

Measuring the measurement problem: controlling decoherence with measurement duration in molecular MCB junctions

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We investigate quantum coherence in molecular MCB junctions operating in a tetrahydrofuran partially wet phase. These systems represent a distinct class of enclosed open quantum systems with unusually long decoherence times at ambient conditions, on the order of 1–20 ms. By tuning the integration time of the current measurement relative to the decoherence time, we observe a transition from quantum interference patterns, manifested as structured bands of data points, to classical behavior characterized by a single averaged response. This offers new insights into quantum decoherence dynamics.

The measurement problem addresses the apparent discontinuity between the deterministic unitary evolution of a quantum state and the probabilistic outcome observed upon a measurement [1]. Making a measurement or observation within the context of the Copenhagen interpretation implies that the quantum world is completely separated from our classical world. Here, the classical world also includes the measurement equipment. A measurement is treated as an external, classical process, leaving the mechanism of wavefunction collapse unresolved. The measurement itself is a physical action, superimposed on the system it measures, which implies that we can describe the two systems combined as a new system. Providing the measurement point in the equipment has not been observed by a human, we can add the human to the combined system, again arriving at a higher-level new system. In this way, we will end up with a Schrödinger’s cat analogy. Decoherence theory provides for a partial resolution by describing how entanglement with an environment suppresses quantum interference, effectively yielding classical outcomes without invoking an explicit collapse.

Open quantum systems, where a quantum system interacts with its environment, are central to understanding decoherence [2, 3]. Before any interaction between the quantum system and the environment has taken place, the total wave function of the quantum system and environment combined can be described as:

$$|\psi\rangle_{\text{OQS}} = |\psi\rangle_{\text{QS}} |\psi\rangle_{\text{E}} \quad (1)$$

with $|\psi\rangle_{\text{OQS}}$ the wavefunction of the open quantum system, comprising the quantum system, $|\psi\rangle_{\text{QS}}$, and the environment, $|\psi\rangle_{\text{E}}$. When the quantum system starts to interact with the environment, the two wavefunctions $|\psi\rangle_{\text{QS}}$ and $|\psi\rangle_{\text{E}}$ start to become entangled and they can no longer be separated, hence the appearance of Eq. 1 becomes much more complex. Physics describes the decohering of the quantum system, how the quantum mechanical character of the quantum system is lost, or transferred to the classical world, due to the interaction with the environment. Thus, quantifying the collapse of a

wavefunction at the quantum to classical transition. Decoherence theory implies that not all coherence is lost at once but over a typical decoherence time, τ_c , as information flows from the quantum system to the environment. As early as 1970, Zeh [4] stressed that realistic quantum systems are never closed, never coherent forever. It was demonstrated that a quantum system obeying the Schrödinger equation becomes intensely entangled with its environment. It is this evolving entanglement that will suppress the quantum character, such as interference and coherence.

Decoherence is a real quantum mechanical effect related to enduring entanglement, for an insightful experiment see Ref. [5]. It is an extremely efficient process; due to the many degrees of freedom of the environment, the ongoing entanglement is often considered irreversible. To use a quantum system for practical purposes, for example quantum computers or quantum sensors, the superposition of states is the critical aspect that is being exploited. It is this superposition in a quantum system which is very susceptible to decoherence and therefore easily destroyed. Decoherence times can be extremely short, much shorter than the times required to reach thermal equilibrium. After Zeh, over several decades, the field of decoherence has been developed to the current level of understanding; see for example Refs. [6–8].

Here we show results from a specific type of open quantum system, the molecular MCB junction in the tetrahydrofuran partially wet phase. A bridging molecule between two electrodes represents the quantum system, the partially wet phase represents a controlled environment, a Faraday cage encloses the entire system from the larger environment (Fig. 1). We will call such a system an “enclosed open quantum system”. Most studies assume Markovian dynamics with unidirectional information flow. Here, we focus on enclosed open quantum systems, where the environment is controlled and isolated, enabling strong non-Markovian effects [9] and a bidirectional exchange of information (Fig. 2). It has been unclear to this point under which exact conditions the enclosed open quantum system shows coherence. Here we experimentally demonstrate that the critical quantity

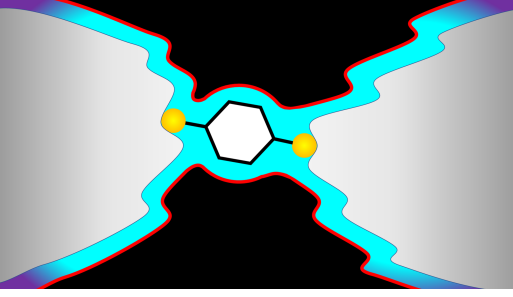


FIG. 1. An enclosed open quantum system with a single molecule representing the quantum system, a controlled environment represented by the blue THF partially wet phase and the enclosing Faraday cage represented by the red liner embodying the surface potential trapping charge carriers at the THF-air interface.

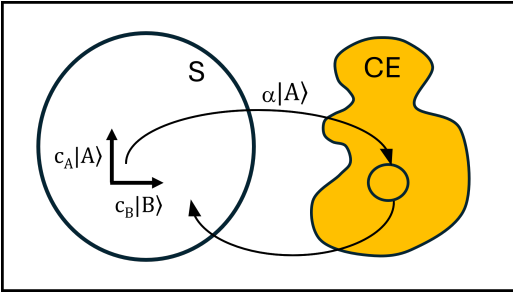


FIG. 2. The information flow between the molecule S and the controlled environment CE. A bi-directional flow of information ensures a memory action of the system.

is the measurement time in relation to the decoherence time of the system.

The experimental set up allows for breaking a gold wire in a tetrahydrofuran (THF) environment at ambient conditions [9]. After a controlled drying process, a single molecule mechanically controllable break (MCB) junction in the partially wet phase is created via self-assembly. The THF partially wet phase provides for a Faraday cage, offering the special conditions for enclosed open quantum systems. We performed the measurements on two bending beam assemblies (BBA1,2), BBA1 in a benzene dithiol (BDT) THF solution and BBA2 in a THF only solution. Current-voltage (IV) characteristics consisting of 1000 data points per scan are voltage biased and are recorded in the partially wet phase. A typical IV curve contains two scans, one with an increasing and one with a decreasing voltage, without a delay at the point of scan reversal. A measurement of the MCB device is defined by the duration of the integration time τ_m , integrating the charge that flows through the junction (Fig. 3). The current is calculated by dividing the integrated charge by τ_m . The delay time τ_{delay} is used to calculate the current from the previous measurement, store this value, and for some wait-time of the new voltage value prior to the next measurement. A single scan

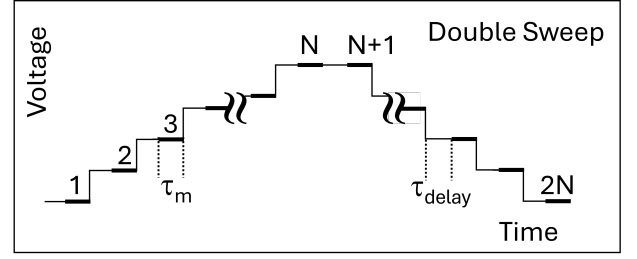


FIG. 3. A measurement consists of an integration time τ_m and a delay time τ_{delay} , $N = 1000$. During τ_m the total charge that flows through the junction is integrated. During τ_{delay} the measurement current is calculated and stored.

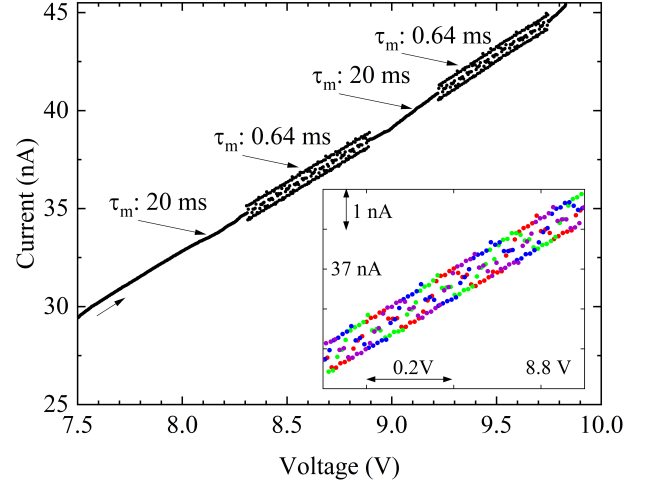


FIG. 4. Toggling the measurement time between $\tau_m = 20$ ms and $\tau_m = 640 \mu\text{s}$ indicates the sensitivity of quantum coherence in relation to the measurement time.

($N=1000$) lasts 45 seconds for $\tau_m = 20$ ms and 5 seconds for $\tau_m = 640 \mu\text{s}$. High-speed measurements, $\tau_m = 640 \mu\text{s}$, lead to a lower resolution and a higher measurement noise band, while slower measurements, $\tau_m = 20$ ms, exhibit the opposite behavior. The noise bandwidth has been measured on a fixed $50 \text{ M}\Omega$ resistor to be 15 pA for $\tau_m = 20$ ms scans and 50 pA for $\tau_m = 640 \mu\text{s}$ scans within a $9\text{--}10 \text{ V}$ range, well below the presented structure in the data.

The IV curve under conditions where τ_m has been toggled in BBA1 between $640 \mu\text{s}$ and 20 ms (Fig. 4) clearly shows structure for the $\tau_m = 640 \mu\text{s}$ sections where data points seem to favor alternating positions at the extremes and the middle position of a band. Every fourth data point creates the data lines at the extremes of the band. Therefore, the data has been separated into four groups consisting of group1: point 1, 5, 9... group2: point 2, 6, 10... and so forth. The inset shows a part of the data with a repetitive pattern, where colored data points change from one extreme to the opposite extreme via the middle position. The 1000 points of this curve

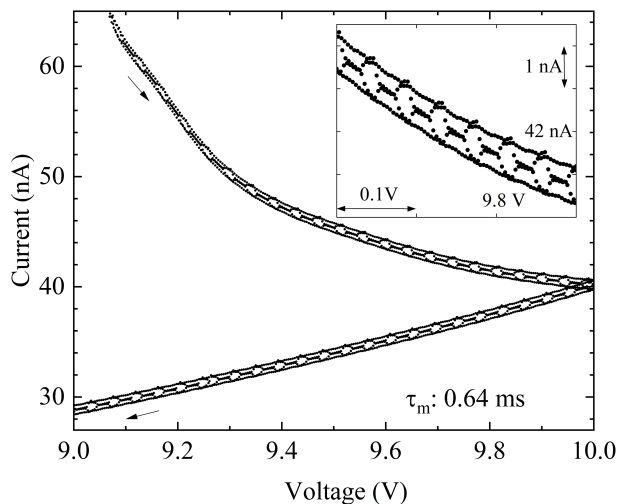


FIG. 5. A consistent bandwidth and a clear repetitive structure, visible as a pattern in the data.

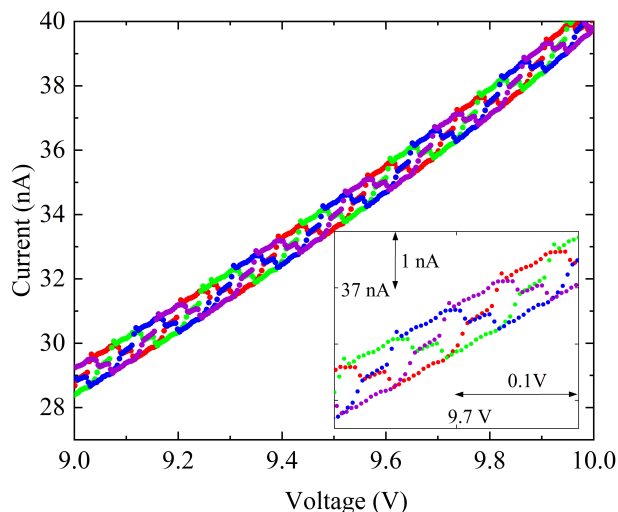


FIG. 6. The lower branch of Fig. 5 split into four groups reveals a recurring interleaved pattern.

extend over 2.5 V. The measurement equipment caters for integer mV values only, thus the voltage separation between sequential data points alternates, 2 mV, 3 mV, 2 mV, 3 mV and so forth, implying a delta of 10 mV between same color points. For all the colored data the measurement sequence in the scan direction is the same: green, blue, purple, red, green and so forth.

The IV curve from BBA2 (Fig. 5) shows a typical behavior towards the end of the partial wet phase with a relatively fast reduction in conductivity. A band of data points is visible in the entire curve. Similarly, as before, four groups of data were created, showing the interleaved pattern in Fig. 6. This graph displays an equivalent repeating pattern of data points as compared to the fast measurement sections in Fig. 4. Here the separation be-

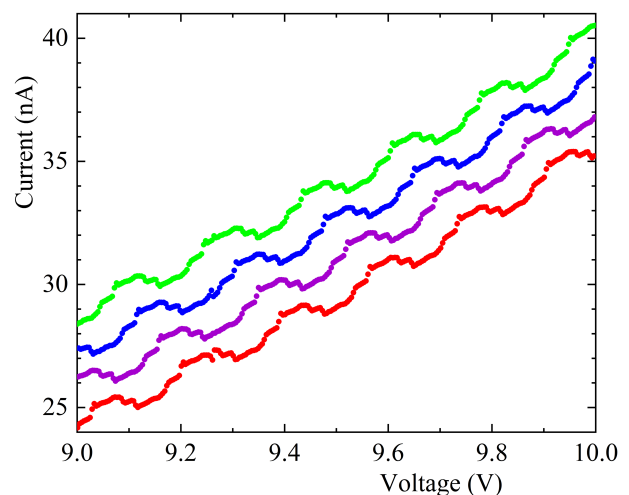


FIG. 7. A further breakdown of Fig. 6, blue, purple, and red groups offset show phase-shifted similarity.

tween sequential data points is 1 mV. In Fig. 7, each group is separately indicated, with the blue, purple and red groups offset by -1.5 nA, -3 nA and -4.5 nA respectively. The data sets of the various groups look identical, but phase-shifted in time. Colored groups of data points alternate between the two current extremes and the middle position of the data band with typically an amplitude of 1 nA for both Figs. 4 and 5. The structure of the colored groups has a periodicity of 160 mV and 172 mV which is equivalent to 0.32 and 0.86 seconds at the scan-speed of 500 mV s^{-1} for the fast-speed measurement sections of Fig. 4 and 200 mV s^{-1} for Fig. 5 respectively.

The data in Fig. 4 are exemplary for toggling τ_m ; there have been at least over a dozen observations of the same effect in as many different bending beam assemblies. This seems a generic effect in THF partially wet phase molecular junctions. Bands of data points in IV curves with $\tau_m = 20 \text{ ms}$ have also been observed [9]. They are, however, less common than at high-speed measurements. Also, in the $\tau_m = 20 \text{ ms}$ bands a structured pattern of data points emerges within a fixed amplitude, however, with a varying periodicity and oscillatory behavior as opposed to a constant periodicity and a more "digital" behavior demonstrated in Figs. 4 and 5. The correspondence between the two types of band curves is the alternation of data points, and clear grouping and interleaving of data points. The alternation of the data, as well as the typical pattern of the data, is a sign that both types of bands belong to a common origin.

A clear image emerges of the conditions under which the band of data points appears. Reducing the measurement time favors the band to occur. For long measurement times, the data band may still show occasionally but far less frequently and in far fewer BBA data sets. Here we argue that with the measurement time we have

a knob which we can tweak, either to cause τ_m to be shorter or longer than the decoherence time of the system. For $\tau_m \ll \tau_c$ the response of the quantum system shows a band of data points, for $\tau_m \gg \tau_c$ a line of data points results. This implies that the typical decoherence time of these quantum systems is in the 1 ms to 20+ ms range, based on many experimental observations with $\tau_m = 640 \mu\text{s}$ or $\tau_m = 20 \text{ ms}$. Since the band of data points does occur at some $\tau_m = 20 \text{ ms}$ measurements, we infer that τ_c for those instances must have been 20+ ms, providing for an approximate upper limit of the decoherence time estimate. The lower limit of 1 ms is based on the generic finding that as a rule the fast measurement, $\tau_m = 640 \mu\text{s}$, toggled with the $\tau_m = 20 \text{ ms}$ measurement provides data as shown in Fig. 4.

For enclosed open quantum systems due to strong non-Markovianity, next to the flow of information from the quantum system to the controlled environment, there must be a flow of information from the controlled environment to the quantum system. The flow of information between the quantum system and the controlled environment is treated as a diffusion process with a typical time constant τ_c . The quantum system is in equilibrium with the controlled environment; performing a measurement acts on the controlled environment and will disturb this equilibrium. Let's assume the quantum system consists of a molecule with states $|A\rangle$ and $|B\rangle$ in a superposition, $|\psi\rangle = c_A |A\rangle + c_B |B\rangle$ where c_A and c_B are the probability amplitudes: $c_A^2 + c_B^2 = 1$. We suppose state $|A\rangle$ has leaked $\alpha |A\rangle$ to the controlled environment, see Fig. 2. A measurement will erase $\alpha |A\rangle$ from the environment, this will proliferate through the measurement system and finish as a data point in the measurement equipment. This process will distort the equilibrium in the controlled environment. Since $|A\rangle$ character is depleted $|B\rangle$ character will predominantly flow from the controlled environment to the quantum system in a typical time τ_c subsequently the $|B\rangle$ vector of the quantum system is bolstered and will start to leak to the controlled environment over the same typical time τ_c . As long as $\tau_m \ll \tau_c$ we will measure the two distinct states. The time resolution of the measurement is below the decoherence time, enabling a measurement of a separate $|A\rangle$ and $|B\rangle$ state. In the other extreme where $\tau_m \gg \tau_c$ at the start of the integration time $\alpha |A\rangle$ will be erased. At some point $|B\rangle$ character starts to flow to the quantum system and will leak $\beta |B\rangle$ to the controlled environment. Multiple information-flow cycles are measured during the same integration interval, hence measuring multiple $\alpha |A\rangle$ and $\beta |B\rangle$ associated current values. Thus, the measurement will display an average of the $\alpha |A\rangle$ and $\beta |B\rangle$ associated current values. The separate quantum values get averaged and thus lost.

The interplay between the measurement equipment and the enclosed quantum system may lead to interference between the measurement-frequency and the information diffusion rate between the molecule and the con-

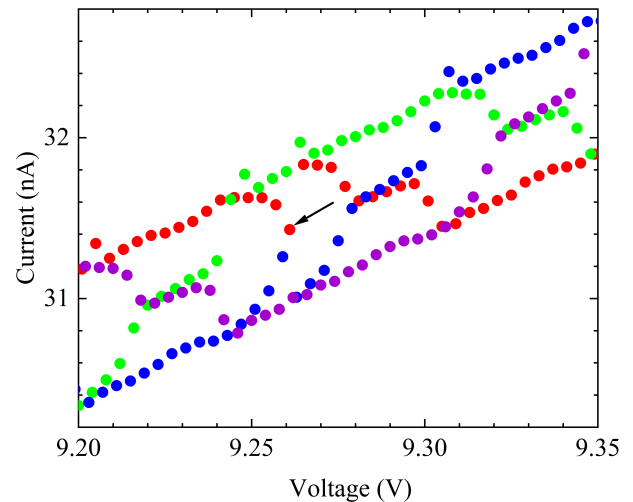


FIG. 8. The shifted pattern starts at the "off pattern" red point indicated by the arrow, which normally would have been located at the upper current extreme. After this the shifted pattern continues as before towards lower voltages.

trolled environment. Alternatively, the information diffusion rate may be locked to the measurement-frequency. To increase our knowledge of the pattern, we will need more variation in the measurement-frequency to study the resulting response from the enclosed quantum system. In addition, more variation in τ_m is required to study the transition from coherence to decoherence. At this stage, the exact nature of the pattern remains unexplained.

In the main graph from Fig. 5 a distinct black and white pattern repeats itself over the entire curve. In the lower curve, however close at 9.25 V, it shows that the pattern is broken. Two white spots are in close proximity; this section is enlarged in Fig. 8. We found that the effect is a "glitch" at one point, see arrow, after which the regular pattern continues as before. Thus, this relates to a phase shift of the entire pattern. We speculate that the pattern may be "hard-coded" in the quantum system measurement combination because of the interplay between the measurement-frequency and the information diffusion rate, surviving a glitch with a resulting pattern shift.

The molecular MCB junction in the partially wet phase provides for a special type of open quantum system, the enclosed open quantum system. In this type of device, information flows in both directions between the quantum system and the controlled environment. Typical decoherence times are in the 1 ms to 20+ ms range at ambient conditions. With the measurement time we have a powerful knob, enabling adjustment in relation to the decoherence time of the system, facilitating a study of the transition from coherence to decoherence at ambient conditions.

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