Direct observation of geometric phase in dynamics around a conical intersection

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Conical intersections are ubiquitous in chemistry, often governing processes such as light harvesting, vision, photocatalysis, and chemical reactivity. They act as funnels between electronic states of molecules, allowing rapid and efficient relaxation during chemical dynamics. In addition, when a reaction path encircles a conical intersection, the molecular wavefunction experiences a geometric phase, which affects the outcome of the reaction through quantum-mechanical interference. Past experiments have measured indirect signatures of geometric phases in scattering patterns and spectroscopic observables, but there has been no direct observation of the underlying wavepacket interference. Here, we observe geometric-phase interference in the dynamics of a nuclear wavepacket travelling around an engineered conical intersection in a programmable trapped-ion quantum simulator. To achieve this, we develop a new technique to reconstruct the two-dimensional wavepacket densities of a trapped ion. Experiments agree with the theoretical model, demonstrating the ability of analog quantum simulators—such as those realised using trapped ions—to accurately describe nuclear quantum effects. These results demonstrate a path to deploying analog quantum simulators for solving some of the most difficult problems in chemical dynamics.

Light drives molecular processes as important as photosynthesis, photocatalysis, and vision. Absorbing a photon promotes a molecule to an excited electronic state, triggering chemical dynamics and reactivity. The molecule will eventually return to the ground state; often, this relaxation happens on ultrafast (fs-ps) timescales at molecular geometries where two electronic energy surfaces have the same energy, known as conical intersections [1, 2]. By acting as funnels between electronic states for the molecular wavefunction, conical intersections enable rapid non-radiative electronic transitions and have a decisive role in chemical dynamics, from charge-transfer processes to photochemical reactions [3].

The path taken during molecular dynamics involving conical intersections can profoundly alter chemical reaction outcomes. In particular, a geometric phase [4] causes quantum interference of nuclear wavepackets encircling a conical intersection [5–7]. Accounting for geometric phase is necessary in quantum chemistry calculations because the resulting interference changes the ratio of reactive and non-reactive outcomes in scattering cross-sections [8–11] and alters vibrational spectra [12–14]. Indeed, recent experiments have detected indirect signatures of geometric phase in reactive scattering [15, 16]. An elegant proposal for revealing spectroscopic signatures of geometric phase involves interference signals from pairs of excitation pulses [17, 18], but it remains unimplemented due to challenging state preparation.

An unambiguous observation of geometric phase remains an outstanding challenge in chemical physics. It requires full reconstruction of nuclear wavepacket dynamics on ultrafast timescales, which is possible in small molecules [19], but has never been used to characterise geometric phase. Analog quantum simulators present a new opportunity to access chemical dynamics on laboratory-accessible timescales [20–23]. In such systems, a one-to-one correspondence between the degrees of freedom of the chemical system and those of the simulator makes it possible to replicate the target dynamics in a controllable and measurable manner.

Several controllable quantum systems have been proposed to engineer conical intersections and study signatures of geometric phase. Theoretical proposals have included using trapped Rydberg ions to simulate electronic populations [24], circuit quantum electrodynamics to simulate emission spectra [25], and cavity quantum electrodynamics to simulate collapse-revival characteristics of a spreading nuclear wavepacket [26]. To date, the only experimental implementation of a conical intersection in a quantum simulator was used to demonstrate branching between different photochemical reaction products with strong dissipation [27].

Here, we present the first observation of the destructive interference of a nuclear wavepacket caused by geometric phase during dynamics around a conical intersection. We implement a controllable conical intersection by engineering a Jahn-Teller Hamiltonian in a trapped-ion quantum simulator that employs a mixed-qudit-boson (MQB) encoding to directly simulate both electronic and nuclear degrees of freedom [28]. Our experiment introduces a resource-efficient reconstruction method to image the wavepacket’s nuclear density, directly showing the destructive interference which is a signature of wavepacket dynamics near a conical intersection. Experimental measurements match theoretical predictions, demonstrating the utility of quantum simulators to give insights into properties that have otherwise been impossible to measure directly for chemical systems.

In an MQB simulator [28] the electronic and vibrational degrees of freedom of the molecule are represented in a qudit and a set of bosonic modes. Our simulator is realised using an $^{171}$Yb$^+$ ion confined in a Paul trap, where the two vibrations are encoded directly in the ion’s transverse vibrational modes (B$_1$ and B$_2$), while the two
The minimum of conical intersection can be directly observed from the vibronic coupling strength, and ionization position coordinate for the electronic modes, described by the potential energy model of geometric-phase effects in molecules [30]. It has recently been employed to predict molecular spectra using time-domain simulations [29], and provides resource-scaling advantages relative to conventional methods [28].

To demonstrate geometric-phase interference, we implement the \( E \otimes e \) Jahn-Teller model [5], a standard model of geometric-phase effects in molecules [30]. It consists of two electronic states coupled with two vibrational modes, described by the potential energy

\[
V_{JT} = \frac{\omega}{2} (Q_1^2 + Q_2^2) + \kappa (\sigma_z Q_1 + \sigma_x Q_2),
\]

where \( \sigma_x \) and \( \sigma_z \) are the Pauli matrices acting on the electronic states and \( Q_j = (a_j^+ + a_j)/\sqrt{2} \) is the dimensionless position coordinate for the \( j \)th vibrational mode, with creation and annihilation operators \( a_j^+ \) and \( a_j \). \( \kappa \) is the vibronic coupling strength, and \( \omega \) is the frequency of both vibrational modes. The Jahn-Teller Hamiltonian is given by \( H_{JT} = \omega (P_1^2 + P_2^2)/2 + V_{JT} \), where \( P_j \) is the conjugate momentum of \( Q_j \). We set \( h = 1 \) throughout.

Diagonalisation of \( V_{JT} \) in the electronic basis leads to cylindrically symmetric potential energy surfaces along \( Q_1 \) and \( Q_2 \), with energies

\[
E_{\pm} = \omega (Q_1^2 + Q_2^2)/2 \pm \kappa \sqrt{Q_1^2 + Q_2^2}
\]

(see fig. 1). The conical intersection is present at the point of highest symmetry (\( Q_1 = Q_2 = 0 \)), where the two potential energy surfaces are degenerate. The minimum of \( E_- \) occurs where \( Q_1^2 + Q_2^2 = (\kappa/\omega)^2 \).

The effects of geometric phase on dynamics around a conical intersection can be directly observed from the nuclear probability density, fig. 1a–d. As the initial wavepacket, we choose the ground state of the non-interacting vibrational Hamiltonian, \( H_0 = \omega (a_1^+ a_1 + a_2^+ a_2) \), displaced to the potential-energy minimum at \( Q_1 = -\kappa/\omega, Q_2 = 0 \) (fig. 1a–b). During the time evolution, the wavepacket splits into two components evolving in opposite directions around the conical intersection. The two components overlap at \( Q_1 > 0 \), causing destructive interference at the nodal line \( Q_2 = 0 \), where their equal and opposite geometric phases lead to the vanishing of the nuclear density (fig. 1c–d). By contrast, if geometric phase were disregarded, the two wavepacket fragments would interfere constructively, reaching maximum amplitude at \( Q_2 = 0 \) (fig. 1e–f).

To map the Jahn-Teller model onto the MQB simulator, we rewrite \( H_{JT} \) in the interaction picture with respect to \( H_0 \),

\[
H_{JT}^{\text{int}} = \frac{\kappa}{\sqrt{2}} \sigma_z (a_1^+ e^{i\omega t} + a_1 e^{-i\omega t}) + + \frac{\kappa}{\sqrt{2}} \sigma_x (a_2^+ e^{i\omega t} + a_2 e^{-i\omega t}),
\]

which can be implemented using tunable light-atom interactions to enact quibit-boson couplings. We achieve this using a coherent state-dependent force (SDF) enacted by stimulated Raman transitions driven with a 355 nm pulsed laser [31, 32]. Driving transitions near bosonic mode \( j \) leads to the Hamiltonian

\[
H_{\text{SDF}}^j (\delta, \Omega, \phi_m) = \frac{\Omega}{2} \sigma_{\phi_m}(a_j^+ e^{i(\phi_m + \delta t)} + a_j e^{-i(\phi_m + \delta t)}),
\]

Figure 1. Directly detecting a geometric phase through wavepacket interference. a, A nuclear wavepacket is initially displaced to the minimum of the potential energy surface, after which it begins to encircle the conical intersection, denoted \( \Sigma \). b, Initial nuclear density in 2D (left), and integrated over \( Q_1 \) (right). c, After sufficient time evolution, the two components of the wavepacket destructively interfere due to geometric phase, giving a nodal line along \( Q_2 = 0 \) (dotted line). d, Nuclear density at the maximum interference time \( T \). e, If the geometric phase were neglected, the two wavepacket components would interfere constructively. f, Nuclear density at \( t = T \) with geometric phase neglected. Contours in b, d, and f correspond to the potential energy surface \( E_- \). g, The Jahn-Teller Hamiltonian \( H_{JT}^{\text{int}} \) is engineered in an ion-trap quantum simulator with a single \( ^{87}\text{Rb} \) ion. The ion (white sphere) is confined in a Paul trap and \( H_{JT}^{\text{int}} \) is realised using two simultaneous laser-induced interactions (purple and pink, corresponding to colour-coded terms in \( H_{JT}^{\text{int}} \)).
where \( \sigma_{\phi_0} = \sigma_x \cos \phi_0 + \sigma_y \sin \phi_0 \) and \( \phi_0 \) and \( \phi_m \) are the phases associated with the qubit and the bosonic mode, respectively (see Methods). \( \Omega \) and \( \delta \) are the Rabi frequency and detuning of the laser from the bosonic mode, respectively. We use the notation \( H^{\text{SDF}}_{\pm} \) and \( H^{\text{SDF}}_{\pm,\beta} \) for SDF interactions where \( \phi_0 = 0 \) and \( \phi_0 = \pi/2 \), respectively. Interactions in the \( \sigma_z \) basis are obtained using a qubit basis rotation, \( H^{\text{SDF}}_{\pm} = R_z(\pi/2)H^{\text{SDF}}_{\pm,\beta} R_z(-\pi/2) \), where \( R_z(\theta) \) are driven qubit rotations around the Bloch sphere. \( H^{\text{JT}} \) can then be implemented in a programmable way using two simultaneous SDFs (see fig. 1g),

\[
H^{\text{JT}}_1 = H^{\text{SDF}}_{1,\pm}(\omega, \sqrt{2} \kappa, 0) + H^{\text{SDF}}_{2,\pm}(\omega, \sqrt{2} \kappa, 0).
\]

The parameters \( \kappa \) and \( \omega \) are chosen to produce a clear wavepacket interference. To achieve this, \( \kappa/\omega \) should be large enough that the wavepacket prepared at the minimum of the potential energy surface (at \( Q_1 = -\kappa/\omega \)) has negligible overlap with the conical intersection. However, \( \kappa/\omega \) should also be kept small enough to mitigate vibrational decoherence that increases with larger vibrational excitations. To balance these considerations, we choose \( \kappa/\omega = 1.5 \), for which the wavepacket has only 1.7% of the density at \( Q_1 \geq 0 \). \( \kappa \), implemented by adjusting the Rabi frequency, is maximised to increase the speed of the dynamics; its value is constrained by the available SDF-laser power to \( \kappa = 2\pi \times 1.00 \text{kHz} \), yielding \( \omega = 2\pi \times 667 \text{ Hz} \). With these parameters, the wavepackets are expected to experience the greatest geometric-phase interference at \( T = 1.59 \text{ ms} \).

We probe the dynamics of the geometric phase around the conical intersection by reconstructing the nuclear probability densities at different evolution times \( t \). The experimental sequence consists of four stages, shown in fig. 2. (i) Preparation of the qubit and cooling of the vibrational modes to their ground states is achieved by optical pumping, Doppler cooling, and sideband cooling. (ii) Initialisation consists of displacing \( B_1 \) to \( Q_1 = -\kappa/\omega \) by applying an SDF interaction \( H^{\text{SDF}}_{1,\pm}(0, \Omega_1, \pi/2) \) for a duration \( \tau \). This applies the displacement operator \( D_1(-\Omega_1 \tau/2) \). (iii) Evolution of the system under \( H^{\text{JT}}_1 \) is achieved by applying the two simultaneous SDF interactions of eq. (4) for an experimentally variable duration \( t \). (iv) Reconstruction of the joint densities of \( B_1 \) and \( B_2 \) is achieved by measuring the characteristic function

\[
\chi(i\beta_1, i\beta_2) = \langle \Psi | D_1(i\beta_1) D_2(i\beta_2) | \Psi \rangle,
\]

where \( | \Psi \rangle \) is the total wavefunction of the system, and \( \beta_1 \) and \( \beta_2 \) are real numbers. See Methods for details.

The joint probability densities are reconstructed using the circuit in fig. 2. Two SDF pulses are sequentially applied on \( B_1 \) and \( B_2 \), and \( \chi(i\beta_1, i\beta_2) \) is scanned over \( \beta_1, \beta_2 \). These measurements yield the joint probability density via the Fourier transform of the measured characteristic function

\[
|\Psi(Q_1, Q_2)|^2 = \iint \frac{d\beta_1 d\beta_2}{2\pi} e^{-i\chi(Q_1 \beta_1 + Q_2 \beta_2)} \chi(i\beta_1, i\beta_2).
\]

In further detail, we measure \( \chi(i\beta_1, i\beta_2) \) by mapping information from the multimode bosonic system onto the qubit using SDF pulses, moving beyond previous works on direct single-mode [33–36] and indirect multimode reconstructions [37]. The reconstruction consists of preparing the qubit in \( |\downarrow\rangle \) and applying two successive SDF interactions, \( H^{\text{SDF}}_{1,\pm}(0, \Omega_2, 0) \) and \( H^{\text{SDF}}_{2,\pm}(0, \Omega_2, 0) \) with durations \( \tau_1 \) and \( \tau_2 \). Doing so results in controlled displacements \( D_1(i\beta_1/2) \) and \( D_2(i\beta_2/2) \), where \( \beta_j = \Omega_j \tau_j \), \( \chi(i\beta_1, i\beta_2) \) is measured for different values of \( \beta_1 \) and \( \beta_2 \).
β2 by varying τ1 and τ2. We reconstruct the characteristic functions of the bosonic modes entangled with the |↓⟩ and |↑⟩ qubit states independently. Reconstructing the |↓⟩ component is achieved by adding a mid-circuit measurement which projects out the |↑⟩ component (see Methods). The experiment is repeated with an additional $R_x(\pi)$ pulse prior to the mid-circuit measurement to reconstruct the |↑⟩ component. The qubit probabilities $p_1$ and $p_1$ are calculated from the success rate of the mid-circuit measurement. After the displacements, measuring the qubit in the $\sigma_z$ basis gives the real part of the characteristic function, $\langle \sigma_z \rangle = \text{Re} \chi(i\beta_1, i\beta_2)$. Repeating the experiment with an additional $R_x(\pi/2)$ pulse prior to the displacements gives the imaginary part, after which the full $\chi(i\beta_1, i\beta_2)$ is obtained by adding the real and imaginary parts associated with both |↓⟩ and |↑⟩.

The reconstructed probability densities in fig. 3a-d demonstrate a direct measurement of the wavepacket interference caused by the nuclear geometric phase. At $t = 0$, the initial wavepacket is prepared at $(Q_1, Q_2) = (-1.5, 0)$. As the wavepacket evolves around the conical intersection, the nodal line becomes visible at $Q_2 = 0$ and is most pronounced at $t = T$; this is a direct observation of destructive interference due to geometric phase. Finally, at $t = 2T$, the two wavepackets recombine close to their initial position. The experimental results agree well with theoretical predictions, reproducing key features of interference and wavepacket recombination.

Further quantitative insight may be gained from the 1-dimensional density $|\Psi(Q_2)|^2$, obtained by omitting the $D_1(i\beta_1)$ displacements from the reconstruction procedure above. In this case, the measurements scanned over $\beta_2$ are Fourier-transformed to give $|\Psi(Q_2)|^2 = \int d\beta_2 e^{-iQ_2\beta_2} \chi(i\beta_2) / \sqrt{2\pi}$. In fig. 3e, we present $|\Psi(Q_2)|^2$ for seven different evolution times. A comparison of experiment and theory shows excellent agreement in the shape and amplitude of the measured density function. We attribute minor discrepancies to the dephasing of the bosonic modes, miscalibrations such as uncompensated AC Stark shifts, and technical imperfections in the protocol implementation.

Our approach avoids the limitations of direct experiments on molecular systems, where only a few observables—such as spectra and scattering cross sections—can be measured. Instead, a fully controllable quantum device, such as an ion-trap MQB simulator [28], can, in principle, read out any observable, including, as we showed here, the full two-dimensional density of the $^{171}$Yb$^+$ ion as it moves in space and time. A further advantage is the ability to slow down femtosecond molecular dynamics to the millisecond trapped-ion timescale, a scaling of about 10 orders of magnitude [29]. This enormously improves the achievable resolution of chemical-dynamics measurements relative to ultrafast observations.

A key general feature of quantum simulations is their programmability [38]. In an MQB simulator, the qudit-boson interaction is fully controllable, meaning that the same device can be programmed to simulate many molecular systems or even theoretical models that do
not occur naturally in any molecular system, simply by changing experimental parameters. In particular, our geometric-phase simulator could be used to simulate dynamics in molecules with conical intersections where the interactions are not as symmetric as in $H_{1T}$, such as the general quadratic vibronic-coupling Hamiltonian [28].

Like any analog simulation—quantum or classical—our approach is ultimately limited by noise and uncorrected errors. In our experiment, the main sources of decoherence and dissipation are motional dephasing and motional heating [29, 39]. However, in MQB simulations of molecular processes, noise can be characterised and even amplified in order to create a realistic model of molecular environments, such as collisions in solution. Since our Jahn-Teller experiment shows only weak effects of decoherence over the full period $2T$, we would need to inject additional noise to simulate conical-intersection dynamics that are otherwise unmeasurable directly according to the displacement are adjusted by varying $\alpha$ and $\tau$. The bosonic modes are cooled in two stages. First, they are Doppler cooled using a laser red-detuned from the 2$^3S_{1/2}$ ground state. The two Raman beams are orthogonal to one another, and configured so that they can be coupled to both radial vibrational modes. Each Raman beam passes through an acousto-optical modulator (AOM), which allows the phase, frequency and amplitude of the beam to be adjusted by altering the RF signal driving the AOM. One of the RF signals is generated by an arbitrary waveform generator (Keysight M8190A), allowing multiple phase-coherent tones to be imprinted on one of the laser beams. We ensure phase coherence between all pulses in the experimental sequence by tracking the phase (relative to the beginning of the pulse sequence) and applying appropriate corrections (detailed in appendix A2).

By tuning the frequency difference of the Raman beams, one can drive carrier, red- and blue-sideband transitions (see appendix A2). Qubit rotations are obtained by driving carrier transitions, while an SDF $H_{\phi_{m}}^{SD}(0, \Omega, m)$ arises from combining the red- and blue-sideband transitions. Applying this interaction for a duration $\tau$ with the qubit in an eigenstate of $\sigma_{\phi}$, displaces bosonic mode $j$ by $D_{j}(\alpha) = \exp(\alpha a_{j}^{\dagger} - \alpha^{*} a_{j})$, where $\alpha = -i \Omega \tau e^{i \phi_{m}}/2$. The amplitude and phase-space direction of the displacement are adjusted by varying $\tau$ and $\phi_{m}$, respectively.

Experimental protocol

Preparation. The bosonic modes are cooled in two stages. First, they are Doppler cooled using a 369.5 nm laser red-detuned from the $2^3S_{1/2} \rightarrow 2^3P_{1/2}$ transition. Second, resolved sideband cooling is used to reach their motional ground states, achieving temperatures of $\bar{n} = 0.04$ measured via sideband thermometry [42]. The qubit is prepared in its ground state $|\downarrow\rangle$ via optical pumping, using another 369.5 nm laser resonant with the $2^3S_{1/2}$ $|F = 1\rangle \rightarrow 2^3P_{1/2} |F = 1\rangle$ transition.

Initialisation. To initialise B$_1$, we apply an SDF interaction $H_{\phi_{m}}^{SD}(0, \Omega, \pi/2)$ for a duration $\tau$, which gives a displacement $D_{1}(\alpha)$ where $\alpha = \Omega \tau/2$. Setting $\tau = \sqrt{\bar{n}}/\omega_{1}$ so that $\alpha = \kappa/\sqrt{\bar{n}}$ displaces the mode.
We measured $\chi$ with an $t$-ion. Since the characteristic function is Hermitian, $\beta$ for $t$-ion resulting in combined pulse durations of up to experiments and, on average, $\beta$ measured as described in the main text. The full characteristic function projects the qubit state to either $|\alpha\rangle$ or $|\beta\rangle$ through state-dependent fluorescence induced by a $369.5 \text{ nm}$ laser beam resonant with the $|F = 1\rangle \rightarrow |F = 0\rangle$ transition. The qubit states are inferred by thresholding the number of photons collected on an avalanche photodiode (measured state preparation and measurement fidelity of 99.5%), and the outcomes of the measurement determine the probabilities $p_2 = |\alpha|^2$ and $p_1 = |\beta|^2$. A measurement outcome of $|\beta\rangle$ induces significant decoherence of the bosonic modes due to photon recoils. Therefore, the reconstruction only proceeds if the measurement outcome is $|\alpha\rangle$, for which no photon is emitted. Doing so projects the bosonic modes to $|\psi_i\rangle_{B_1} |\xi_i\rangle_{B_2}$. To retrieve $|\psi_f\rangle_{B_1} |\xi_f\rangle_{B_2}$ instead, we insert an $R_x(\pi)$ pulse that flips the qubit before the measurement. After the mid-circuit measurement, the characteristic functions $\chi_{i}(i\beta_1, i\beta_2)$ and $\chi_{j}(i\beta_1, i\beta_2)$ corresponding to each qubit state are measured as described in the main text. The full characteristic function is then the sum of both contributions, $\chi(i\beta_1, i\beta_2) = p_1 \chi_{i}(i\beta_1, i\beta_2) + p_2 \chi_{j}(i\beta_1, i\beta_2)$. The values $\beta_1 = \Omega T \tau$ are scanned by varying the SDF-pulse duration $\tau$. The Rabi frequency was recalibrated between experiments and, on average, $\Omega = 2\pi \times 2.31 \text{ kHz}$, resulting in combined pulse durations of up to $553 \mu s$. We measured $\chi(i\beta_2)$ for $\beta_2 \in [0, 5]$ and $\chi(i\beta_2)$ for $\beta_1, \beta_2 \in [0, 4]$ by varying the SDF pulse duration. Since the characteristic function is Hermitian, $\chi(i\beta_1, i\beta_2)^* = \chi(-i\beta_1, -i\beta_2)$, we used symmetry to find $\chi(i\beta_2)$ for $\beta_2 < 0$ and $\chi(i\beta_1, i\beta_2)$ for $\beta_1 < 0$ or $\beta_2 < 0$. We did not measure the vanishing imaginary part of $\chi(i\beta_2)$ nor $\chi_j$ at $t = 0$. The measured characteristic functions are shown in appendix A1.

**Data acquisition.** The characteristic functions were measured in a way to average out the effects of drift. In each run of the experiment, we randomised the order of the displacement-pulse durations in which $\chi$ was reconstructed. For each run, the quantum circuit to obtain $\chi_\downarrow$ and $\chi_\uparrow$ was repeated until the mid-circuit measurement succeeded 500 times, resulting in 500 measurement repetitions of the reconstruction routine and 1000 measurements to obtain a value of the full $\chi$. Furthermore, the order of the displacements was randomised. Overall, each of the 1- and 2-dimensional experiments was repeated, respectively, four and two times and the results of the runs averaged for a total of 2000 and 1000 measurements for each duration. The bosonic mode frequencies were calibrated every 6 min, while the full system parameters were recalibrated after the second experimental run (see appendix A3 for more details). The 1-dimensional and the four 2-dimensional ($t = \{0, 0.9T, T, 2T\}$) experiments were done on five separate days with total durations of 15.6, 2.8, 8.8, 8.7 and 10.6 hours, respectively.

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APPENDICES

Appendix A1: Characteristic functions

Figure A1 shows the measured characteristic functions used to reconstruct the wavepacket probability densities. The two-dimensional characteristic functions (fig. A1a–d) require four measurements at each t to obtain the real and imaginary parts of p(t)(i β1, i β2) and p(t)χ(i β1, i β2). The one-dimensional characteristic functions (fig. A1c) require two measurements to determine p(t)χ(i β2) and p(t)χ(i β2), as the vanishing imaginary part is not measured. In both one- and two-dimensional reconstructions, only positive values of β1 and β2 are sampled; the characteristic function in other ranges is
Characteristic functions of the wavepacket measured for various evolution times. a–d Joint two-dimensional characteristic function \(\chi(i\beta_1, i\beta_2) = p_1 \chi_1(i\beta_1, i\beta_2) + p_2 \chi_2(i\beta_1, i\beta_2)\) measured at times \(t = \{0, 0.9T, 2T\}\) using the full pulse sequence of fig. 2. The real (left) and imaginary (right) parts are measured with and without an \(R_x(\pi/2)\) pulse in the reconstruction. The top row shows theoretical predictions and the bottom experimental results. \(\chi_1(i\beta_2)\) and \(\chi_2(i\beta_2)\) were measured in the range \(\beta_1, \beta_2 \in [0, 4]\) with \(11 \times 11\) equidistant samples (dashed quadrant). Values in the remaining three quadrants are obtained from the symmetry of \(\chi(i\beta_1, i\beta_2)\). e One-dimensional characteristic functions \(\chi_1(i\beta_2)\) and \(\chi_2(i\beta_2)\) obtained by omitting displacements on \(B_1\) in the reconstruction. \(\beta_2\) was uniformly sampled in the range \([0, 5]\) with 26 points. Each two- and one-dimensional characteristic function was averaged over 1000 and 2000 measurements, respectively. Error bars in e represent one standard deviation based on quantum projection noise.

Appendix A2: Phase coherence in the pulse sequence

This appendix describes the experimental procedure to track the qubit and motional phases, ensuring phase coherence between sequential spin-motional interactions.

The laser-induced excitations interacting with an ion with a qubit frequency \(\omega_0\) and a motional mode with frequency \(\omega_m\) are the carrier (c), red-sideband (rsb), and blue-sideband transitions (bsb). Their interaction Hamiltonians, after dropping high-frequency terms, are

\[
H_c = \frac{\Omega_c}{2} \sigma^+ e^{i\phi_c} e^{i(\omega_0 - \omega_0)t} + \text{h.c.}, \quad (A1)
\]

\[
H_{rsb} = \frac{\eta \Omega_{rsb}}{2} \sigma^+ a e^{i\phi_{rsb}} e^{i(\omega_r - \omega_0 + \omega_m)t} + \text{h.c.}, \quad (A2)
\]

\[
H_{bsb} = \frac{\eta \Omega_{bsb}}{2} \sigma^+ a^e e^{i\phi_{bsb}} e^{i(\omega_r - \omega_0 - \omega_m)t} + \text{h.c.}, \quad (A3)
\]

where \(\eta\) is the Lamb-Dicke parameter and \(\Omega_{c,rsb}\) are the respective Rabi frequencies. \(\omega_{c,rsb}\) and \(\phi_{c,rsb}\) correspond, respectively, to the frequency differences and the phase differences of the two orthogonal Raman beams. Simultaneously driving the red- and blue-sidebands with \(\Omega = \eta \Omega_c = \eta \Omega_{rsb}\) gives

\[
H_{SDF} = H_{rsb} + H_{bsb} = \frac{\Omega}{2} \sigma^+ \left[ a e^{i\phi_r} e^{i(\omega_r - \omega_0 + \omega_m)t} + a^e e^{i\phi_{bsb}} e^{i(\omega_r - \omega_0 - \omega_m)t} + \text{h.c.}\right]. \quad (A4)
\]

We consider \(\omega_r\) and \(\omega_0\) to be set near-resonant with the...
Likewise, eq. (A5) and eq. (A6) give the motional detuning as $\delta \omega_m = (\omega_b - \omega_r)/2 - \omega_m$. To avoid unwanted phase lags associated with $\phi_m$, we enforce $(\omega_b - \omega_r)/2 = \omega_m$ for all SDF pulses throughout the circuit, where $\omega_m$ is the experimentally measured motional frequency, whose calibration procedure is detailed in appendix A3. There is an unavoidable phase lag due to the detuning $\delta$ required in the SDF interactions during the time evolution. To correct this, we add a motional phase offset of $(t + \tau_1)\delta$ to the SDF interaction during the initialisation. Furthermore, a motional phase offset of $(t + \tau_1)\delta$ is added to the reconstruction SDF pulses, where $t$ is the duration of the time evolution.

Appendix A3: Calibration of motional frequencies

We used a calibration scheduling routine to recalibrate parameters during each experiment and ensure high-fidelity implementations of the pulse sequence. Moreover, we optimised the scheduler to maximise the experimental duty cycle by analysing the temporal noise behaviours.

The data quality of the reconstructed densities depends on correctly setting the laser frequencies for the motional sideband interactions that enact SDF interactions. The motional frequencies $\omega_1$ and $\omega_2$ associated with B1 and B2 are calibrated as previously reported [29]. To do so, both SDF interactions are applied, but we set the fields associated with $\omega_2$ to be sufficiently off resonant while calibrating $\omega_1$. We prepare the state $|1\rangle [0]_{B1}$ and apply two sequential SDF pulses with a relative phase shift $\phi_m = \pi$. In the absence of frequency errors, the mode returns to its original state and a qubit measurement yields zero population in $|\uparrow\rangle$. However, in the presence of errors, the motion remains entangled with the qubit, giving a non-zero measured probability. The SDF fields’ frequencies are then scanned, and a fit to the measurements yields the correct mode frequency. We repeat this procedure to calibrate $\omega_2$ by setting the SDF field associated with $\omega_1$ to be off resonant.

Figure A2 shows the drifts in the radial mode frequencies over time, which varied in a range of $2\pi \times 2.2$ kHz over 2 days. From numerical simulations, we determined that an error tolerance of about 10% is required for the detuning $(\delta \simeq 2\pi \times 667$ Hz) in the time evolution to obtain adequate results. Given that typical experiments lasted tens of hours, frequent recalibrations of the motional mode frequencies were necessary. To this end, we implemented a scheduling algorithm to interleave calibrations and experiments [43]. The scheduling rate was determined by choosing a time interval for which the Allan deviation was sufficiently small. From fig. A2b, we chose an interval of 6 minutes, corresponding to an Allan deviation of $2\pi \times 26$ Hz and satisfying the required tolerance. We also found highly correlated noise between the radial modes (see fig. A2), suggesting a common noise source (e.g., trap RF amplitude fluctuations). Therefore, to increase the experiment duty cycle, frequency offsets measured on B1 were also used to correct for B2.