \[25\] which typically results from background charge noise \[23, 24\] and might be described in terms of sub-Ohmic noise \[26, 27\] with \( s = 0 \) and \( \omega_c \ll \Delta \). Such slow noise is a major dephasing source in realistic devices and causes solely dephasing. We aim at unraveling the relaxation mechanisms present rather than the dephasing sources. Relaxation is not influenced by slow noise and thus we describe slow noise here using a simplified Ohmic slow noise model \[25\] numerically results in Fig. 3, the main result, where we plot the steady state occupation \( \bar{P}_{(1,0)} \) versus voltage pulse time and plateau detuning for a tunnel coupling \( \Delta/h = 4.5 \text{ GHz} \). In comparison, the inset shows the instantaneous probability \( \bar{P}_{(1,0)}(t = t_o) \) with the color code stretched by a factor of two, i.e. yellow = 1. The continuous current measurement in the QPC reduces the visibility for the coherent dynamics by a factor of 2. In comparison to the undamped case in Fig. 2 we observe an overall reduction in oscillation amplitude. On top, the LZS oscillations smear out with both, more negative detunings \( \epsilon_p \) and longer voltage pulse times \( t_o \); \( \bar{P}_{(1,0)} \) is flat in the upper right corner of Fig. 3. The same behaviour was experimentally observed, but not explained, by Petersson et al. \[1\]. Our results indicate that the LZS oscillations smear out as a result of relaxation but the remaining asymmetry with respect to \( \epsilon_p \) is a result of adiabaticity. Thus, the experimentally observed feature is a clear signature of LZS oscillations. The overall qualitative agreement between our simulation results and the experimental data \[1\] is very good. The quantitative overestimation of the oscillation amplitude being a factor of 2 is likely due to uncertainties regarding experimental details \[32\].

**Interdot tunnel coupling** Petersson et al. \[1\] observe an increased (decreased) oscillation amplitude for a smaller (larger) interdot tunnel coupling, i.e. \( \Delta/h = 3.3 \text{ GHz} (6.6 \text{ GHz}) \). We find the same tendency. It results from the fact that the relaxation rate [eq. (4)] increases by a factor of two, i.e. yellow = 1. The continuous current measurement in the QPC reduces the visibility for the coherent dynamics by a factor of 2. In comparison to the undamped case in Fig. 2 we observe an overall reduction in oscillation amplitude. On top, the LZS oscillations smear out with both, more negative detunings \( \epsilon_p \) and longer voltage pulse times \( t_o \); \( \bar{P}_{(1,0)} \) is flat in the upper right corner of Fig. 3. The same behaviour was experimentally observed, but not explained, by Petersson et al. \[1\].}

\[ G(\omega) = 2\alpha_1 \omega^3 e^{-\omega/\omega_c} + \sum_{j=2}^{3} 2\alpha_j \omega e^{-\omega/\omega_{c,j}} \]  

with \( \omega_{c,1}, \omega_{c,3} \gg \Delta \) and \( \omega_{c,2} \ll \Delta \). Choosing \( \alpha_1 \Delta^2 = 8.09 \cdot 10^{-5} \), \( \alpha_2 = 3.53 \cdot 10^{-2} \) and \( \alpha_3 = 2.73 \cdot 10^{-3} \) for the coupling strengths reproduces the experimental observations \[1\] of \( T_1(\epsilon_0) = 10 \text{ ns}, T_2(\epsilon = 0) \sim 7 \text{ ns} \) and the decoherence time versus detuning with measured data in Fig. 4c of Ref. \[1\] and our predicted decoherence rate plotted as black full line in Fig. 1(right).

**Dynamics of the driven dissipative DQD** Solving the nonequilibrium Bloch equations \[1\] with the fluctuation spectrum \[6\] numerically results in Fig. 3, the main result, where we plot the steady state occupation \( \bar{P}_{(1,0)} \) versus voltage pulse time and plateau detuning for a tunnel coupling \( \Delta/h = 4.5 \text{ GHz} \). In comparison, the inset shows the instantaneous probability \( \bar{P}_{(1,0)}(t = t_o) \) with the color code stretched by a factor of two, i.e. yellow = 1. The continuous current measurement in the QPC reduces the visibility for the coherent dynamics by a factor of 2. In comparison to the undamped case in Fig. 2 we observe an overall reduction in oscillation amplitude. On top, the LZS oscillations smear out with both, more negative detunings \( \epsilon_p \) and longer voltage pulse times \( t_o \); \( \bar{P}_{(1,0)} \) is flat in the upper right corner of Fig. 3. The same behaviour was experimentally observed, but not explained, by Petersson et al. \[1\]. Our results indicate that the LZS oscillations smear out as a result of relaxation but the remaining asymmetry with respect to \( \epsilon_p \) is a result of adiabaticity. Thus, the experimentally observed feature is a clear signature of LZS oscillations. The overall qualitative agreement between our simulation results and the experimental data \[1\] is very good. The quantitative overestimation of the oscillation amplitude being a factor of 2 is likely due to uncertainties regarding experimental details \[32\].

**Conclusions** We studied theoretically the dissipative nonequilibrium dynamics of a single electron DQD driven by voltage pulses. We couple the DQD additionally to environmental fluctuations causing relaxation and dephasing. Extending standard Born-Markov approaches to driven systems, we derive nonequilibrium Bloch equations exhibiting time dependent rates and the momentum equilibrium. This approach allows efficient numerical simulations of the full nonequilibrium real time dynamics and a comprehensive analysis of the experimental data \[1\]. We identify an asymmetric occupation of the left dot in respect to the detuning between the dots in ref. \[1\] as a clear experimental signature of LZS dynamics. A full analysis of the LZS dynamics furthermore allows us to specify the full environmental fluctuation spectrum acting on the DQD studied by Petersson et al. \[1\] as sum of three processes. Besides slow noise causing
the detuning dependent strong dephasing, super-Ohmic fluctuations, as typically originating from phonons, are the main relaxation channel for a detuned DQD. At zero detuning, however, Ohmic fluctuations, which might be caused by gate voltage noise, dominate relaxation and are also the main cause for the decoherence at zero detuning.

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