Observation of Wave-Packet Branching through an Engineered Conical Intersection

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Analog quantum simulators, which efficiently represent model systems, have the potential to provide new insight toward naturally occurring phenomena beyond the capabilities of classical computers. Incorporating dissipation as a resource unlocks a wider range of out-of-equilibrium processes such as chemical reactions. Here, we operate a hybrid qubit-oscillator circuit quantum electrodynamics simulator and model nonadiabatic molecular dynamics through a conical intersection. We identify dephasing of the electronic qubit as the mechanism that drives wave-packet branching when the corresponding oscillator undergoes large amplitude motion. Furthermore, we directly observe enhanced branching when the wave-packet passes through the conical intersection. Thus, the forces that influence a chemical reaction can be viewed from the perspective of measurement backaction in quantum mechanics—there is an effective measurement-induced dephasing rate that depends on the position of the wave packet relative to the conical intersection. Our results set the groundwork for more complex simulations of chemical dynamics using quantum simulators, offering deeper insight into the role of dissipation in determining macroscopic quantities of interest such as the quantum yield of a chemical reaction.

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I. INTRODUCTION

Quantum processors applied to quantum chemistry have mostly focused on calculating static electronic energies [1], though experimental demonstrations have been limited to variational methods in the current near term intermediate-scale quantum era of devices [2,3]. Modeling molecular dynamics that ensue from interaction with light, however, is a driven-dissipative many-body problem that goes beyond just calculating the fermionic electronic and includes the dynamics of bosonic rotational and vibrational degrees of freedom as well. Electronic transport dynamics have been investigated using quantum processors in the context of light harvesting in photosynthesis, where individual sites are approximated as two-level systems and encoded in qubits [4,5]. Vibrational dynamics and vibronic spectra have also been simulated using bosonic modes that can support multiphoton states [6–8], but only under the adiabatic Born-Oppenheimer approximation.

In many polyatomic molecules, nonadiabatic couplings between potential energy surfaces influence a wide range of photoinduced chemical processes [9]. Central to nonadiabatic dynamics are features known as conical intersections (CIs), which indicate an electronic degeneracy that invalidates the Born-Oppenheimer approximation and results in strong hybridization between the electronic and nuclear degrees of freedom. This hybridization has immense consequences for excited state dynamics when the molecular wave packet traverses these intersections, producing, for example, ultrafast radiationless reactions. Using quantum simulators to study nonadiabatic dynamics has been proposed with digital quantum computers [10] and trapped-ion systems [11–13], but experimental demonstrations have remained elusive.

In this work, we experimentally demonstrate dissipative nonadiabatic dynamics using a circuit quantum electrodynamics (circuit QED) quantum simulator. Using microwave drives, we engineer a CI in a system containing one qubit and two harmonic oscillators. The qubit represents two electronic energy levels that are modulated by two generalized nuclear coordinates encoded in harmonic oscillators.

Our approach uses a simple and optimal one-to-one mapping of the three modes of the model to three modes of our simulator; this native mapping enables the approach’s hardware efficiency and can be straightforwardly scaled up using existing multimode systems [14,15]. Importantly, we engineer intrinsic qubit coherence times an order of magnitude longer than the timescale of the
engineered dissipation. This corresponds to the physical scenario where the dynamics are not dominated by spontaneous emission of the electronic excitation. By simultaneously measuring the qubit and performing Wigner tomography [16] of one of the oscillators as they evolve under a control Hamiltonian, we correlate qubit dephasing events with wave-packet branching. Finally, when the CI is active, we observe enhanced branching when the wave packet passes through the CI. This can be understood as a competition between the measurement-induced dephasing strength and a position-dependent electronic energy gap. Our results demonstrate that superconducting circuits possess all of the capabilities required to implement faithful simulations of nonadiabatic dynamics.

II. MODEL CONICAL INTERSECTION HAMILTONIAN

The variety of numerical methods and associated models developed for addressing nonadiabatic molecular dynamics on classical computers is vast [17–19]. Common to all of these methods are various approximations which reduce the computational cost compared to exact solutions, which of course scale exponentially with system size. For quantum simulation of nonadiabatic problems, we seek models that treat all degrees of freedom quantum mechanically; this effectively translates to a basis choice. Vibronic coupling (VC) model Hamiltonians [20] use a diabatic basis for the electronic eigenfunctions in order to avoid divergence issues associated with the standard adiabatic basis in the vicinity of a CI [21]. See Appendix A for a more detailed discussion of the choice of basis. VC Hamiltonian parameters may be either empirically fit to reproduce experimental data [22] or obtained from ab initio calculations such as for pyrazine [23] and pyrene [24]. Computational studies investigating the influence of various forms of rovibrational damping on nonadiabatic dynamics utilize VC Hamiltonians with different approximations for the bath [25–27].

In this work, we consider a linear vibronic coupling (LVC) model—the simplest VC Hamiltonian that contains a conical intersection—with two electronic states $|\pm\rangle$ coupled to two generalized rovibrational modes $\hat{a}$ and $\hat{b}$:

\[
\hat{H}_a = \frac{\hbar}{2} g_a (\hat{a}^\dagger \hat{a} + \sigma_x (\hat{a} + \hat{a}^\dagger)),
\]

\[
\hat{H}_b = \frac{\hbar}{2} g_b (\hat{b}^\dagger \hat{b} + \sigma_y (\hat{b} + \hat{b}^\dagger)),
\]

FIG. 1. Model molecular Hamiltonian with a conical intersection and experimental setup. (a) Eigenspectrum of $\hat{H}_a$, which consists of two displaced quantum harmonic oscillator Fock ladders (dashed lines) of the tuning mode depending on if the qubit is in $|+\rangle$ (blue) or $|-\rangle$ (red). (b) In the basis of $|\pm\rangle$ for the qubit, the addition of $g_b \sigma_y (\hat{b} + \hat{b}^\dagger)$ on top of $\hat{H}_a$ couples the two Fock ladders of the coupling mode (with energy $\Delta_b \hat{b}^\dagger \hat{b}$) via $\sigma_y = |+\rangle \langle -| + |-\rangle \langle +|$ (dashed arrows) for each Fock state of the tuning mode. Hence, dynamics of the coupling mode can cause transitions between $|\pm\rangle$. (c) Schematic of the circuit QED device used in this experiment. The tuning and coupling modes are 3D $\lambda/4$ coaxial resonators which are coupled to an electronic qubit (encoded in a transmon) with a readout resonator and Purcell filter [31]. An additional ancilla module with a transmon mode and readout resonator is coupled to the tuning mode for independent state tomography. The coupling mode is intentionally overcoupled to a 50 $\Omega$ transmission line, resulting in a linewidth $\kappa_b = 2\pi \approx 320$ kHz. (d) Relative mode frequencies and drive configuration used to enact $\hat{H}_{LVC}$. A full table of system parameters is given in Appendix C.
\[ \hat{H}_{\text{LVC}} = \hat{H}_a + \hat{H}_b, \]

where our electronic states \( |\pm\rangle \) are eigenstates of \( \hat{\sigma}_z \). Our model contains first-order intrastate and interstate coupling strengths \( g_a \) and \( g_b \), respectively, as well as generalized rovibrational frequencies \( \Delta_a \) and \( \Delta_b \). We refer to modes \( \hat{a} \) and \( \hat{b} \) as tuning and coupling modes, respectively, as the dimensionless coordinate \( \hat{q}_t = \hat{a} + \hat{a}^\dagger \) “tunes” the energy between the electronic states \( |\pm\rangle \) and \( \hat{q}_c = \hat{b} + \hat{b}^\dagger \) mediates coupling between the two electronic states via \( \hat{\sigma}_x \) as the states of \( \hat{q}_c \) couples the electronic states via the states of \( \hat{q}_t \) when expressed in the driven eigenbasis spanned by \{\( \hat{g} \), \( \hat{\bar{e}} \)\} that adiabatically connects to the undriven transmon eigenstates \{\( |\hat{g}\rangle \), \( |\hat{e}\rangle \)\}. We refer to modes \( \hat{q}_t \) and \( \hat{q}_c \) as tuning and coupling modes, respectively, as the dimensionless coordinate \( \hat{q}_t = \hat{a} + \hat{a}^\dagger \) “tunes” the energy between the electronic states \( |\pm\rangle \) and \( \hat{q}_c = \hat{b} + \hat{b}^\dagger \) mediates coupling between the two electronic states via \( \hat{\sigma}_x \) as the states of \( \hat{q}_c \) couples the electronic states via the states of \( \hat{q}_t \) when expressed in the driven eigenbasis spanned by \{\( \hat{g} \), \( \hat{\bar{e}} \)\} that adiabatically connects to the undriven transmon eigenstates \{\( |\hat{g}\rangle \), \( |\hat{e}\rangle \)\}. We note that \( |\hat{g}\rangle \approx (1/\sqrt{2})(|\hat{e}\rangle + |\hat{\bar{e}}\rangle) \) and \( |\hat{\bar{e}}\rangle \approx (1/\sqrt{2})(|\hat{g}\rangle - |\hat{e}\rangle) \), with minor contributions from higher excited states, when expressed in the basis of undriven transmon eigenstates. The conditional displacement interactions that we engineer in our experiment \{\( \hat{H}_a \), \( \hat{H}_b \)\} are conditioned on Pauli operators whose eigenstates lie on the equator of the driven Bloch sphere (i.e., \( \hat{\sigma}_x \) and \( \hat{\sigma}_y \) of the Rabi drive and adiabatically prepare the driven qubit eigenstates. This enables large Rabi frequencies while suppressing leakage to higher levels of the driven transmon (see Appendix B).

A. Simulator calibrations

A key requirement of our quantum simulator is the ability to initialize and perform tomography of the electronic qubit, which in our experiment is encoded by a driven transmon with an effective Rabi frequency \( \Omega_R \) that is defined by the amplitude \( \epsilon_R \) and static detuning \( \Delta_R \) of the Rabi drive. In the frame of a single Rabi drive on the transmon, the driven Hamiltonian has the form \( \hat{H}_d = (\Omega_R/2)\hat{\sigma}_x \) where \( \hat{\sigma}_x = |\hat{g}\rangle \langle \hat{g}| - |\hat{\bar{e}}\rangle \langle \hat{\bar{e}}| \) when expressed in the driven eigenbasis spanned by \{\( |\hat{g}\rangle \), \( |\hat{\bar{e}}\rangle \)\} that adiabatically connects to the undriven transmon eigenstates \{\( |\hat{g}\rangle \), \( |\hat{e}\rangle \)\}. We note that \( |\hat{g}\rangle \approx (1/\sqrt{2})(|\hat{e}\rangle + |\hat{\bar{e}}\rangle) \) and \( |\hat{\bar{e}}\rangle \approx (1/\sqrt{2})(|\hat{g}\rangle - |\hat{e}\rangle) \), with minor contributions from higher excited states, when expressed in the basis of undriven transmon eigenstates. The conditional displacement interactions that we engineer in our experiment \{\( \hat{H}_a \), \( \hat{H}_b \)\} are conditioned on Pauli operators whose eigenstates lie on the equator of the driven Bloch sphere (i.e., \( \hat{\sigma}_x \) and \( \hat{\sigma}_y \), and thus will precess around the equator at a rate \( \Omega_R \). We can use this high frequency precession as a way of precisely calibrating \( \Omega_R \), which in turn lets us measure the expectation value of any Pauli operator aligned on the equator of the driven Bloch sphere over time. As long as the Rabi frequency is constant as a function of time, this technique is compatible with any interaction Hamiltonian that involves our driven qubit including \( \hat{H}_a \), \( \hat{H}_b \), and \( \hat{H}_{\text{LVC}} \). For the free evolution case where only the Rabi drive is on and no cavity sidebands are active, we measure \( \langle \hat{\sigma}_z \rangle \) as a function of time and extract a corresponding driven coherence time \( T_{2p} = 27 \mu s \) [34]. This further elucidates the reasoning behind our chosen convention for the Pauli operators in Eqs. (1) and (2); we prefer the eigenstates of \( \hat{\sigma}_x \) to be stationary in the frame of the drive. Full details of this measurement scheme are described in Appendix D.

The goal of our experiment is to understand the dynamics of the subsystem consisting of the electronic qubit and tuning mode under the influence of the conical intersection and dissipation in the coupling mode. The symmetry between the two interactions \( \hat{H}_a \) and \( \hat{H}_b \) is broken by their disparate dissipation rates, which results in qualitatively different dynamics. Specifically, we work in the
coherent regime where \( g_s \gg \kappa_s \) for the tuning mode and the dissipative regime \( g_s \ll \kappa_s \) for the coupling mode. As such, we proceed to calibrate and characterize each interaction independently before combining them together to study the full Hamiltonian \( \hat{H}_{LVC} \) of interest that includes the conical intersection.

We first consider the conditional displacement interaction \( \hat{H}_u \) on the tuning mode, which determines how coherent wave packets in the tuning mode propagate for each electronic state in the absence of the coupling mode. For clarity, we will describe coherent wave packets in the tuning mode as reactive wave packets following our earlier designation that the position of the tuning mode is a reactive coordinate of interest in a molecular system that undergoes large amplitude motion. This Hamiltonian produces two distinct harmonic potential energy surfaces whose equilibrium positions are offset from the origin by an amount \( \pm \alpha_g \), where \( \alpha_g = g_s / \Delta_g \) for each electronic state \( |\mp\rangle \). Moreover, we are interested in the regime where \( |\alpha_g| \gtrsim 1 \) such that there are two macroscopically distinct ground states. Reactive wave packets that are prepared with the qubit in \( |\pm\rangle \) at any location in phase space should oscillate around the respective ground state [Fig. 2(a)].

We demonstrate this first for an initial vacuum state, which is a displaced state with respect to the minimum of each of the driven potentials. We initialize the driven qubit in \( |\rightarrow\rangle \) and then ramp on two sideband drives quickly with respect to \( 1/\Omega_R \) with the appropriate phases (see Appendix D). By adjusting the average of the sideband frequencies relative to the Stark-shifted cavity frequency, we tune \( \Delta_c \), taking care to satisfy the resonance conditions at each frequency configuration. We observe the resulting dynamics by measuring the projection onto the vacuum state as a function of time using a photon-number-selective \( \pi \) pulse [35] on a separate transmon ancilla [Fig. 2(b)]. At the same time, we measure \( \langle \hat{\sigma}_x \rangle \) of the qubit using our previously described measurement scheme and observe that it largely remains in its initial eigenstate \( |\rightarrow\rangle \) up to decoherence.

In the context of the LVC model, we have \( \hat{\sigma}_x = 2(\hat{P}_+ - 1) \) and \( \hat{\sigma}_y = -2(\hat{P}_+ - 1) \), where \( \hat{P}_\pm = |\pm\rangle \langle \pm| \) are projectors onto the diabatic states and \( \langle \hat{P}_\pm \rangle \) are the corresponding diabatic populations. This demonstrates that we are able to

![Figure 2](image)

**FIG. 2.** Calibrating coherent and dissipative conditional displacement interactions on the tuning mode (a), (b) and the coupling mode (c). (a) Depiction of tuning mode dynamics under \( \hat{H}_u \). The oscillator’s equilibrium position shifts by \( +\alpha_g \) when the qubit is in \( |\rightarrow\rangle \) (red) and \( -\alpha_g \) when the qubit is in \( |\leftarrow\rangle \) (blue). Top: an initial wave packet \( |\rightarrow\rangle \otimes |\alpha = 0\rangle \) oscillates along the red potential surface associated with \( |\rightarrow\rangle \). Bottom: in phase space, the state performs a circular trajectory around the equilibrium location, indicated by the red triangle for the top schematic. Smaller and larger equilibrium positions are indicated by the circle and square, respectively. (b) Coherent state revivals during evolution under \( \hat{H}_u \). We prepare an initial state \( |\rightarrow\rangle \otimes |\alpha = 0\rangle \) and simultaneously measure the vacuum projector of the tuning mode \( \hat{P}_\alpha = |0\rangle \langle 0| \) (top) and \( \hat{\sigma}_x \) (bottom) for different calibrated values of \( \Delta_c \). Solid lines are fits to a simple analytic model, extracting values \( g_s / 2\pi = 450 \text{ kHz} \), \( \Delta_c / 2\pi = 457 \text{ kHz} \) (circles), \( 355 \text{ kHz} \) (triangles), and \( 246 \text{ kHz} \) (squares) which determine the values of \( \alpha_g \). Approximate locations of the coherent state for each calibration at a delay time marked by the magenta arrow are shown in the bottom panel of (a). At the same time, the qubit largely remains in \( |\rightarrow\rangle \) up to intrinsic decoherence. (c) Top: coupling mode state trajectories starting in vacuum \( |0\rangle \) for various values of \( \Delta_b \) (rates from the bottom panel) with fixed values of \( \hat{g}_b \) and \( \kappa_b \), splitting in phase space depending on if the qubit is in \( |\pm\rangle \). Full coherent state distributions are not depicted because we are in the weak measurement regime where there is significant overlap between the coherent states. Bottom: measurement-induced dephasing of \( |\rightarrow\rangle \) with \( g_s / 2\pi \approx 117 \text{ kHz} \), \( \Delta_b / 2\pi \approx 0 \text{ (circles), 400 (triangles), 800 (squares)} \) kHz and \( \kappa_b / 2\pi \approx 320 \text{ kHz} \) obtained from independent calibrations. Solid lines represent equivalent time-domain master equation simulation results (see Appendix F). All error bars indicate standard deviations and may not be visible compared to marker size.
engineer $\hat{H}_a$ with in situ control of the Hamiltonian parameters.

Next, we independently calibrate $\hat{H}_b$ and its dissipative dynamics in the absence of coupling to the tuning mode. From a quantum optics perspective, the combination of a conditional displacement interaction $\hat{H}_b$ and single photon loss with rate $\kappa_b/2\pi \approx 320$ kHz results in measurement-induced dephasing of the qubit along the axes orthogonal to the interaction [36,37]. We verify this behavior by preparing an eigenstate of $\hat{\sigma}_x$, tuning the qubit axis of the interaction to be $\hat{\sigma}_y$ by adjusting the relative phase of the sidebands on the coupling mode (see Appendix B), and measuring $\langle \hat{\sigma}_y \rangle$ as a function of time [Fig. 2(c)]. The measurements reveal dephasing rates that are suppressed when $\Delta_\beta$ increases, as expected. Similar to the interaction on the tuning mode, the conditional displacement strength $g_f$ and detuning $\Delta_b$ can be controlled in situ. Ultimately, this yields a tunable measurement-induced $\hat{\sigma}_y$ dephasing with a finite bandwidth, which we will proceed to activate in conjunction with the coherent interaction on the tuning mode. Full experimental calibration details for both interactions $\{\hat{H}_a, \hat{H}_b\}$ are provided in Appendix D.

**B. Demonstration of wave-packet branching**

Before combining the two conditional displacement interactions to enact $\hat{H}_{\text{LVC}}$, we first perform a control experiment to observe branching of a reactive wave packet due to the intrinsic noise of our quantum simulator. Under $\hat{H}_a$, where the eigenstates of $\hat{\sigma}_x$ determine the two potential surfaces, we define branching of a reactive wave packet to be a process (either coherent or noisy) that flips $|+\rangle \leftrightarrow |-\rangle$, which causes the wave packet to branch to the opposite potential surface [Fig. 3(a)]. Note that this process bears resemblance to trajectory-based surface hopping algorithms [38,39], except that in those algorithms nuclei are treated classically. We can probe this effect, while also simultaneously verifying that we have control over two simultaneous conditional displacement interactions, by enacting the following control Hamiltonian:

$$\hat{H}_x = \Delta_\alpha \hat{a}^\dagger \hat{a} + \Delta_\beta \hat{b}^\dagger \hat{b} + \hat{\sigma}_x [g_f (\hat{a} + \hat{a}^\dagger) + g_b (\hat{b} + \hat{b}^\dagger)].$$

We engineer $\hat{H}_x$ by adjusting the sideband drive phases to align the interaction of both modes to $\hat{\sigma}_x$ (see Appendix B). In this scenario, the eigenstates of $\hat{H}_x$ are product states in the uncoupled basis $\{|\pm\rangle \otimes \hat{D}(\pm \alpha_g) |n\rangle_a \otimes \hat{D}(\pm \beta_g) |n\rangle_b\}$, where dissipation modifies the value of $\beta_g = g_f/\sqrt{\Delta_\beta^2 + k_b^2}/4$ and $\{|n\rangle_{a/b}\}$ are Fock state ladders of each mode. As such, any measurement-induced dephasing from the coupling mode should not perturb the dynamics of a reactive wave packet since they do not cause transitions between $|\pm\rangle$. Thus, our previous description of branching remains valid in the case of white noise and we do not have to consider the dynamics of the coupling mode.

For our control experiment, we enact $\hat{H}_x$ with calibrated parameters $g_f/2\pi = 410$ kHz, $\Delta_\alpha/2\pi = 324$ kHz, $g_b/2\pi \approx 156$ kHz, and $\Delta_\beta/2\pi \approx 0$ kHz. We prepare a ground state of the tuning mode $|\cdots\rangle \otimes |\underbar{0}\rangle_\alpha \otimes |\underbar{0}\rangle_\beta$ (see Appendix D) and let the system evolve under $\hat{H}_x$ for a delay time $\tau = 10$ µs that is short compared to the intrinsic coherence time of the qubit $\tau < T_{2p}^\alpha$ (here we label the decoherence with a superscript x to indicate it is measured with $\hat{H}_x$ active). Next, we simultaneously measure the qubit in the $\hat{\sigma}_x$ basis and perform Wigner tomography on the cavity using the ancilla transmon [16]. In the language of
quantum dynamics, the Wigner distribution is function in a phase space that contains full information about the quantum state. The marginal distributions represent wave-packet probability distributions of the tuning mode along position and momentum, which in our case is represented by the real and imaginary components of the electromagnetic field \{Re(\alpha), Im(\alpha)\}. Correlating the tomography results with the qubit measurement outcome reveals that the wave packet remains coherent if the qubit did not experience a dephasing event, i.e., stays in \ket{\alpha}, or fully dephases otherwise if environmental noise caused a flip from \ket{-} to \ket{+} [Fig. 3(b)]. Because the time of each individual dephasing event is unknown to the experimentalist, the oscillator state will be in a uniformly distributed mixed state with constant total energy in the displaced frame (reminiscent of a donut in phase space centered around the opposite ground state). Thus, we have verified that wave-packet branching indeed occurs alongside qubit dephasing in our quantum simulator, and that we are able to faithfully enact two conditional displacement Hamiltonians simultaneously.

C. Dissipative dynamics through a conical intersection

The wave-packet branching observed under \(\hat{H}_x\) and a white noise dissipator \((\gamma_{sp}^2/2) \times \hat{D}(\sigma_j)\hat{\rho}\), where \(2\pi \times \gamma_{sp} = 1/T_{sp}^3\), largely does not depend on the location of the reactive wave packet, and thus on the corresponding energy gap \(\propto \hat{\sigma}_z\). Under \(\hat{H}_\text{LVC}\) and the conical intersection, this is no longer the case. To demonstrate this, we activate \(\hat{H}_\text{LVC}\) with calibrated parameters \(g_\text{c}/2\pi = 158.0\ kHz\), \(\Delta_\text{a}/2\pi = 125.8\ kHz\), \(g_\text{c}/2\pi = 115\ kHz\), and \(\Delta_\text{b}/2\pi \approx 0\ kHz\). With our confirmed understanding that qubit dephasing along \(\hat{\sigma}_x\) drives wave-packet branching, we prepare the system with reactive wave packets at different locations \((-\ket{\alpha}_a) \otimes \ket{\alpha}_{j,a}, \ket{2\alpha}_{j,a} \otimes \ket{0}_b\) and directly monitor \(\langle \hat{\sigma}_x \rangle\) as a function of time [Fig. 4(a)]. We observe that the qubit dephasing is both highly nonexponential and stronger upon passage of the reactive wave packet through the intersection. Additionally, we further verify that this behavior indeed correlates with dephasing of the cavity state by taking Wigner functions of an initially displaced wave packet before and after passage through the conical intersection. In this instance, we reduce the interaction strengths of the reactive potential surface to get a clear signature of branching over the course of one period of motion, and choose zero detuning on the coupling mode’s conditional displacement to achieve the strongest dephasing, representing a very slow intranuclear mode.

In order to qualitatively understand the dissipative dynamics that we observe, we can imagine treating the tuning mode classically. Here, we are left with a simplified Hamiltonian,

\[
\hat{H}_\text{LVC} \propto \begin{array}{c}
\langle \hat{\sigma}_x \rangle \\
\text{Delay time } \tau \text{ (\mu s)}
\end{array},
\]

\[
\begin{array}{c}
\text{Re}(\alpha)
\end{array}
\]

\[
\begin{array}{c}
\text{Im}(\alpha)
\end{array},
\]

\[
\begin{array}{c}
\text{Parity}
\end{array}
\]

\[
\Gamma_\text{meas} \gg E, \Gamma_\text{meas} \approx E, \Gamma_\text{meas} \ll E
\]

FIG. 4. Branching through a conical intersection. (a) Top: measured expectation value of \(\hat{\sigma}_x\) with standard error bars over time for the three different initial states \(-\ket{\alpha}_a \otimes \ket{\alpha}_{j,a}, \ket{2\alpha}_{j,a} \otimes \ket{0}_b\), where \(\alpha = g_\text{c}/\Delta_\text{a} \approx 1.26\). The magenta wave packet prepared at the CI immediately dephases, whereas the other two dephase more slowly, as they are farther away. After half of an oscillation period \(\tau \sim 1/(2\Delta_\text{a})\), the orange wave packet arrives at the CI and dephases. Solid lines are predictions from a master equation simulation using independently fitted parameters. Dashed lines represent the negligible background decoherence due to \(T_{sp}^3\) on the timescale of the interaction and dissipation. Bottom: unconditional Wigner tomography on the tuning mode at \(\tau = 2 \text{ \mu s}\) (left) and 6 \(\mu\) s (right) for preparing \(\ket{\alpha}_a \otimes \ket{2\alpha}_{j,a}\), revealing a coherent wave packet before and dephased state after passage through the CI. The distortion of the Wigner function from a Gaussian at \(\tau = 2 \text{ \mu s}\) suggests the presence of a residual self-Kerr nonlinearity in the oscillator. (b) A semiclassical interpretation of the dephasing versus location of the three initial wave packets prepared in (a). The potential surfaces are shown in dashed lines to emphasize that they are no longer exact in the diagonalized basis of \(\hat{H}_\text{LVC}\). The dissipative dynamics arises from a competition between the \(\hat{\sigma}_x\) energy gap \(E\) with the finite strength of the dephasing \(\Gamma_\text{meas}\).
\[ \hat{H}_{\text{Zeno}}/\hbar = E(x)\hat{\sigma}_x + \Delta_b\hat{b}^\dagger\hat{b} + g_x\hat{\sigma}_x(\hat{b} + \hat{b}^\dagger), \]

subject to single photon loss on mode \( \hat{b} \) at a rate \( \kappa_b \). The function \( E(x) \) can be interpreted as the position-dependent energy gap (in frequency units) between qubit states for a conditional displacement interaction. At the CI, i.e., at energy gap (in frequency units) between qubit states for a particular in the regime \( E(x) \approx 0 \), this energy gap vanishes and we are left with the environment measuring \( \hat{\sigma}_x \), with a measurement strength \( \Gamma_{\text{meas}} = g^2_k/\kappa_b/2 + \Delta_b^2/\kappa_b^2 \) in the steady state where \( g_x = \kappa_b \). Away from the origin, the qubit has a finite energy along an axis orthogonal to that of the measurement and we recover a scenario reminiscent of Zeno dynamics of a driven qubit [41]. This reduces the effective measurement strength, resulting in slower decoherence and thus reduced branching events [Fig. 4(b)]. As such, a semiclassical approximation would correspond to choosing a time-dependent trajectory \( x(t) \) for an initial Gaussian wave packet. Interestingly enough, this model contains regions in parameter space where the steady-state qubit polarization is not 0.5 as one may naively expect. A full in-depth analysis is provided in Appendix G. In our full model, wave packets in the tuning mode will diffuse in phase space due to the branching, resulting in dynamics that are quantitatively different from the above simplified model.

\section*{IV. OUTLOOK}

Modifications to our experimental setup can be made to expand the scope of dynamics in our LVC model. For instance, by overcoupling the tuning mode to a transmission line, the reactive coordinate will experience dissipation which would eventually localize an initially excited wave packet into the two ground states and define a branching ratio. The addition of \( \kappa_a \) as a tuning knob, particularly in the regime \( \kappa_a \sim \kappa_b \), expands the landscape of competing forces in our model and represents a more realistic description of a reaction. We note the possibility of tuning the decay rate of each oscillator \textit{in situ} via mechanical means in our 3D architecture, which would enable a flexible way to explore the wider range of parameter space.

On a broader scale, it will be necessary to both scale up our simulator and add complexity to our model in order to simulate larger molecules and challenge classical algorithms. As described in Appendix B, our method of enacting \( \hat{H}_{\text{LVC}} \) can easily be extended to existing multimode systems where a large number of cavity modes (\( \sim 10 \)) are dispersively coupled to a single transmon, such as in Refs. [14,15].

Expanding from the LVC model to include higher order couplings, however, will require the development of new control techniques that have yet to be proposed for any system. This would allow for a more accurate modeling of realistic systems, whose dynamics are heavily influenced by nuclear topography. The flexibility of engineering the Josephson potential to enact mixing processes of various orders poses promise for this task [42,43]. Furthermore, systems that have Franck-Condon regions far from the CI will translate to larger photon numbers in our simulator. As such, precisely controlling the desired nonlinear reaction Hamiltonian over the domain of larger photon numbers will be a central challenge to address in future experiments.

Beyond quantum chemistry, the successful implementation of our quantum simulator motivates the investigation of novel cavity QED physics [44,45]. The primary advantage of our approach is the \textit{in situ} tunability of the constituent interactions in a quantum simulator to explore system dynamics in various parameter regimes.

\section*{V. CONCLUSION}

In summary, our results highlight the interplay between coherent evolution and engineered dissipation in a system whose energy landscape contains a CI. We achieve this via careful Hamiltonian engineering of a circuit QED processor involving five simultaneous microwave drives and engineered dissipation, along with the appropriate state preparation and measurement protocols to observe branching dynamics. In particular, branching events arise when dissipation in the coupling mode induces flips on the electronic state and correspondingly causes the reactive wave packet to jump onto the opposite potential surface. These branching events are at the heart of chemical reaction dynamics, such as the \textit{cis-trans} isomerization reaction of rhodopsin [46], a central process in human vision, and occur more frequently upon passage through the CI. Our experiment demonstrates and further confirms the immense flexibility of this platform to perform quantum information processing tasks by dressing microwave modes with continuous drives [33,47,48], and constitutes an important step toward investigating more complex chemical phenomena with higher degrees of accuracy. It is worth noting that the techniques developed in our work may readily be applicable to control multimode bosonic systems coupled to one or a few qubits [15,49]. Looking ahead, incorporating additional features into our simulator such as additional controlled nonlinearities and structured dissipators, while scaling up the number of electronic states and rovibrational modes, will enable more accurate simulation of larger molecules. More broadly, this expands the landscape of controllable qubit-oscillator interactions in a circuit QED platform, which may be useful for bosonic quantum computation, error correction, and simulation.

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C. S. W., M. H. D., and R. J. S. conceived the experiment. C. S. W. and N. E. F. performed early stage experimental design. C. S. W. and B. J. C. fabricated the qubits and assembled the device. C. S. W. developed the experimental protocol, performed measurements, and carried out numerical simulations with N. E. F., B. J. C., and R. J. S providing input. S. P. and S. M. G. provided theoretical support. C. S. W. and R. J. S. wrote the paper with feedback from all authors.

APPENDIX A: CONSTRUCTING A DIABATIC HAMILTONIAN CONTAINING A CONICAL INTERSECTION

In this appendix, we describe the general formalisms behind obtaining various model and/or ab initio molecular Hamiltonians that involve strong vibronic coupling. We begin with a brief review of adiabatic potential energy surfaces and highlight the difficulties that arise in the vicinity of conical intersections. This motivates the use of diabatic electronic states, which form the basis of the Hamiltonians that we consider [50].

The standard molecular Hamiltonian is

$$\hat{H}_{\text{mol}} = \hat{T}(\vec{r}) + \hat{V}(\vec{r}, \vec{R}) + \hat{e},$$  \hspace{1cm} (A1)

where $\hat{T}$ and $\hat{V}$ correspond to kinetic and Coulomb potential energies, respectively, and the subscripts $n$ and $e$ denote nuclei and electrons. $\vec{r}$ and $\vec{R}$ represent the positions of the electrons and nuclei, respectively.

The conventional approach based on the Born-Oppenheimer approximation begins with noting that the electron masses $m_e$ are much smaller than the nuclear masses $m_N$. This motivates momentarily dropping $\hat{T}_n(\vec{r}) \propto 1/m_N$ altogether, leaving behind a reduced Hamiltonian of the electrons only with parametric dependence on the nuclear coordinates $\vec{R}$. By choosing an ansatz for the total molecular wave function $\Psi(\vec{r}, \vec{R}) = \sum \phi_l(\vec{r})\chi_l(\vec{R})$, we get a reduced electronic Schrodinger equation:

$$\hat{H}_e \phi_l(\vec{r}, \vec{R}) = E_l(\vec{R})\phi_l(\vec{r}, \vec{R}),$$  \hspace{1cm} (A2)

where $E_l(\vec{R})$ is the potential energy surface for the $l$th electronic eigenstate. Here, we have identified a complete set of diabatic electronic eigenfunctions $\phi_l(\vec{r}, \vec{R})$. Returning to the full Schrödinger equation,

$$\hat{H}_{\text{mol}}\Psi(\vec{r}, \vec{R}) = [\hat{T}_n(\partial_{\vec{r}}) + \hat{H}_e] \sum \phi_l(\vec{r}, \vec{R})\chi_l(\vec{R}),$$  \hspace{1cm} (A3)

we can obtain a reduced equation for the nuclear motion by integrating over a complete set of adiabatic electronic eigenfunctions $\phi^*_l(\vec{r}, \vec{R})$. This brings out terms such as

$$\langle \phi_j(\vec{r}, \vec{R})|\nabla_{\vec{R}}\hat{H}_e|\phi_l(\vec{r}, \vec{R})\rangle \over E_j(\vec{R}) - E_l(\vec{R}),$$  \hspace{1cm} (A4)

which arise from the application of $\hat{T}_n(\partial_{\vec{r}}) \propto -(1/2m_N)\nabla^2_{\vec{R}}$ on $\phi_l(\vec{r}, \vec{R})$. These are commonly referred to as nonadiabatic coupling terms in the literature. It is clear that in the vicinity of a conical intersection, these terms diverge as the denominator becomes very small and the adiabatic electronic basis fails to be an appropriate basis for calculations and analyses.

Given the aforementioned issue, one can consider instead a diabatic electronic basis $\psi_{\alpha}(\vec{r})$ such that the molecular wave function can be expressed as

$$\Psi(\vec{r}, \vec{R}) = \sum_k \psi_{\alpha}(\vec{r})\chi_{\alpha}(\vec{R}),$$  \hspace{1cm} (A5)

where the diabatic states are, by definition, diagonal in the nuclear kinetic energy operator. Off-diagonal couplings between electronic states must of course exist, but now they arise via the potential $\langle \phi_j|\hat{V}(\vec{r}, \vec{R})|\phi_l\rangle$ and do not involve wave-function derivatives.

This forms the basis for a general vibronic coupling Hamiltonian:

$$\hat{H}_{\text{VC}} = \sum_n |\phi_n\rangle [\hat{T}_n + W_{nn}(\vec{R})]|\phi_n\rangle + \sum_{n\neq m} |\phi_n\rangle W_{nm}(\vec{R})|\phi_m\rangle.$$  \hspace{1cm} (A6)

In our experiment, we consider a minimal model for a two-dimensional linear vibronic coupling Hamiltonian, where we have for two normal modes:

$$\hat{T}_n = {\hat{p}_n^2 \over 2m_t} + {\hat{p}_e^2 \over 2m_e},$$  \hspace{1cm} (A7)

$$\hat{W} = \left( \begin{array}{cc} \sum_{i=l,e} \frac{1}{2} m_i \Delta_i^2 \hat{Q}_i^2 + g_e \hat{q}_e & g_e \hat{q}_e \\ g_e \hat{q}_e & \sum_{i=l,e} \frac{1}{2} m_i \Delta_i^2 \hat{Q}_i^2 - g_e \hat{q}_e \end{array} \right).$$  \hspace{1cm} (A8)
with momentum \( \hat{P}_{t/c} \) and position \( \hat{Q}_{t/c} \) with effective masses \( m_{t/c} \) and frequencies \( \Delta_{t/c} \), respectively. By recasting these coordinates into dimensionless creation and annihilation operators,

\[
\hat{P}_{t/c} = p_{ZPF} \hat{P}_{t/c}, \quad (A9)
\]

\[
\hat{Q}_{t/c} = q_{ZPF} \hat{Q}_{t/c}, \quad (A10)
\]

where \( \hat{p}_t = i(\hat{a}^\dagger - \hat{a}) \), \( \hat{p}_c = i(\hat{b}^\dagger - \hat{b}) \), \( \hat{q}_t = \hat{a} + \hat{a}^\dagger \), \( \hat{q}_c = \hat{b} + \hat{b}^\dagger \), and \( q_{ZPF} = \sqrt{\hbar/2m_{t/c}} \), \( p_{ZPF} = \sqrt{\hbar m_{t/c}/2} \) for \( i \in \{t, c\} \), we arrive at \( \hat{H}_{LVC} \) [Eq. (3) of the main text].

**APPENDIX B: ENGINEERING CONDITIONAL DISPLACEMENTS**

Here, we describe how we engineer the conditional displacement interactions that make up \( \hat{H}_{LVC} \). As discussed in the main text, this Hamiltonian consists of two simultaneous conditional displacement interactions \( \{ \hat{H}_q, \hat{H}_b \} \) of a single qubit to two different cavity modes, where the qubit coupling axes are orthogonal. To simplify the derivation, we will begin by focusing on how we enact just one of these interactions. As we will see, adding additional interactions to other cavity modes is relatively straightforward, and the qubit coupling axis is freely adjustable in the effective \( x-y \) plane.

We expand upon the derivation provided by Ref. [33] by incorporating the finite anharmonicity of the transmon mode. We begin with the static Hamiltonian of a transmon mode \( \hat{q} \) dispersively coupled to a cavity mode \( \hat{c} \):

\[
\hat{H}_{\text{static}}/\hbar = \omega_c \hat{c}^\dagger \hat{c} + \omega_q \hat{q}^\dagger \hat{q} - \frac{\alpha_q}{2} \hat{q}^\dagger \hat{q}^\dagger \hat{q} \hat{q} - \chi \hat{c}^\dagger \hat{c}^\dagger \hat{q} \hat{q}, \quad (B1)
\]

where \( \alpha_q \) is the transmon anharmonicity and \( \chi \) is the dispersive shift. At a high level, we will see that the conditional displacement interaction arises by transforming the cross-Kerr interaction between the transmon and the cavity. Thus, our approach will be to consider how driving each mode transforms the static interaction. Specifically, we drive the system with one tone coupled to the transmon and two coupled to the cavity:

\[
\hat{H}_d/\hbar = 2e_R \cos(\omega_q + \Delta_R t) (\hat{q} + \hat{q}^\dagger) - 2ie_1 \sin(\omega_t + \phi_1) (\hat{c} - \hat{c}^\dagger) - 2ie_2 \sin(\omega_t + \phi_2) (\hat{c} - \hat{c}^\dagger), \quad (B2)
\]

such that the full system Hamiltonian is described by \( \hat{H} = \hat{H}_{\text{static}} + \hat{H}_d \). For convenience, we regroup the terms such that we can write \( \hat{H} = \hat{H}_q(\hat{q}, \hat{q}^\dagger) + \hat{H}_c(\hat{c}, \hat{c}^\dagger) + \hat{H}_{\text{int}}, \) where \( \hat{H}_{\text{int}}/\hbar = -\chi \hat{c}^\dagger \hat{c}^\dagger \hat{q} \hat{q} \). We first go into the rotating frame of the transmon drive via \( \hat{H} \rightarrow U \hat{H} U^\dagger + i \dot{U} \dot{U}^\dagger \), where \( U = e^{i(\omega_q + \Delta_e)\hat{q}\hat{q}^\dagger} \):

\[
\hat{H}/\hbar = -\Delta_R \hat{q}^\dagger \hat{q} - \frac{\alpha_q}{2} \hat{q}^\dagger \hat{q}^\dagger \hat{q} \hat{q} + e_R (\hat{q} + \hat{q}^\dagger) + \hat{H}_c/\hbar - \chi \hat{c}^\dagger \hat{c}^\dagger \hat{q} \hat{q}, \quad (B3)
\]

noting that we have performed the rotating wave approximation and discarded terms rotating at \( \mathcal{O}(\omega_q) \). Furthermore, the cross-Kerr term remains unaffected since it is proportional to \( \hat{q}^\dagger \hat{q} \). Now, we diagonalize \( \hat{H}_q \) and reexpress it in the resulting eigenbasis:

\[
\hat{H}_q/\hbar = \sum_i e_i |i\rangle \langle i|, \quad (B4)
\]

where we label \( i \in \{+, -, \tilde{f}, \ldots\} \) in correspondence with the fact that we will be working in a regime where the two lowest driven eigenstates strongly resemble those of a standard qubit that is driven on resonance, but now incorporate a weak dressing with higher levels of the transmon. We identify the Rabi frequency to be the energy difference between the lowest two eigenstates \( \epsilon_+ - \epsilon_- = \Omega_R \) and define an effective anharmonicity as \( \epsilon_- - \epsilon_f = \Omega_R + \tilde{g} \). At this stage, we turn to numerics and construct a unitary basis transformation between the undriven and driven transmon eigenstates for a finite truncation of the transmon Hilbert space. We then reexpress the cross-Kerr interaction in the driven basis, giving us

\[
\hat{H}/\hbar = \sum_i e_i |i\rangle \langle i| + \hat{H}_c/\hbar - \chi \hat{c}^\dagger \hat{c} \sum_{jk} u_{jk} |j\rangle \langle k|, \quad (B5)
\]

We can further simplify this by going into the frame of the driven transmon \( \dot{U} = e^{i\epsilon_q t/\hbar} \), which performs the transformations \( |j\rangle \rightarrow e^{i\epsilon_q t/\hbar} |j\rangle \), resulting in

\[
\hat{H}/\hbar = \hat{H}_c/\hbar - \chi \hat{c}^\dagger \hat{c} \sum_{jk} u_{jk} e^{i(\epsilon_f - \epsilon_-)/\hbar} |j\rangle \langle k|. \quad (B6)
\]

where \( u_{jk} = u_{kj} \). We consider the terms associated with the lowest three levels explicitly:

\[
\hat{q}^\dagger \hat{q} = u_{++} |+\rangle \langle +| + u_{+-} |+\rangle \langle -| + u_{\tilde{f}j} |\tilde{f}\rangle \langle \tilde{f}| + u_{+-} e^{i(\Omega_R + \tilde{g})t/\hbar} |+\rangle \langle -| + u_{\tilde{f}j} e^{i(\Omega_R + \tilde{g})t/\hbar} |\tilde{f}\rangle \langle \tilde{f}| + u_{+-} e^{i(2\Omega_R + \tilde{g})t/\hbar} |+\rangle \langle +| + u_{\tilde{f}j} e^{i(2\Omega_R + \tilde{g})t/\hbar} |\tilde{f}\rangle \langle \tilde{f}|. \quad (B7)
\]

At this stage, we pause and turn to simplify \( \hat{H}_c \). First, we choose to parametrize the two drive frequencies \( \omega_{1/2} = \omega_e - \Delta_e \mp \Omega_R \). By going into the rotating frame
at the average of the drive frequencies \( \hat{U} = e^{i(\omega_c - \Delta_c)\hat{c}\hat{\varphi}} \), we arrive at

\[
\hat{H}/\hbar = \hat{H}_{\text{int}}/\hbar + \Delta_c \hat{c}^\dagger \hat{c} - e_2 (\hat{c}e^{i\Omega_0 t + i\phi_1} + \hat{c}^\dagger e^{i\Omega_0 t - i\phi_1}) \\
- e_2 (\hat{c}e^{i\Omega_0 t + i\phi_2} + \hat{c}^\dagger e^{-i\Omega_0 t - i\phi_2}).
\] (B8)

Finally, we assume that the drive strengths are equal \( e_2 = -e_1 = e \) and parametrize the drive phases as their sum and differential components \( \phi_2 = (\phi_1 + \phi_2)/2 \) and \( \phi_4 = (\phi_1 - \phi_2)/2 \). This allows us to further simplify our Hamiltonian to

\[
\hat{H}/\hbar = \hat{H}_{\text{int}}/\hbar + \Delta_c \hat{c}^\dagger \hat{c} - 2ie \sin(\Omega_R t - \phi_4) \\
\times (\hat{c}e^{i\phi_2} - \hat{c}^\dagger e^{-i\phi_2}).
\] (B9)

We can observe here that the sum phase of the two sidebands contributes simply as a static rotation of \( \hat{c} \); therefore we can always align to this frame by experimentally adjusting this phase. Hence, we will set \( \phi_4 = 0 \) here on out to simplify our expressions. At this stage, we aim to eliminate this time-dependent drive term by performing a displacement transformation \( \hat{U} = e^{i(\xi(t) - \xi^*(t))\hat{c}} \). This is achieved by choosing \( \xi(t) = (2e/\Omega_R)\cos(\Omega_R t + \phi_4) = \xi_0 e^{i(\Omega_0 t + \phi_0)} + e^{-i(\Omega_0 t + \phi_0)} \), where \( \xi_0 = (e/\Omega_R) \), which also transforms \( \hat{c} \to \hat{c} + \xi(t) \). This gives

\[
\hat{H}/\hbar = \Delta_c [\hat{c}^\dagger + \xi^*(t)] [\hat{c} + \xi(t)] \\
- \chi [\hat{c}^\dagger + \xi^*(t)][\hat{c} + \xi(t)] \sum_{jk} u_{jk} e^{i(\gamma_j - \gamma_k)/\hbar} j\rangle \langle k| \\
= \Delta_c [\hat{c}^\dagger + \xi(t)] [\hat{c} + \hat{c}^\dagger] + \xi^2_0 \\
- \chi [\hat{c}^\dagger + \xi_0 e^{i(\Omega_0 t + \phi_0)} + e^{-i(\Omega_0 t + \phi_0)}] [\hat{c}^\dagger + \hat{c}] + \xi^2_0 \\
\times \sum_{jk} u_{jk} e^{i(\gamma_j - \gamma_k)/\hbar} j\rangle \langle k|. 
\] (B10)

By substituting the expansion for \( \hat{\alpha}^\dagger \hat{\alpha} \), discarding terms that rotate at \( \Omega_R \) and higher, and neglecting constant offsets, we are left with an effective static interaction Hamiltonian:

\[
\hat{H}/\hbar = \Delta_c \hat{c}^\dagger \hat{c} \\
- \chi \xi_0 u_{+-} (e^{-i\phi_1} |+\rangle \langle -| + e^{i\phi_1} |+\rangle \langle +|)(\hat{c} + \hat{c}^\dagger) \\
- \chi \xi_0 u_{++} |+\rangle \langle +| + u_{--} |\rangle \langle +| + u_{+\dagger} [\hat{f}] \langle \hat{f}^\dagger]. 
\] (B11)

1. Optimizing the static cross-Kerr

Here, we consider Eq. (B10) and ask the following questions. How large do we need the Rabi frequency to be in order to safely discard all the rotating terms? Does the answer to this question inform any design choices with regard to our static Hamiltonian? To answer these questions, we first consider all of the different terms that rotate at \( \Omega_R \), neglecting any phases:

\[
\Delta_c \xi_0 (\hat{c} + \hat{c}^\dagger) - \chi u_{+-} \hat{c} \hat{\sigma}_+ - \chi \xi_0^2 u_{+-} \hat{\sigma}_+ e^{\pm i\Omega_0 t}. 
\]

Note that there are also terms that rotate at \( \tilde{\alpha} + 2\Omega_R + \tilde{\alpha} \), \( 2\Omega_R + \tilde{\alpha} \), and \( 3\Omega_R + \tilde{\alpha} \), but since the prefactors will all be of the same order, we consider the smallest rotating frequency for the most stringent condition. Importantly, we also require that \( \tilde{\alpha} > \Omega_R \), otherwise other terms involving \( \hat{f} \) will be activated and we can no longer restrict ourselves to a qubit subspace. This sets a limit on how large of a Rabi frequency can be used for a fixed anharmonicity \( \alpha_q \). The above terms reveal that our conditions for the RWA are

\[
\Omega_R \gg \left\{ \Delta_c \xi_0 \langle (\hat{c} + \hat{c}^\dagger) |, \frac{\chi}{2} (\hat{c}^\dagger \hat{c}), \frac{\chi^2}{2\xi_0^2} \right\}, 
\] (B13)

which notably depends on the state of the cavity. It is clear from this that as the conditional displacement interaction...
strength \( g \approx (\chi \xi_0 / 2) \) increases, the approximation becomes less valid. However, we can instead rewrite the condition for a fixed \( g \):

\[
\Omega_R \gg \left\{ \Delta_\omega |\xi_0 (\langle \hat{c}^\dagger \hat{c} \rangle) \rangle, \frac{g}{\xi_0} \langle \hat{c}^\dagger \hat{c} \rangle, g \xi_0 \right\}, \tag{B14}
\]

which reveals that there is indeed an optimal value for \( \xi_0 \) given a fixed \( g \). For considering photon numbers \( \langle \hat{c}^\dagger \hat{c} \rangle \sim \mathcal{O}(1) \) and \( g \approx \Delta_\omega \), we best satisfy all these conditions by choosing \( \xi_0 \approx 1 \). This, in turn for a fixed \( g \), suggests that we should roughly target a static cross-Kerr strength of \( \chi \approx 2g \).

2. Choice of static detuning of the Rabi drive

The final term in Eq. (B11) represents an effective cross-Kerr interaction between cavity photons and the driven transmon eigenstates. For a true two-level system driven on resonance, which is a good approximation for transmons in the regime that the Rabi frequency is much weaker than the resonance, which is a good approximation for transmons in transmon eigenstates. For a true two-level system driven on resonance, which is a good approximation for transmons in transmon eigenstates. For a true two-level system driven on resonance, which is a good approximation for transmons in transmon eigenstates. For a true two-level system driven on resonance, which is a good approximation for transmons in transmon eigenstates. For a true two-level system driven on resonance, which is a good approximation for transmons in transmon eigenstates.

By adding an additional static detuning knob \( \Delta_R \) on the Rabi drive, we can determine an optimal working configuration that nulls this effective cross-Kerr. We show this optimization in Fig. 5. The presence of this static detuning thus dictates that we perform an adiabatic preparation of our driven qubit eigenstates. This has the further benefit of eliminating leakage events associated with large Rabi frequencies and finite transmon anharmonicity, but has an additional challenge which we address in Appendix D 5.

APPENDIX C: EXPERIMENTAL DETAILS

A list of the static system parameters is given in Table I. A schematic of the wiring diagram for this experiment is depicted in Fig. 6. We comment below on notable features not explicitly shown in the wiring diagram.

1. Wiring diagram

A field-programmable gate array-based quantum controller synchronizes multiple modules that contain DACs and ADCs for generating the pulses (I and Q control) and digitizing the readout signals, respectively. rf switches are only open while pulses are played on a corresponding mode. The control line for the tuning mode is split between the resonant drive (left) and sideband pumps (right), which are never played simultaneously; thus a closed loop where both rf switches are open together is never formed. The band-stop filter on the pump line is centered on the tuning mode’s resonance frequency, suppressing pump-induced noise that may lead to heating of the cavity mode. dc blocks are placed around each active component, as well as on each line at the boundary of the cryostat (i.e., the line separating 300 and 4 K in the schematic). All components in the 20 mK region are thermally anchored to the mixing chamber plate via OFHC copper braids. We use Josephson Parametric Converters (JPCs) as quantum limited amplifiers—only coupling to the signal port is shown.

2. Temperature stabilization

As the derivations of Appendix B suggest, the resonance condition for enforcing our reaction Hamiltonian [Eq. (3) of the main text] relies on matching the sideband detunings to the Rabi frequency. The Rabi frequency depends linearly.

![FIG. 5. Optimizing the static detuning. By numerically diagonalizing \( \hat{H}_q / \hbar \) for various values of \( \Delta_R \), we can plot the dimensionless factor that contributes to the residual cross-Kerr for \( \Omega_R / 2\pi = 80 \text{ MHz} \) and \( \alpha / 2\pi = 244 \text{ MHz} \). We find an optimal value of \( \Delta_R / 2\pi \approx 7 \text{ MHz} \).](image-url)

<table>
<thead>
<tr>
<th>System quantity</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transmon frequency</td>
<td>( \omega_a / 2\pi )</td>
<td>4850 MHz</td>
</tr>
<tr>
<td>Transmon anharmonicity</td>
<td>( \alpha_q / 2\pi )</td>
<td>244 MHz</td>
</tr>
<tr>
<td>Transmon relaxation</td>
<td>( T_1^q )</td>
<td>80 \mu s</td>
</tr>
<tr>
<td>Transmon decoherence</td>
<td>( T_2^q )</td>
<td>7 \mu s</td>
</tr>
<tr>
<td>Tuning mode frequency</td>
<td>( \omega_q / 2\pi )</td>
<td>5436 MHz</td>
</tr>
<tr>
<td>Tuning mode linewidth</td>
<td>( \kappa_2 / 2\pi )</td>
<td>0.23 kHz</td>
</tr>
<tr>
<td>Coupling mode frequency</td>
<td>( \omega_b / 2\pi )</td>
<td>6506 MHz</td>
</tr>
<tr>
<td>Coupling mode linewidth</td>
<td>( \kappa_3 / 2\pi )</td>
<td>320 kHz</td>
</tr>
<tr>
<td>Tuning mode–Transmon coupling</td>
<td>( \chi_{\omega_q} / 2\pi )</td>
<td>295 kHz</td>
</tr>
<tr>
<td>Coupling mode–Transmon coupling</td>
<td>( \chi_{\omega_b} / 2\pi )</td>
<td>210 kHz</td>
</tr>
<tr>
<td>Ancilla frequency</td>
<td>( \omega_a / 2\pi )</td>
<td>4509 MHz</td>
</tr>
<tr>
<td>Ancilla relaxation</td>
<td>( T_1^{\omega_a} )</td>
<td>60 \mu s</td>
</tr>
<tr>
<td>Ancilla decoherence</td>
<td>( T_2^{\omega_a} )</td>
<td>10 \mu s</td>
</tr>
<tr>
<td>Tuning mode–Ancilla coupling</td>
<td>( \chi_{\omega_a} / 2\pi )</td>
<td>845 kHz</td>
</tr>
</tbody>
</table>
on the amplitude of the Rabi drive [roughly speaking, using the two-level approximation that $\Omega_R = \sqrt{\epsilon_R^2 + \Delta_R^2}$ and we operate in a regime where $\epsilon_R \gg \Delta_R$, resulting in $\Omega_R \approx \epsilon_R[1 + \frac{1}{2} \Delta_R^2\epsilon_R^2]$], and thus is susceptible to amplitude fluctuations such as those caused by variations in the gain at any stage of our microwave control chain. A dominant source of these variations is due to ambient temperature fluctuations in the lab. To this end, we suppress these fluctuations by anchoring the components (particularly a Marki LXP IQ mixer and MITEQ low noise amplifier; see turquoise box in Fig. 6) to a Thorlabs optical breadboard and placing the breadboard in a cardboard box. We then actively stabilize the temperature of the air in the box via an op-amp-based proportional-integral-derivative
feedback controller that heats an Ohmite ceramic resistor ($R = 2.5 \, \Omega$) based on a differential measurement of the temperature using a 100 kΩ thermistor referenced to a set point. Figure 7 shows the typical performance of our stabilization and correlates the temperature variations with the amplitude variations as measured via the Rabi frequency. Over the course of 24 hours, we achieve an absolute temperature stability within 50 mK and a relative amplitude stability of $10^{-3}$, suggesting that we have a relative amplitude sensitivity of 1% per 500 mK. We note that the timescale for a typical calibration and measurement of a dataset presented in this paper is roughly a few hours, meaning we can operate in a window where the relative amplitude stability can be much better than $10^{-3}$.

APPENDIX D: CALIBRATION PROCEDURES

In this appendix, we describe the various calibration experiments in detail along with their pulse sequences.

1. Tomography of the Rabi qubit

We initialize eigenstates $|\pm\rangle$ of $\hat{\sigma}_x$ in the driven frame by first preparing the corresponding states of the undriven transmon via a standard $\pi/2$ rotation and then adiabatically ramping on the Rabi drive $\tau_{\text{ramp}} \gg 1/\Omega_R$. In order to measure the Rabi qubit in the driven basis, we need a decoding operation that maps the driven states $|\pm\rangle$ onto our measurement basis $|g,e\rangle$. Given that our decoding operation is a $\pi/2$ rotation performed using the same microwave clock as the initial $\pi/2$ rotation, we need to know the phase evolution of the $|\pm\rangle$ states in the frame of this drive centered at the bare $g-e$ transition frequency $\omega_q$ of the transmon. Notably, this phase depends on the Rabi frequency $\Omega_R$, the static detuning $\Delta_R$, the evolution time $\tau$, and a fixed offset phase $\phi_0$:

$$\varphi_{\text{decode}} = (\Omega_R - \Delta_R)\tau + \phi_0. \quad (D1)$$

By calibrating this rate appropriately, we are able to continuously measure $\langle \hat{\sigma}_x \rangle$ as a function of time and extract a corresponding driven coherence time (see Fig. 8) [34]. In all of the experiments in this paper, we choose to map $|+\rangle \rightarrow |e\rangle$ and $|-\rangle \rightarrow |g\rangle$. As such, we have the mapping $\langle \hat{\sigma}_x \rangle = 2\langle \hat{P}_x \rangle - 1$. 

![Graph](image-url)

**FIG. 7.** Tracking system stability over time. Top panel: simultaneous measurements of the Rabi frequency and the ambient temperature (inferred from the measured thermistor resistance) where the active microwave components are held. The data shows that the two are correlated. Middle panel: extracting driven coherences $T_{2p}$ suggests a stable amplitude noise spectrum within an acquisition time $\tau_{\text{acq}} = 3 \, \text{min}$ (bottom left-hand panel: a typical time-domain Ramsey trace), with a single instance where the amplitude drift was large (bottom right-hand panel), as confirmed via looking at the raw data binned 10 shots at a time over $\tau_{\text{acq}}$. **OBSERVATION OF WAVE-PACKET BRANCHING THROUGH AN … PHYS. REV. X 13, 011008 (2023)**
system, we have $\hat{c} \in \{\hat{a}, \hat{b}\}$. The qualitative behavior of each individual sideband interacting with the driven qubit will be different given that we are operating in the regime where $g > \kappa_d$ and $g \lesssim \kappa_b$. The former will result in either creating and annihilating two excitations simultaneously $|\tilde{g}, 0\rangle \leftrightarrow |\tilde{e}, 1\rangle$ (red sideband) or a coherent exchange between an excitation in the driven qubit and a photon in the cavity $|\tilde{e}, 0\rangle \leftrightarrow |\tilde{g}, 1\rangle$ (blue sideband) [51]. The latter will stabilize the qubit in either the excited state $|\tilde{e}\rangle$ (red sideband) or the ground state $|\tilde{g}\rangle$ (blue sideband) [52]. To solidify the connection with these previous experiments, we note that $|\tilde{g}\rangle \approx ((1/\sqrt{2})(|g\rangle + |e\rangle)$ and $|\tilde{e}\rangle \approx (1/\sqrt{2})(|g\rangle - |e\rangle)$ when expressed in the basis of undriven transmon eigenstates.

In order to calibrate the interaction strengths, we will operate in the restricted two-dimensional subspace of the joint Hilbert space of the cavity and qubit as described above. This allows us to simplify our analysis and replace the qubit raising and lowering operators $\sigma^\ddagger$ with general bosonic creation and annihilation operators $\hat{d}^{(i)}$. We then follow Refs. [47,53] and capture the full range of dynamics by solving the equations of motion for $\hat{d}$ under $\hat{H}_{\text{blue}}$ and incorporating a cavity damping rate $\kappa$. We also include a static detuning term $\delta \hat{c}$ to capture the effect of sweeping the pump frequency that enables the interaction. The resulting field has the form:

$$\hat{d}(t) = \frac{\hat{d}(0)}{\Omega} e^{-\kappa_{eff} t/4} \left[ \Omega \cosh \left( \frac{\Omega t}{4} \right) + \kappa_{eff} \sinh \left( \frac{\Omega t}{4} \right) \right],$$

(D4)

where $\Omega = \sqrt{\kappa_{eff}^2 - (4g)^2}$ and $\kappa_{eff} = \kappa + 2i\delta$.

For each interaction, we prepare our system in either $|\tilde{e}, 0\rangle$ (blue sidebands) or $|\tilde{g}, 0\rangle$ (red sidebands) and scan the frequency of the cavity sideband and the delay time for a given pump amplitude. By measuring the qubit population, we extract $\langle \hat{d}^{\dagger}(t)\hat{d}(t) \rangle$ and can fit the resulting data using Eq. (D4). For the exchange interaction under the blue sideband, we have $\langle \hat{d}^{\dagger}(0)\hat{d}(0) \rangle_{\text{blue}} = 1$ for the qubit initially in its excited state, giving us an expression for $\langle \hat{d}^{\dagger}(t)\hat{d}(t) \rangle_{\text{blue}}$. For the red sideband, the features are qualitatively identical, with the exception that the qubit starts out in the ground state, giving us $\langle \hat{d}^{\dagger}(t)\hat{d}(t) \rangle_{\text{red}} = 1 - \langle \hat{d}^{\dagger}(t)\hat{d}(t) \rangle_{\text{blue}}$. Notably, this assumes that the effective interaction strength $g$ is independent of the pump detuning $\delta$, which is strictly not true but should be a very good approximation in our regime given that the scale of the chevron features, set by $g$, is much smaller than the absolute detuning from the cavity resonance $\sim k_R$. We allow for an overall amplitude, global offset, and time offset in our fit, leaving $g$ and $\kappa$ to be the only free parameters. In the case of the coupling mode, we first perform this fit for a range of interaction strengths $g \approx \kappa_b$.

2. Individual sideband interactions

The sideband interactions have the form:

$$\hat{H}_{\text{red}}/\hbar = g\hat{c}\hat{\sigma}^- + g^*\hat{c}^\dagger \hat{\sigma}^+, \quad (D2)$$

$$\hat{H}_{\text{blue}}/\hbar = g\hat{c}\hat{\sigma}^+ + g^*\hat{c}^\dagger \hat{\sigma}^-, \quad (D3)$$

for a general cavity annihilation operator $\hat{c}$, and $\hat{\sigma}^{\pm(-)} = \langle \tilde{e}(\tilde{g}) \rangle \langle \tilde{g}(\tilde{e}) \rangle$ are the raising and lowering operators of the two driven qubit eigenstates that adiabatically connect to the ground and first excited state of the transmon. For our
and extract a decay rate $\kappa_b/2\pi \approx 320$ kHz. For the remainder of the calibrations where $g < \kappa_b$, we fix this quantity and let the interaction strength $g$ be the only free parameter to be fitted. The full calibration for different pump amplitudes is shown in Fig. 9.

3. Conditional displacements

The combination of simultaneous red and blue cavity sidebands enacts a conditional displacement interaction. This requires that two conditions are fulfilled: (1) the interaction strengths of each individual sideband interaction be equal and (2) the frequency difference between the two sidebands equals twice the Rabi frequency. If these two conditions are met, then we can model the interaction using Eq. (B12).

In practice, the presence of each sideband will Stark shift both the transmon and cavity modes. Thus, to capture the dominant effect of all of these Stark shifts (which influences the resonance condition), we perform individual sideband calibrations with the opposite sideband on but detuned by an amount larger than the interaction strength we are using (i.e., by an additional 2 MHz in our experiments).

We scan the pump amplitudes and match the individual sideband strengths before bringing both sidebands into resonance. This relies on the assumption that over a variation of $\sim 2$ MHz, the relative change in the cavity Stark shift, which influences the dimensionless pump strength that determines the interaction strength, is negligible. Finally, we fine-tune the difference frequency of the two sidebands while keeping the average value fixed [which fixes $\Delta_c$ in Eq. (B12) in order to account for any change in the Rabi frequency which we are very sensitive to]. This sensitivity is revealed by measuring the transverse relaxation time $T_2^\rho$, and choosing a calibration point where this value is maximized, suggesting that the resonance conditions are fulfilled as well as possible. This calibration procedure gives us the data shown in Fig. 2(b) of the main text.

For the tuning mode, the dynamics of an initial vacuum state $|0\rangle_a$ evolving under a conditional displacement...
interaction will be a circular trajectory in phase space around the location of the ground state  \( \alpha_g = g_x / \Delta_a \). By measuring the population in \( |n = 0\rangle \), we are effectively measuring the overlap of a coherent state with itself as it oscillates in time. This justifies the use of a simple model, where the state autocorrelation function is \( \langle \hat{g} e^{-i\Delta\tau} \hat{g} \rangle = e^{2|\beta|^2 |\cos(\Delta\tau)| - 1} \). The corresponding probability is

\[
P_0 = \langle \hat{g} e^{-i\Delta\tau} \hat{g} \rangle^2 = e^{2|\beta|^2 |\cos(\Delta\tau)| - 1},
\]

To make the connection with our model, we choose \( \beta = \alpha_g = g_x / \Delta_a \). We use Eq. (D5) (along with an overall amplitude and offset to account for preparation and measurement errors) as our fitting function for the data in Fig. 2(b) of the main text.

4. Cavity displacements along the interaction axis

As described in Appendix B, the cavity phase of the conditional displacement [i.e., the phase which defines the position operator \( \hat{x} \propto (\hat{e} e^{i\phi_{\text{disp}}} + \hat{e}^\dagger e^{-i\phi_{\text{disp}}}) \)] is determined by the sum phase of the red and blue sidebands. Given that we are turning on the conditional displacement interaction suddenly (\( \tau_{\text{ramp}} \ll 1/g \)), the phase of our initial displacement operation \( \hat{D}(\alpha_0) \) on the tuning mode will determine the location in the driven phase space where the wave packet begins. Displacements whose phase is aligned to the conditional displacement cavity phase will prepare wave packets along the position axis, whereas care needs to be taken to prepare wave packets with various momentums that are located at one of the two ground state positions. In our experiment, we prepare coherent states with no initial momentum along the reaction coordinate.

In order to calibrate the displacement phase, we begin with a calibrated conditional displacement where we have extracted \( g_x \) and \( \Delta_a \), which gives us a value for the ground state amplitude \( \alpha_g = g_x / \Delta_a \) [Fig. 2(b) of the main text]. Note that this does not rely on any displacement phase since we are starting off in a vacuum state. Next, we scan the phase of an initial displacement of \( 2\alpha_g \) and turn on the conditional displacement interaction for various delay times. The optimal phase will be the one where we recover revivals that are half a period out of phase from those in Fig. 2(b) of the main text. This can be interpreted as follows. A vacuum state in the lab frame \( |0\rangle_{\text{lab}} \) looks like a displaced state \( |\alpha_g\rangle_{\text{disp}} \) with respect to the displaced ground state \( |\alpha_g\rangle_{\text{lab}} = |0\rangle_{\text{disp}} \), and thus will oscillate around the ground state, reaching \( |\alpha_g\rangle_{\text{disp}} \) after half a period. By determining the phase that enables us to prepare \( |\alpha_g\rangle_{\text{disp}} \) (which is \( 2|\alpha_g\rangle_{\text{lab}} \) in the lab frame and will return to the vacuum state \( |\alpha_g\rangle_{\text{disp}} \) after half a period), we can prepare any state along the position axis, including the displaced ground state, as shown in Fig. 10(b).

5. Echoing the residual entanglement during ramping of the Rabi drive

The adiabatic preparation of our driven qubit eigenstates as motivated in Appendix B 2 has two benefits: for a fixed transmon anharmonicity, we can (1) use larger Rabi frequencies while canceling the residual cross-Kerr and (2) avoid leakage events to higher transmon levels (up to...

---

**FIG. 10.** Calibrating conditional displacements. (a) Pulse sequence. The displacement pulse is played after the Rabi drive is turned on and the cross-Kerr is nulled. An echo sequence eliminates the residual entanglement between the qubit and cavity during the ramp off of the Rabi drive. (b) Top: coherent revivals for various initial displacements along the interaction axis, probed through measurement of the vacuum projector. The contrast is maximized when the wave packet passes through the origin, and oscillations vanish when the ground state \( \alpha_g \approx 1.3 \) is prepared. The data in Fig. 3(b) are taken at the location of the yellow star. Bottom: simultaneous measurement of \( T_2^p \) reveals driven coherence times with a slight dependence on \( \langle \hat{a} \hat{a} \rangle \).
is fully ramped on, i.e., during the time when the cross-Kerr interaction is nulled [see Fig. 10(a)]. For addressing the entanglement during the ramp off of the Rabi drive, we implement a simple and short echo sequence of the qubit to undo the interaction. This works because the entanglement is fully determined by $\chi$ and the ramp time $\tau_{\text{ramp}}$ and not the cavity photon distribution. This is important as we do not want a scheme which depends on the cavity state that we are manipulating. Figure 11 shows how implementing this protocol eliminates spurious features that arise from this entanglement when performing a decode calibration experiment [Fig. 8(b)] when $\hat{H}_J$ is active. In practice, this calibration is only done with respect to photons in the tuning mode. Given that we operate the coupling mode in a regime where $g_y < \kappa$, the photon distribution remains relatively small and thus any residual entanglement effects are negligible.

**APPENDIX E: EXTENDED DATA AND POSTSELECTING ON LEAKAGE EVENTS**

In this appendix, we present additional data (Fig. 12) that support what is shown in the main text, specifically focusing on leakage statistics. Our measurement of the transmon is able to distinguish between $\{|g\rangle, |e\rangle, |f\rangle\}$ on a single-shot basis, which gives us information on leakage events outside the qubit manifold that we use for our experiments. In the absence of decoherence, our adiabatic preparation scheme should ideally eliminate any leakage to the second excited state $|f\rangle$ and higher (assuming there are no accidental resonances induced by the drives). In practice, any relaxation or heating between undriven transmon levels will lead to transitions between the driven eigenstates that have finite support across multiple undriven basis states. In all of the data presented, we postselect away outcomes where the transmon is measured to be in $|f\rangle$. We compile the postselection statistics in Table II, and note that overall the leakage probabilities are small.

**APPENDIX F: MASTER EQUATION SIMULATIONS**

The theoretical predictions for $\langle \hat{\sigma}_x \rangle$ in Figs. 2(d) and 4(b) are obtained by performing a numerical simulation of a full time-domain master equation using a PYTHON-based open source software (QuTiP):

$$\dot{\hat{\rho}} = -\frac{i}{\hbar} [\hat{H}_{\text{int}}, \hat{\rho}] + \kappa_b \mathcal{D}[\hat{b}]\hat{\rho} + \frac{\gamma_y}{2} \mathcal{D}[\hat{\sigma}_y] \hat{\rho},$$

where the left-hand side of the equation is the time derivative of the system’s density matrix and $\mathcal{D}[\hat{A}]\hat{\rho} = \hat{A} \hat{\rho} \hat{A}^\dagger - \frac{1}{2} \hat{A}^\dagger \hat{A} \hat{\rho} - \frac{1}{2} \hat{\rho} \hat{A}^\dagger \hat{A}$.

We extract $\langle \hat{\sigma}_x \rangle$ by plotting $\text{Tr}[\hat{\rho} \hat{\sigma}_x]$, where $\hat{\rho}_{\text{qubit}} = \text{Tr}_{a,b} \hat{\rho}$ is the reduced density matrix of the qubit only. For the measurement-induced...
dephasing in Fig. 2(d), we use $\hat{H}_{\text{sim}} = \hat{H}_b$ from Eq. (2) and an initial state $|\!\rangle \otimes |0\rangle_b$ for various values of $\Delta_b$. For the full system dynamics through the conical intersection in Fig. 4(b), we use $\hat{H}_{\text{sim}} = \hat{H}_{\text{LVC}}$ from Eq. (3) and initial states $|\!\rangle \otimes |\alpha_0\rangle_a \otimes |0\rangle_b$ for various values of $\alpha_0$.

Across these two simulations, the parameters $g_x, \Delta_a, g_y, \kappa_b$ are determined via independent calibrations, and $\Delta_b$ is estimated via the frequencies of the sidebands on the coupling mode. The value of $\gamma_y$ is set by an independent measurement of $T_{2\rho}^x$, specifically via $\gamma_y = 1/T_{2\rho}^x$. Finally, the amplitude and offset are adjusted using the measured experimental values from the corresponding $T_{2\rho}^x$ control measurement, leaving zero free parameters.

APPENDIX G: DISSIPATION ANALYSIS AND ZENO DYNAMICS

In this appendix, we describe the oscillator-induced dissipation of the qubit resulting from the evolution under the master equation,

$$\dot{\rho} = -\frac{i}{\hbar} [\hat{H}_{\text{LVC}}, \rho] + \kappa_b \left( b \rho \hat{b}^\dagger - \frac{1}{2} \hat{b}^\dagger \hat{b} \rho - \frac{1}{2} \rho \hat{b}^\dagger \hat{b} \right). \quad (G1)$$

TABLE II. Extended data information. Quantities in brackets correspond to identifiers within a dataset. Different values of $\Delta_c, c \in \{a, b\}$, require a fine-tuning calibration on the drive configuration to match Rabi frequency shifts, but preparing different initial states does not.

<table>
<thead>
<tr>
<th>Dataset</th>
<th>Drives (+Rabi)</th>
<th>Figures</th>
<th>$\Omega_k[\Delta_c]/2\pi$ (MHz)</th>
<th>$T_{2\rho}$ $[\alpha_0]$ (µs)</th>
<th>Leakage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coherent state revivals</td>
<td>Tuning mode sidebands</td>
<td>2(c)</td>
<td>82.532 [0.457]</td>
<td>Not measured</td>
<td>Fig. 12(a)</td>
</tr>
<tr>
<td>Measurement-induced</td>
<td>Coupling mode sidebands</td>
<td>2(d)</td>
<td>79.915 [0]</td>
<td>36.6</td>
<td>Fig. 12(b)</td>
</tr>
<tr>
<td>Dephasing</td>
<td>Tuning and coupling mode sidebands</td>
<td>10(b)</td>
<td>80.045</td>
<td>see Fig. 10(b)</td>
<td>Fig. 12(c)</td>
</tr>
<tr>
<td>Coherent revivals + state</td>
<td>Tuning and coupling mode sidebands</td>
<td>4(a)</td>
<td>81.013</td>
<td></td>
<td>Fig. 12(d)</td>
</tr>
<tr>
<td>preparation</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
with
\[ \hat{H}_{Zeno} / \hbar = E(x) \hat{\sigma}_z + \Delta_b \hat{b}^\dagger \hat{b} + g \hat{\sigma}_z (\hat{b}^\dagger + \hat{b}) \] (G2)

from Eq. (5) of the main text. The dynamics of the system are effectively the same as Zeno dynamics of a driven qubit generated by the competition between \( E(x) \hat{\sigma}_z \), which tries to lock the state of the qubit to an eigenstate of \( \hat{\sigma}_z \), and \( g \hat{\sigma}_z (\hat{b}^\dagger + \hat{b}) \), which in combination with photon loss in the oscillator tries to project the state of the qubit onto an eigenstate of \( \hat{\sigma}_x \). Note that here we choose the dissipation to be along \( \hat{\sigma}_x \) as opposed to \( \hat{\sigma}_z \). The resulting dissipative dynamics is crucially dependent on the parameters \( E(x), \Delta_b, g, \kappa_b \). While an exact analytic expression for the time dependence of the qubit density matrix cannot be obtained in general, here we consider a few extreme parameter regimes which can be easily analyzed and provide a window to the vast range of qubit dynamics possible.

This simplest case is when \( E(x) = 0 \). In this case, it is possible to use Fokker-Planck equations to calculate an effective dephasing rate. However, when \( E(x) \neq 0 \), this approach will fail to give an analytical expression for the dephasing rate. In this case, we will invoke additional constraints which will allow us to apply the Born-Markov approximation and derive an effective master equation for the qubit.

1. \( E(x) \neq 0 \)

To get a simple intuitive understanding of the oscillator-induced dephasing when \( E(x) \neq 0 \), we consider two cases: (a) \( \Delta_b = 0, \kappa_g \gg g \) and (b) \( |2\Delta_b - E(x)| \ll |2\Delta_b + E(x)|, |2\Delta_b + E(x)| \gg \kappa_b \gg |2\Delta_b - E(x)|, g \).

Case (a)—In this case, the oscillator mode can be eliminated using the standard Born-Markov approximation and a simple effective master equation for the qubit is obtained:
\[ \dot{\rho}_q = -i[\hat{H}_q, \rho_q] + \kappa_g (\hat{\sigma}_q \rho_q \hat{\sigma}_q - \rho_q), \] (G3)
with \( \hat{H}_q = E(x) \hat{\sigma}_z, \kappa_g = g^2 \kappa_b |4E(x)^2 + \kappa_b^2/4| \). Note that we have set \( \hbar = 1 \) and will do so from here onwards. In this case, the state of the qubit will be an equal mixture of \( |+\rangle \) and \( |-\rangle \) or \( |g\rangle \) and \( |e\rangle \).

Case (b)—In this case, the standard Born-Markov approximation yields the following master equation:
\[ \dot{\rho}_q = -i[\hat{H}_q, \rho_q] \]
\[ + \kappa_g \left( |+\rangle \langle +| \rho_q |+\rangle \langle +| - \frac{1}{2} |+\rangle \langle +| \rho_q |+\rangle \langle +| + \frac{1}{2} |\rangle \langle \rangle \rho_q |\rangle \langle \rangle + |\rangle \langle \rangle \rho_q |\rangle \langle \rangle \right). \] (G4)

Unlike in case (a), here, the steady state of the qubit will be the pure state \(|+\rangle \). The difference between the master equations Eqs. (G3) and (G4) can be understood by observing that when \( 2|\Delta_b + E(x)| \gg \kappa_b \gg |2\Delta_b - E(x)| \), then the transition between \(|+\rangle \otimes |0\rangle \) and \(|-\rangle \otimes |1\rangle \) becomes more likely than that between \(|+\rangle \otimes |1\rangle \) to \(|-\rangle \otimes |0\rangle \) because the former, happening at frequency \( 2\Delta_b - E(x) \), lies within the bandwidth \( \kappa_b \). The first term in the tensor product refers to the state of the qubit and the second term refers to the vacuum \( |0\rangle \) and single photon Fock state \(|1\rangle \) of the oscillator mode. Consequently, if the photon is subsequently lost from the oscillator, the qubit will be preferentially projected to the state \(|-\rangle \).

Thus, the steady-state value of \( \langle \hat{\sigma}_x \rangle \) is 0 for case (a) and -1 for case (b). Clearly, we see that the qubit dynamics depends on the parameters \( \{E(x), \Delta_b, g, \kappa_b\} \), and in certain cases can have a steady-state value of \( \langle \hat{\sigma}_x \rangle \) between 0 and -1.

We note here that by considering our full model [Eq. (3) of the main text] with not only dissipation on the coupling mode \( \kappa_b \) but also dissipation on the tuning mode \( \kappa_g \), we can expect a more complex landscape of steady-state dynamics which may qualitatively fall into the above regime.

2. \( E(x) = 0 \)

In this case, we follow the approach in Ref. [41] and begin by writing the density matrix of the qubit and oscillator as
\[ \dot{\rho} = |g\rangle \langle g| \otimes \dot{\rho}_{gg} + |e\rangle \langle e| \otimes \dot{\rho}_{ee} + |g\rangle \langle e| \otimes \dot{\rho}_{ge} |e\rangle \langle g| \] (G5)
where \(|g,e\rangle\) represent the qubit states and \( \dot{\rho}_{ij} \) acts on the oscillator Hilbert space conditioned on the qubit state. From Eqs. (G1) and (G2) with \( E(x) = 0 \) and Eq. (G5), we get
\[ \dot{\rho}_{ge} = -ig(\hat{b}^\dagger + \hat{b})\rho_{ge} - ig\hat{g}e(\hat{b}^\dagger + \hat{b}) \]
\[ - i\Delta_b (\hat{b}^\dagger \hat{b} \rho_{ge} - \rho_{ge} \hat{b}^\dagger \hat{b}) + \kappa_b \hat{b} \hat{g}e \hat{b}^\dagger \]
\[ - \frac{\kappa_b}{2} \hat{b} \hat{b} \rho_{ge} - \frac{\kappa_b}{2} \hat{b} \hat{b} \rho_{ge} \hat{b}^\dagger. \] (G6)

Next, we use the positive-P representation \( \dot{\rho}_{ge} = \int \mathcal{P}(\alpha, \alpha^*, \xi) \hbar d\alpha d\alpha^* d\xi \) and write the effective Fokker-Planck equation:
\[ \frac{\partial P}{\partial \xi} = -2igP(\alpha + \alpha^*) + i(g + \Delta_b \alpha) \frac{\partial P}{\partial \alpha} \]
\[ + i(g - \Delta_b \alpha^*) \frac{\partial P}{\partial \alpha^*} + \kappa_b P \frac{\kappa_b \alpha - \kappa_b \alpha^*}{2} \frac{\partial P}{\partial \alpha} \]
\[ + \kappa_b \alpha^* \frac{\partial P}{\partial \alpha^*}. \] (G7)

Now we must solve the above equation with some given initial condition. In the setup of interest we start with the
oscillator mode in vacuum so that \( P(\alpha, \alpha^*, 0) = \delta^2(\alpha) = \lim_{x \to 0} (1/x^2) \exp[-|\alpha|^2/x] \) and the qubit in the state \(|+\rangle\). We can make the Gaussian ansatz, \( P(\alpha, \alpha^*, t) = \exp[-a(t) + b(t)\alpha + c(t)\alpha^* - d(t)|\alpha|^2] \) and write the equivalent differential equations for \( a, b, c, d \) to get

\[
\dot{a} = igb + igc + \kappa_b, \\
\dot{d} = -\kappa_b d, \\
\dot{b} = -2ig + \left( i\Delta_b + \frac{\kappa_b}{2} \right) b - igd, \\
\dot{c} = -2ig + \left( -i\Delta_b + \frac{\kappa_b}{2} \right) b - igd. \\
\tag{G8}
\]

These time-dependent equations can be easily solved with initial conditions now written as \( a(0) = \ln x \), \( b(0) = 0 \), \( c(0) = 0 \), \( d(0) = 1/x \), but the analytic expressions are considerably simplified when \( \Delta_b = 0 \). Once we get \( a, b, c, d \) we are able to reconstruct \( P \) and hence \( \hat{\rho}_{qe} \).

The relevant quantity of interest is the time dependence of the expectation value of the operator \(|g\rangle\langle e|\):

\[
\langle |g\rangle\langle e| (t) \rangle = \text{Tr}[\hat{\rho}_{qe}] = \int P(\alpha, \alpha^*, t) d^2 \alpha. \\
\tag{G9}
\]

For \( \Delta_b = 0 \) and an initial condition where the qubit is in \(|+\rangle\) and the oscillator in vacuum, we have

\[
\langle |g\rangle\langle e| (t) \rangle = \frac{1}{2} \exp \left[ -\frac{8\gamma^2 t}{\kappa_b} + \frac{16\gamma^2 t}{\kappa_b} (1 - e^{-\kappa_b t/2}) \right]. \\
\tag{G10}
\]

Next, we consider the rate equation for \( \hat{\rho}_{gg} \):

\[
\dot{\hat{\rho}}_{gg} = -ig(\hat{b}^\dagger + \hat{b})\hat{\rho}_{gg} + ig\hat{\rho}_{gg}(\hat{b}^\dagger + \hat{b}) \\
- i\Delta_b (\hat{b}^\dagger\hat{b}\hat{\rho}_{gg} - \hat{\rho}_{gg}\hat{b}^\dagger\hat{b}) + \kappa_b \hat{b}^\dagger \hat{b} \hat{\rho}_{gg} - \frac{\kappa_b}{2} \hat{b}^\dagger \hat{b}\hat{\rho}_{gg} \\
- \frac{\kappa_b}{2} \hat{\rho}_{gg} \hat{b}^\dagger \hat{b}. \\
\tag{G11}
\]

Like before, we use the positive-\( P \) representation \( \hat{\rho}_{gg} = \int P(\alpha, \alpha^*, t) d\alpha d\alpha^* \), so that

\[
\frac{dP}{dt} = -ig \frac{\partial P}{\partial \alpha^*} + ig \frac{\partial P}{\partial \alpha} - i\Delta_b \frac{\partial P}{\partial \alpha^*} + i\Delta_b \frac{\partial P}{\partial \alpha} \\
+ \kappa_b \alpha \frac{\partial P}{\partial \alpha} + \kappa_b \alpha^* \frac{\partial P}{\partial \alpha^*}. \\
\tag{G12}
\]

We can again make the Gaussian ansatz \( P(\alpha, \alpha^*, t) = \exp[-a(t) + b(t)\alpha + c(t)\alpha^* - d(t)|\alpha|^2] \), and solve the corresponding differential equations for \( a, b, c, d \) to get \( P(\alpha, \alpha^*, t) = \delta^2(\alpha - \alpha(t)) \), where \( \alpha(t) = 2ig[1/2 - \exp(-\kappa_b t/2)]/\kappa_b \) (for \( \Delta_b = 0 \)). Thus, \( \langle |g\rangle\langle g| (t) \rangle = 1/2 \).

Similarly we can show that \( \langle |e\rangle\langle e| (t) \rangle = 1/2 \). Hence the diagonal terms of the qubit density matrix do not decay with time—only the off-diagonal terms do. Thus, in this case the qubit undergoes pure dephasing due to its coupling with the oscillator mode and at steady state becomes an equal mixture of \(|g\rangle\) and \(|e\rangle\).


