

Demonstration of Measurement-Enhanced State Preparation and Erasure Conversion in a Molecular Tweezer Array

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Programmable optical tweezer arrays of molecules are an emerging platform for quantum simulation and quantum information science. For these applications, the reduction and mitigation of errors remain major challenges. In this work, we leverage the rich internal structure of molecules to mitigate two types of errors—internal state preparation and qubit leakage errors. First, we demonstrate robust measurement-enhanced tweezer preparation at a record fidelity using site-resolved error detection followed by tweezer movement. Second, using a new hyperfine qubit encoding well suited for use as a quantum memory, we demonstrate site-resolved detection of qubit leakage errors (erasures) induced by blackbody radiation. This approach constitutes the first demonstration of erasure conversion in molecules, a capability that has found recent interest in quantum error correction. Our work opens the door to new possibilities with molecular tweezer arrays: Measurement-enhanced preparation opens access to mesoscopic defect-free molecular arrays that are important for quantum simulation of interacting many-body systems; erasure conversion in molecular arrays lays the technical groundwork for midcircuit detection, an important capability for explorations in quantum information processing.

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I. INTRODUCTION

Programmable optical tweezer arrays of ultracold molecules are an emerging platform for quantum science [1]. They combine the microscopic detection and control capabilities offered by programmable optical tweezer arrays [2,3] with the richness of molecules such as their complex internal structures and long-ranged and anisotropic interactions [4–6]. These features enable applications in many areas ranging from quantum information processing [7–9] and quantum simulation of novel many-body Hamiltonians [1,10–13] to precision measurement experiments probing fundamental physics beyond the Standard Model [11,13–16].

Starting with the first demonstrations of trapping and high-fidelity detection of single molecules in optical tweezer traps [17–21], recent work has demonstrated highly coherent rotational qubits [6,22,23], observed coherent dipolar interactions between individually held

molecules, and demonstrated on-demand entanglement and two-qubit gates between rotational molecular qubits [24–27]. These works established the building blocks for quantum information processing and quantum simulation with molecular tweezer arrays.

A major experimental challenge for molecular tweezer arrays—and for any quantum platform—is mitigation of errors that occur during the preparation and subsequent evolution of the quantum system. In addition to reducing sources of these errors, a powerful and complementary approach is to mitigate errors via detection and subsequent postselection or feedback. In particular, for programmable optical tweezer arrays of atoms and molecules, site-resolved detection of errors, in some cases, allows their subsequent removal simply through tweezer rearrangement. In previous work with atomic tweezer arrays, site-resolved detection has been used to increase internal state preparation fidelities [28], increase robustness to radiative decays of Rydberg states [28,29], and lower motional temperatures [30]. In recent work with bialkali molecules, site-resolved error detection was used to enhance molecular formation efficiencies and reduce array defect rates [31,32]. In addition to mitigating these classical errors, site-resolved detection of qubit errors with known locations, so-called quantum erasures [33,34], is also useful. In particular, recent work has shown that conversion of qubit errors into

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erasures can substantially reduce overheads and raise thresholds for fault tolerance [35], leading to both theoretical [35–38] and experimental investigations into erasure conversion across a variety of qubit platforms [28–30,39–42].

In this paper, we demonstrate new schemes to mitigate two types of errors in molecular tweezer arrays: state preparation errors and qubit leakage errors. Our general strategy is to leverage the rich internal structure of molecules and site-resolved imaging for local error detection midsequence. Specifically, we encode errors into a set of “bright” internal states whose optical detection minimally affects error-free molecules residing in “dark” internal states.

To mitigate state preparation errors, we demonstrate a robust scheme that detects internal state errors locally and subsequently corrects them via tweezer rearrangement. This process allows us to achieve tweezer preparation fidelities substantially surpassing previous work [24,43]. Crucially, the fidelities achieved allow immediate access to defect-free mesoscopic arrays and open the door to quantum simulation in the many-body regime with molecular arrays.

To mitigate qubit leakage errors, we demonstrate a new scheme that detects qubit leakages midsequence for a new noninteracting qubit encoding suitable for use as a quantum memory. We encode errors in states that can be detected locally while minimally affecting qubit population and coherence, and we show that a significant source of errors arising from absorption of blackbody radiation can be converted into locally detectable errors, i.e., erasures. We show that excision of erasures significantly improves qubit lifetimes and coherence times, constituting the first demonstration of erasure conversion in molecules. Our erasure conversion work adds an important capability to the molecular quantum information processing toolkit. It lays the groundwork for midcircuit measurement and quantum error correction in molecular tweezer arrays.

II. OUR PLATFORM: PROGRAMMABLE OPTICAL TWEezer ARRAYS OF LASER-COOLED MOLECULES

Our experiment uses laser-cooled CaF molecules trapped in a programmable optical tweezer array. A CaF molecular beam is created using a cryogenic buffer gas cell [44], laser slowed [45], and trapped in a magneto-optical trap (MOT) [46,47]. Subsequently, the molecules are laser cooled to sub-Doppler temperatures [48,49], optically trapped [50], and transferred into a rearrangeable 1D array of optical tweezer traps [17,19]. Because of the light-assisted collisions, each tweezer is either empty or holds one molecule. The loading process is stochastic, with a typical loading probability of 20% to 30%.

Molecules in the $X^2\Sigma(v=0, N=1)$ rovibrational manifold [where v (N) denote the vibrational (rotational) quantum number] can be cooled and optically detected

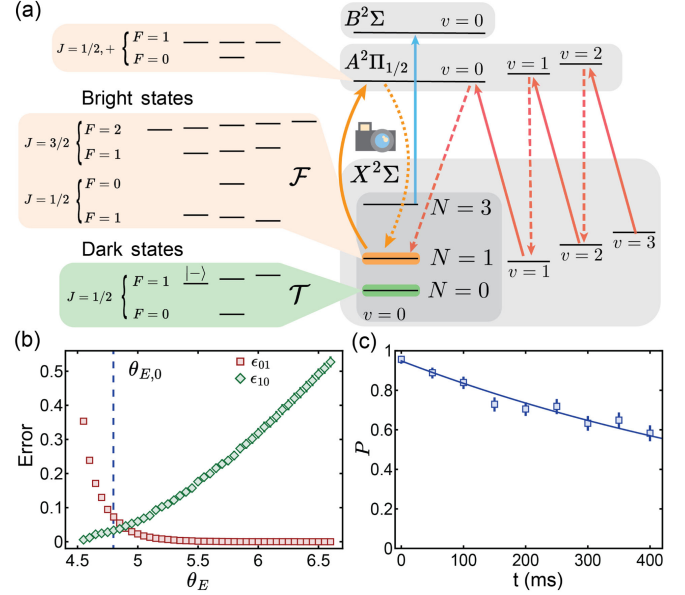


FIG. 1. Site-resolved detection using rapid resonant imaging. (a) Imaging scheme. Molecules in bright states $\mathcal{F} = X^2\Sigma(v=0, N=1)$ scatter light resonant with the $X^2\Sigma(v=0, N=1) \rightarrow A^2\Pi_{1/2}(v=0, J=1/2, +)$ transition (orange arrows). Vibrational (red solid arrows) and rotational repumping light (blue solid arrow) is applied. The imaging light is off resonant for all other states including the ones in the ground rotational manifold $\mathcal{T} = X^2\Sigma(v=0, N=0)$. (b) Error identification performance. The false-positive probability ϵ_{01} (red squares) and false-negative probability ϵ_{10} (green diamonds) are shown versus the classification threshold θ_E . The vertical dashed line marks $\theta_E = \theta_{E,0} = 4.8$, the value used in this work. (c) Population P in $|-\rangle = |X^2\Sigma(v=0, N=0), F=1, m_F=-1\rangle$ versus imaging duration t . An exponential fit gives a $1/e$ lifetime of $\tau = 790(60)$ ms, much longer than the imaging duration of 3 ms. The error bars in panel (b) represent a ± 1 standard deviation and are obtained using a bootstrapping procedure [19]. The error bars in panel (c) represent 68% Wilson-score intervals.

using the $X^2\Sigma(v=0, N=1) \rightarrow A^2\Pi_{1/2}(v=0, J=1/2, +)$ closed optical cycling transition [Fig. 1(a)]. Consequently, molecules occupy all twelve hyperfine states of $X^2\Sigma(v=0, N=1)$ following tweezer loading. To determine each tweezer’s occupation, we apply laser-cooling light on the same cycling transition and collect the emitted fluorescence onto an EMCCD camera. This protocol detects molecules in all $X^2\Sigma(v=0, N=1)$ hyperfine states [Fig. 1(a)].

III. LEVERAGING MOLECULAR STRUCTURE FOR ERROR DETECTION

The first step in our work is to devise a scheme that allows molecules with errors to be detected in a site-resolved manner without affecting error-free ones. To this end, we make use of the rich set of available internal molecular states. We encode errors into a set of bright states ($\mathcal{F}$) that can be optically detected while keeping error-free

molecules in dark states ($\mathcal{T}$) that are minimally affected during detection. Specifically, we make use of bright and dark states whose optical transitions are well separated compared to their linewidths. This strategy is generally possible in a quantum system with sufficiently rich internal structure. For example, work in alkaline-earth atomic arrays [28–30,35] has used ground and metastable electronic manifolds with widely separated optical transitions.

Instead of two different electronic manifolds, our strategy makes use of two different rotational manifolds. The rotational manifold $\mathcal{F} = X^2\Sigma(v=0, N=1)$, which permits optical cycling, is used to flag errors, while error-free molecules reside in the ground rovibrational manifold $\mathcal{T} = X^2\Sigma(v=0, N=0)$ [Fig. 1(a)]. The optical transitions of the bright states in $\mathcal{F}$ are separated from those in the dark states $\mathcal{T}$ by $\Delta \approx 2\pi \times 20$ GHz, which is much larger than the excited state linewidth of $\Gamma \approx 2\pi \times 10$ MHz. Optical detection of errors encoded as population in $\mathcal{F}$ should therefore minimally affect population and coherence in $\mathcal{T}$.

IV. NEW SITE-RESOLVED IMAGING SCHEME TO DETECT ERRORS

Experimentally, we first show that molecules in the bright states $\mathcal{F}$ can indeed be detected while minimally affecting the population of error-free molecules residing in the dark states $\mathcal{T}$. To this end, we devise a new fluorescence detection scheme that is optimized for minimally affecting the dark states. This method is in contrast to previous imaging schemes that prioritize detection fidelity [17,19,48].

In our scheme, we seek to minimize exposure to imaging light in order to minimize off-resonant scattering by error-free molecules in dark states. At the same time, high photon scattering rates of the bright states improve the signal-to-noise ratio of fluorescence detection. We therefore use light resonant with the $X^2\Sigma(v=0, N=1) \rightarrow A^2\Pi_{1/2}(v=0, J=1/2, +)$ transition. This approach provides the best ratio of fluorescence emission to off-resonant scattering and also enables rapid detection compared to previous imaging schemes using detuned laser-cooling light [17,19,48].

We quantify the performance of our detection scheme using two metrics: (1) the classification fidelity and (2) the effect on error-free dark states. To classify a tweezer site as having occupation in $\mathcal{F}$, i.e., having an error, we binarize the fluorescence counts with a threshold $\theta_{E,0}$. Sites with counts above $\theta_{E,0}$ are classified as errors; those below $\theta_{E,0}$ are classified as error-free. The classification fidelity is determined by the false-negative probability ϵ_{10} , where a site with an error is classified as error-free, and the false-positive probability ϵ_{01} , where an error-free site is classified as having an error. Both misclassification probabilities depend on $\theta_{E,0}$. Here, we prioritize minimizing ϵ_{10} rather than the overall fidelity because, for error mitigation via postselection, ϵ_{01} only affects the data rate while ϵ_{10} directly affects the ability to reject errors. After optimization, we find that, at an imaging intensity of

$I = 4.5$ mW/cm², an imaging duration of 3 ms, and a tweezer depth of $k_B \times 930(20)$ μ K, we achieve a false-negative probability $\epsilon_{10}(\theta_{E,0}) = 0.033(4)$ at a threshold of $\theta_E = \theta_{E,0} = 4.8$ [Fig. 1(b)]. This imaging performance allows up to a 30-fold reduction of intrinsic error rates.

Next, we quantify the effect of our imaging procedure on the dark state population. We prepare molecules in one of the dark states $|-\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=1, m_F=-1\rangle$ and measure the population loss versus imaging duration [Fig. 1(c)]. At the imaging duration used (3 ms), we measure a population loss of $3.8(3) \times 10^{-3}$ per image, confirming that the error-free states are minimally affected. The population loss rate is in agreement with theoretical estimates. For our scheme, the ratio of the off-resonant scattering rate to the fluorescence rate is approximately $9\Gamma^2/\Delta^2 \sim 10^{-6}$ (Appendix E). Because the dark states do not optically cycle, they leak to other internal states after $\mathcal{O}(1)$ scattering events. Since we typically scatter about 1000 photons to achieve a sufficient signal-to-noise ratio, the population leakage errors are expected to be at about the 10^{-3} level. These leakage errors could, in principle, be corrected with more elaborate optical pumping and detection schemes.

V. ROBUST AND ENHANCED TWEEZER PREPARATION USING SITE-RESOLVED ERROR DETECTION

Having established that bright molecules in $\mathcal{F}$ can be detected without affecting dark ones in $\mathcal{T}$, we next demonstrate how this finding can be used to detect and correct for internal state preparation errors. In previous work with laser-cooled molecular arrays, the best reported fidelity of having a tweezer loaded with a molecule in a single internal state was approximately 80%, limited by imperfect optical pumping and microwave transfers [24,25]. Here, we present a scheme that detects internal state preparation errors, allowing them to be subsequently removed either midsequence or through postselection.

Our scheme, shown in Fig. 2(a), works as follows. After initial tweezer loading, molecules are optically pumped into a single hyperfine state $|D'\rangle = |X^2\Sigma(v=0, N=1), J=3/2, F=2, m_F=-2\rangle$ within $\mathcal{F}$. Subsequently, molecules are state-selectively shelved into one of the dark states $|-\rangle$ with a microwave pulse at approximately 20.5 GHz. All errors due to imperfect optical pumping and microwave transfer leave a molecule in the bright states $\mathcal{F}$, which is then optically detected with our resonant imaging scheme without substantially affecting the correctly prepared molecules that reside in the dark state $|-\rangle$.

We quantify the performance of our measurement-enhanced state preparation scheme as follows. We first perform internal state preparation followed by error detection, which provides a list of sites that are flagged as having an error. Subsequently, we transfer correctly prepared

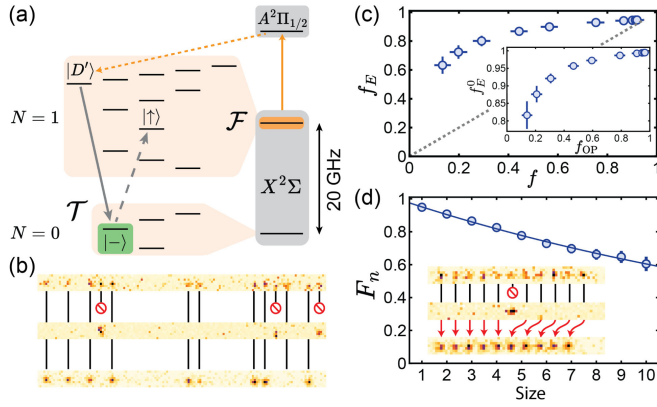


FIG. 2. Robust and enhanced state preparation using error detection. (a) Procedure consisting of the following: (i) optical pumping to $|D'\rangle$ (orange arrow), (ii) population transfer to $|-\rangle$ via a state-resolved microwave pulse (gray solid arrow), and (iii) error detection of leftover molecules in $\mathcal{F}$. To detect the error-free population, a state-selective microwave pulse (gray dashed arrow) transfers $|-\rangle$ to a bright state $|+\rangle$. (b) Verifying tweezer preparation. A nondestructive image (top) identifies initially loaded sites. Measurement-enhanced state preparation is applied, producing an error image (middle) that flags state preparation errors. To verify error detection, we state-selectively transfer molecules from $|-\rangle$ to $|+\rangle$ for detection (bottom). (c) Error-excised overall preparation fidelity f_E (probability of being occupied and in the correct internal state) versus bare overall preparation fidelity f . The inset shows the error-excised internal state preparation fidelity f_E^0 versus the bare internal state preparation fidelity f_{OP} . (d) Removing state preparation errors midsequence. The probability F_n for preparing a defect-free and internally pure array is shown versus array size n . The solid line shows a fit to $(f_R)^n$ with $f_R = 0.951(1)$. Inset: exemplary images for $n = 10$. The error bars in panel (c) are propagated from 68% Wilson-score intervals of the underlying data. The error bars in panel (d) represent 68% Wilson-score intervals.

molecules from $|-\rangle$ to the bright state $|+\rangle = |X^2\Sigma(v=0, N=1), J=1/2, F=0, m_F=0\rangle$ for a final detection image. The measurement-enhanced fidelity f_E can be obtained by measuring the fraction of molecules that appear in the final image out of the occupied sites that were not flagged during error detection. To test our scheme's ability to reject errors, we intentionally lower the bare optical pumping fidelity f_{OP} by decreasing the optical pumping duration. This process also lowers the overall tweezer preparation fidelity f , which is additionally affected by errors in the form of empty tweezers. In Fig. 2(c), we show the error-excised overall preparation fidelity f_E versus f . Additionally, the inset of Fig. 2(c) shows the error-excised internal state preparation fidelity f_E^0 versus f_{OP} (see Appendix C for details on fidelity estimation). Both plots show that excising detected errors substantially improves preparation fidelities. We achieve an error-excised internal state preparation fidelity of $f_E^0 = 0.995(1)$ and an error-excised overall preparation fidelity of $f_E = 0.952(3)$, at a data rate reduction of 7.2(2)%.

The overall fidelity f_E improves significantly upon previous reported values ($f \approx 80\%$) [24,25,43,51]. In addition to improved fidelities, our method importantly provides *robustness* to fluctuations in f_{OP} . For example, at an optical pumping fidelity of $f_{OP} = 0.46$, the error-excised internal state fidelity remains at $f_E^0 \approx 0.96$. We note that the overall preparation fidelity is now limited by occupation errors arising from population loss during the initial nondestructive image (about 2%) used for initial rearrangement and during optical pumping (about 2%). Thus, future work that reduces these losses could allow substantial improvement.

Lastly, our scheme enables midsequence removal of preparation errors via tweezer rearrangement. We first create regularly spaced defect-free arrays of molecules in $\mathcal{F}$ and nondestructively detect their occupation [24]. Subsequently, we apply our measurement-enhanced state preparation protocol. Tweezers identified to have an error are then switched off, and the error-free ones are rearranged into a defect-free configuration. Figure 2(d) shows the measured success probability F_n versus array size n . Fitting the probability to $F_n = (f_R)^n$, where f_R is the single-site overall preparation fidelity, we find $f_R = 0.951(1)$, in agreement with the value of $f_E = 0.952(3)$ without tweezer rearrangement. This finding demonstrates that no detectable additional errors occur during tweezer rearrangement.

While previous experiments with atomic arrays have incorporated similar techniques to postselect for no errors [28], to our knowledge, our work is the first demonstration of midsequence removal of internal state preparation errors. This capability will be crucial for quantum simulation and computation applications that require large-scale arrays at low defect rates. Already, our measurement-enhanced preparation scheme opens the door to quantum simulation of interacting many-body Hamiltonians in defect-free mesoscopic molecular arrays. We achieve an overall fidelity of 60(4)% for arrays of size $n = 10$, which makes practical postselection for perfect arrays possible with only a modest data rate reduction of around 40%.

VI. DETECTING QUBIT LEAKAGE ERRORS

The second part of our work concerns detecting qubit leakage errors. First, we demonstrate a new qubit encoding using two noninteracting hyperfine states in the dark state manifold $\mathcal{T}$. Second, we devise a scheme (“erasure imaging”) to optically detect population in the bright states $\mathcal{F}$ while minimally affecting both population and coherence of the dark qubit. Lastly, we show that a significant source of error for the new qubit—leakage into other molecular states induced by blackbody radiation—can be converted to population in bright states, which can be detected as local errors. These local qubit errors are known as erasures [33,34], a topic that has attracted much attention in both theoretical [35–38] and experimental work in

quantum error correction recently [28–30,39–41]. Our work constitutes the first demonstration of erasure conversion in molecules.

A. New hyperfine qubit in CaF

In previous work using CaF molecules, coherent interactions and two-qubit gates were demonstrated using a rotational qubit encoding [24,25]. Specifically, two *interacting* states from two neighboring rotational manifolds, $|\uparrow\rangle = |X^2\Sigma(v=0, N=1), J=1/2, F=0, m_F=0\rangle$ and $|\downarrow\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=1, m_F=0\rangle$, were used.

Here, we utilize a new *noninteracting* qubit encoding suitable for storing quantum information. Specifically, our encoding uses the two hyperfine states $|1\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=1, m_F=0\rangle$ and $|0\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=0, m_F=0\rangle$, both from the ground rovibrational manifold $X^2\Sigma(v=0, N=0)$ [Fig. 3(a)]. These states are predicted to have long coherence times because of the absence of tensor and Stark shifts and their low magnetic moments. In addition, they are naturally compatible with our error mitigation scheme, as they belong to the dark state manifold $\mathcal{T}$. We note that hyperfine qubit encodings in molecules have been previously proposed for use in quantum information processing [8], and hyperfine coherence times have been measured previously in the bialkali molecules NaK [52], RbCs [53], and NaRb [54]. Very recent work concurrent with ours has also demonstrated a hyperfine qubit encoding in bialkali NaCs molecules trapped in an optical tweezer array [27].

We first demonstrate coherent control over this new hyperfine qubit and show that it has long coherence times. We coherently control the qubit by driving an M1 transition (at approximately 122 MHz) through a two-photon Raman process. Specifically, we use light detuned halfway between the $X - A^2\Pi_{1/2}$ and $X - A^2\Pi_{3/2}$ transitions (single-photon detuning of $\Delta_{XA} \approx 2\pi \times 1.1$ THz) [Fig. 3(b)]. The two frequencies in the Raman light copropagate and have σ^+ projections. To demonstrate coherent control, we prepare molecules in $|1\rangle$ and apply Raman light for a variable duration. As shown in Fig. 3(c), we observe Rabi oscillations at $\Omega_R \approx 2\pi \times 15$ kHz.

We next measure the qubit coherence time using a Ramsey sequence consisting of two Raman $\pi/2$ pulses separated by a variable duration. To minimize inhomogeneous light shifts between sites due to tweezer trapping light, we operate at a tweezer depth of $U_{\text{hf}} = k_B \times 39(1)$ μK . By measuring the decay rate of the amplitude of the Ramsey fringe, we find a bare coherence time of $T_2^* = 19.5(7)$ ms, which can be increased to $T_2 = 288(15)$ ms with a single spin echo. We further extend the coherence time to 1710(60) ms by applying dynamical decoupling with an XY8 pulse sequence (pulse separation of 33.3 ms) [Fig. 3(d)].

In addition to having long coherence times, this hyperfine qubit can be interfaced with the rotational qubit used in

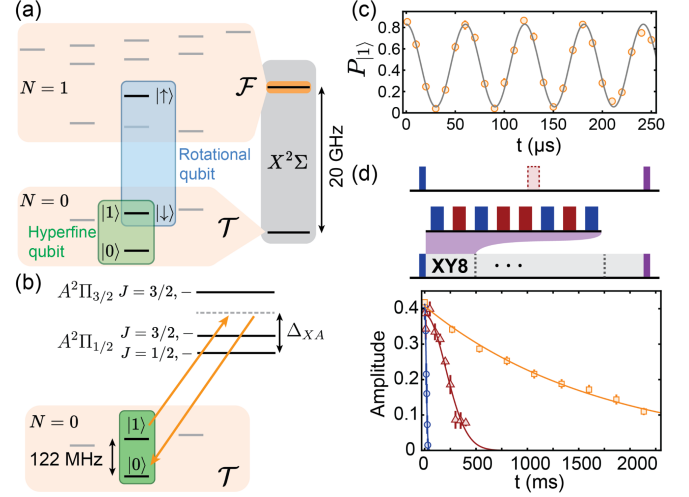


FIG. 3. New hyperfine qubit. (a) Hyperfine qubit encoding using two noninteracting states, $|1\rangle = |X^2\Sigma(v=0, N=0), F=1, m_F=0\rangle$ and $|0\rangle = |X^2\Sigma(v=0, N=0), F=0, m_F=0\rangle$, from the ground rotational manifold $X^2\Sigma(v=0, N=0)$. These states are dark (not in the bright manifold $\mathcal{F}$) with respect to our error detection scheme. (b) Coherent control achieved using a two-photon Raman process with light detuned halfway between the $X - A^2\Pi_{1/2}$ and $X - A^2\Pi_{3/2}$ transitions. (c) Rabi oscillation with an on-resonant Raman drive. We show the population $P_{|1\rangle}$ in $|1\rangle$ versus the Raman pulse duration t . The gray line is a fit to a sinusoid. (d) Qubit coherence versus hold time measured via a Ramsey sequence (top panel). The bottom panel shows the measured Ramsey amplitudes versus hold time t . Data without (with) a spin-echo pulse are shown by blue circles (red triangles) with corresponding solid lines showing fits to Gaussian decay curves. Data with XY8 dynamical decoupling pulses (orange squares) are fit to an exponential decay curve (orange line). The fitted $1/e$ lifetimes are 19.5(7) ms, 288(15) ms, and 1710(60) ms, respectively. The error bars in panel (c) are 68% Wilson-score intervals. The error bars in panel (d) are 1σ fitting errors obtained from fitting Ramsey data.

previous work [22,24,25]. Specifically, we have verified that a quantum state can be coherently transferred from the rotational encoding ($|\uparrow\rangle, |\downarrow\rangle$) to the hyperfine encoding ($|1\rangle, |0\rangle$) (Appendix I).

B. New detection scheme that preserves qubit coherence—composite erasure detection

Having established long coherence times and coherent control of our new hyperfine qubit that is suitable for a quantum memory, we next investigate how to image population in bright states without affecting qubit population and coherence. This is a much more stringent requirement than simply preserving population, which the rapid imaging scheme in Sec. IV satisfies.

We base our new detection scheme off of the rapid imaging procedure described in Sec. IV. To achieve high detection fidelities of bright state population, a tweezer depth of $U_{\text{ED}} = k_B \times 930(20)$ μK is needed, which is about 20 times higher than that used to obtain long

hyperfine coherence times. At the imaging depth, the qubit frequency shifts by $2\pi \times 0.40(6)$ kHz, leading to an approximately 1-rad phase shift during the 3-ms image. Inhomogeneous tweezer depths also cause the qubit to dephase during the image. To counteract this dephasing, we devise a new “composite erasure detection” scheme that incorporates an echo pulse halfway through the image. As shown in Fig. 4(a), our scheme involves 4 steps: (1) The tweezer depth is ramped from U_{hf} to U_{ED} , (2) local errors are detected using rapid resonant imaging, (3) a π pulse is applied to the hyperfine qubit halfway through the image, and (4) the tweezer depth returns to U_{hf} .

To determine how well this procedure preserves qubit population and coherence, we perform Ramsey measurements with a variable number of composite erasure detections between the two Ramsey $\pi/2$ pulses. Figures 4(b) and 4(c) show the offset (amplitude) of the resulting Ramsey fringe versus the number of erasure images, which is proportional to the qubit population (coherence). Fitting to an exponential decay curve, we find that the population and coherence errors per image are $\epsilon_p = 5.1(3) \times 10^{-3}$ and $\epsilon_c = 9.3(8) \times 10^{-3}$, respectively.

To understand whether these errors are fundamental, we next investigate their origins. We independently measure

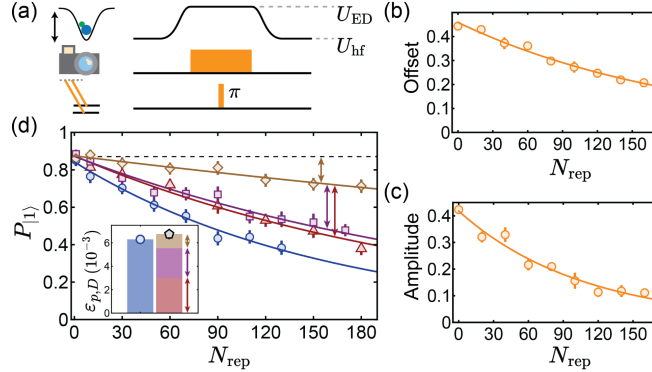


FIG. 4. Composite erasure detection. (a) Composite erasure detection sequence consisting of a tweezer depth increase followed by a rapid resonant image and a tweezer depth decrease. A Raman π pulse is applied midsequence. (b) Ramsey fringe offset versus number of composite erasure detections, N_{rep} . (c) Ramsey fringe amplitude versus N_{rep} . (d) Population loss measurements with various combinations of tweezer ramp (T), error detection light (E), and $v = 1$ repumping light (V). The surviving population $P_{|1\rangle}$ versus N_{rep} cycles of TEV (blue circles), EV (red triangles), and T (purple squares) is shown. Brown diamonds show measurements at a shallow tweezer depth without TEV. The brown, purple, and red arrows indicate contributions from background vacuum and blackbody loss, tweezer ramp, and erasure detection. The inset shows the population loss per detection image $\epsilon_{p,D}$. The sum of each source of loss is consistent with the total loss (blue bar and circle). The error bars in panels (b) and (c) are 1σ fitting errors. The error bars in panel (d) are 68% Wilson-score intervals; those in the inset are 1σ fitting errors.

population loss for each step of the composite erasure detection sequence. These losses include those due to (1) off-resonant scattering of the Raman π -pulse light, (2) off-resonant scattering of imaging light, and (3) tweezer depth ramps. Our measurements indicate that the Raman π pulse contributes a loss rate of $4.1(3) \times 10^{-5}$ per image and is therefore negligible. Off-resonant scattering of imaging light and tweezer ramps contribute $3.0(3) \times 10^{-3}$ and $2.6(3) \times 10^{-3}$, respectively. In addition, we measure a background loss rate of $1.2(1) \times 10^{-3}$ per imaging duration, consistent with the estimated leakage rate to higher vibrational states due to blackbody radiation at room temperature [19,55]. Since leakage due to blackbody radiation can, in principle, be suppressed in a cryogenic setup and loss during the tweezer ramps can likely be reduced with colder molecular temperatures, our scheme is likely only fundamentally limited by off-resonant scattering of imaging light. Shorter imaging durations could potentially allow one to achieve errors per image at the low 10^{-3} level.

C. Erasure conversion and detection of qubit leakage errors induced by blackbody radiation

Having quantified the performance of our composite erasure imaging scheme, we next use it to mitigate a significant error for our hyperfine qubit—leakage into higher vibrational states due to blackbody radiation at room temperature. Specifically, blackbody radiation drives $\Delta v = 1$ vibrational-changing E1 transitions, leading to leakage from the qubit states (encoded in $v = 0$) to the vibrationally excited states $\mathcal{E} = X^2\Sigma(v = 1, N = 1)$. The estimated leakage rate is $\Gamma_{01} \approx 0.3 \text{ s}^{-1}$ [56], which is a slow but significant source of qubit loss. As we will show, these errors can be converted to population in the bright states $\mathcal{F}$ and detected in a site-resolved manner. In other words, the leakage errors can be converted into erasures and detected.

To perform erasure conversion, we simply apply $v = 1$ vibrational repumping light that repumps molecules into the bright manifold $\mathcal{F}$ with 98% probability [Fig. 5(a)]. There are, however, two subtleties. First, vibrationally excited molecules in $X^2\Sigma(v = 1)$ decay back to the ground vibrational manifold $X^2\Sigma(v = 0)$ at a rate $\Gamma_{10} \approx 5.3 \text{ s}^{-1}$, which is much quicker than the blackbody excitation rate $\Gamma_{01} \approx 0.3 \text{ s}^{-1}$. Therefore, erasure conversion should occur at a rate much faster than Γ_{10} . It is not necessary, however, to detect the error flags in $\mathcal{F}$ quickly since they lie in the long-lived ground vibrational manifold. Second, blackbody radiation causes secondary leakage of molecules out of the detectable bright states $\mathcal{F}$ into the undetected dark states $X^2\Sigma(v = 1, N = 0, 2)$ [Fig. 5(b)]. These states then decay both back to $\mathcal{F}$ and also to another dark rotational manifold $X^2\Sigma(v = 0, N = 3)$. We close this leakage pathway by applying rotational repumping light ($N = 3$ repumper) that brings $X^2\Sigma(v = 0, N = 3)$ molecules back to the error flag manifold $\mathcal{F}$ [Fig. 5(b)].

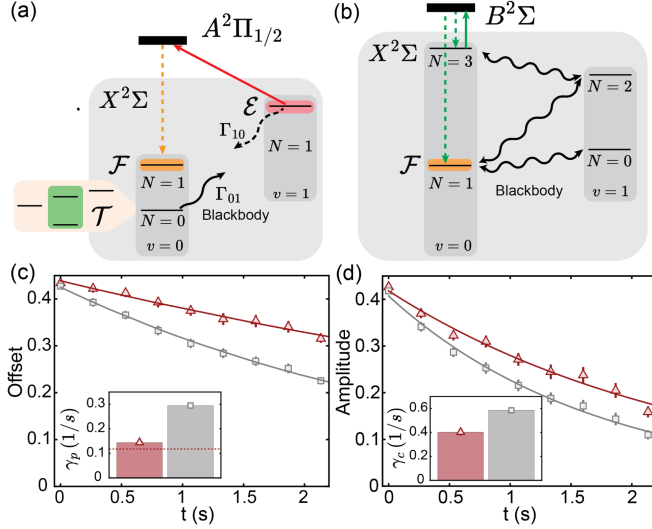


FIG. 5. Erasure conversion of blackbody errors. (a) Erasure conversion scheme. Blackbody radiation (black solid wavy line) predominantly excites molecules from $\mathcal{T}$ to $\mathcal{E}$ at a rate Γ_{01} , which radiatively decays (black dashed wavy line) back to $v = 0$ at a rate Γ_{10} . We erasure convert to $\mathcal{F}$ using $v = 1$ repumping light (red arrow). (b) Robustness of $\mathcal{F}$ to blackbody leakage errors. Blackbody leakage errors (black wavy line) primarily populate $X^2\Sigma(v = 0, N = 3)$ states, which are converted back to $\mathcal{F}$ by the rotational repumping light present during erasure detection (solid green line). (c) Ramsey fringe offset versus hold time t without erasure excision (gray squares) and with a single final error detection image. Solid curves are exponential decay fits. Inset: fitted $1/e$ loss rates γ_p , with the red dashed line indicating the prediction from separately measured rates (Appendix J). (d) Ramsey fringe amplitude versus hold time with (red triangles) and without (gray squares) error excision. Inset: amplitude loss rate γ_c from the exponential fits. The error bars in panels (c) and (d) and their insets are 1σ fitting errors.

We first experimentally confirm that secondary blackbody leakage is indeed suppressed by $N = 3$ repumping light. We indeed find that with the extra repumping light, the error flags $\mathcal{F}$ have a population lifetime of $\tau = 13.5(5)$ s, well exceeding the blackbody-limited lifetime by a factor of approximately 4. The residual loss is attributed to background gas collisions and double blackbody excitation events into dark rotational manifolds with $N \geq 5$ (Appendix J).

We next experimentally implement our blackbody erasure conversion scheme and investigate whether it extends qubit population lifetimes and coherence times. We measure the offset and amplitude of Ramsey fringes with and without erasure excision. For these measurements, erasure conversion is repeatedly applied midcircuit, with erasure detection occurring once at the end of the sequence. During the Ramsey hold time, XY8 dynamical decoupling is applied.

Regarding qubit population loss, we measure a bare loss rate of $\gamma_p = 0.30(1)$ s $^{-1}$, consistent with the estimated

blackbody excitation rate. Excising erasures, the loss rate decreases to $\gamma_{p,E} = 0.14(1)$ s $^{-1}$ [Fig. 5(c)]. We find that a loss rate model that incorporates vacuum loss, blackbody excitation, erasure conversion and detection efficiency, and the internal state preparation fidelity agrees well with our measurements [Fig. 5(c)]. We use this model to infer that approximately 80% of blackbody errors are erasure converted and detected correctly (Appendix J). Regarding qubit coherence, we measure a bare loss rate of $\gamma_c = 0.58(2)$ s $^{-1}$, which decreases to $\gamma_{c,E} = 0.40(2)$ s $^{-1}$ upon excising erasures. This improvement is consistent with the improvement in population loss, suggesting that blackbody radiation does not lead to additional decoherence beyond population loss. Overall, we find that erasure conversion of blackbody errors allows excision of 51(3)% (31(5)%) of *all* qubit population (coherence) errors, which includes loss due to a finite vacuum lifetime.

Looking ahead, because these blackbody erasures can be detected midsequence, they can, in principle, be corrected, as previously demonstrated in molecular ions [55]. For molecular tweezer arrays, blackbody erasures can be removed via reloading a site or by reinitializing the molecular state [57,58].

VII. DISCUSSION AND OUTLOOK

In summary, we have demonstrated two error mitigation schemes for state preparation errors and qubit leakage errors. Specifically, we demonstrated (1) a measurement-enhanced state preparation scheme that enables robust and record-level fidelities for preparing defect-free and internally pure molecular arrays, and (2) an erasure conversion and detection scheme for blackbody-induced leakage errors of a hyperfine qubit. Both parts of our work represent key new capabilities in the quantum control of molecules. Our measurement-enhanced state preparation work is the first demonstration of removal of internal state errors following midsequence error detection; our erasure conversion work constitutes the first demonstration of erasure conversion in molecules.

Our error mitigation work opens several possibilities. First, state preparation fidelities of approximately 95% open the door to quantum simulation of many-body Hamiltonians with low defect rates. In particular, for mesoscopic systems of $\mathcal{O}(10)$ particles, we can postselect on perfectly prepared arrays with a tolerable level of data reduction.

Second, our work lays the foundations for exploring erasure conversion in the context of quantum error correction with molecular qubits. For quantum error correction, erasure conversion is useful when the dominant errors—often from two-qubit gates—are converted and detected [35]. Our current work concerns erasure conversion of a significant source of leakage errors in non-interacting hyperfine qubits. For quantum memories stored in these qubits, our work could thus substantially

mitigate idling errors. Beyond idling errors, our erasure conversion approach could be extended to mitigate two-qubit gate errors in specific regimes. To date, entangling two-qubit gates in molecular tweezer arrays have all relied on the electric dipolar interaction, whose strength depends sensitively on molecular separation. In several experiments including ours [24,25,27], two-qubit gate fidelities were limited by thermal motion within tweezers, which led to variations in the interaction strength and, consequently, gate errors. Very recent work from Ref. [26] substantially reduces this source of gate errors by placing molecules far apart, where interaction strength variations due to motion become small. However, at these far separations, gate times became significantly increased, which led to significant leakage errors (about 5%) arising from off-resonant scattering of trapping light. In this far-separated regime, leakage was the dominant source of gate infidelity. Because leakage errors are erasure convertible, improving molecular two-qubit gate fidelities in the far-separated regime is a clear use case for erasure conversion.

Third, the composite erasure detection scheme used in our erasure conversion work could open the door to midcircuit measurements in molecular tweezer arrays, a powerful tool for quantum error correction and exploring exotic many-body phases of matter. Specifically, our composite erasure detection shows that site-resolved measurements of bright state populations can be made without affecting quantum information encoded in dark states. By combining this readout capability with site-selective transfer from the hyperfine qubit encoding into bright states implementable via local light shifts, site-selective midcircuit measurements with crosstalk errors at the 10^{-3} level could be achieved.

In addition to the above possibilities opened by our error mitigation results, the new technical capabilities developed in our work also open additional possibilities. First, our new noninteracting hyperfine qubit encoding can be integrated with the previously used interacting rotational encoding, directly enabling applications using three-level systems that include quantum information processing with qutrits, simulation of $S = 1$ quantum spin models, bosonic t - J models [59], and lattice gauge theory models [60]. Second, our work on detecting blackbody excitations is directly relevant to investigations of molecule-based temperature standards [61]. Third, our work on enhanced tweezer preparation and catching blackbody errors could aid molecular tweezer-based precision measurement experiments [15,16], both through reducing spatial disorder in molecular arrays and through using metrologically useful entangled states such as spin-squeezed states [62,63] that can be naturally generated in interacting defect-free arrays of many molecules [64,65].

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APPENDIX A: GENERAL FRAMEWORK FOR SITE-RESOLVED ERROR DETECTION

In this appendix, we describe a general framework for site-resolved errors in optical tweezer arrays of atoms and molecules. This framework provides a unified way to understand the requirements for site-resolved error detection in the following tasks: (1) preparation of defect-free arrays, (2) measurement-enhanced state preparation, and (3) erasure conversion and detection.

In atomic and molecular tweezer array experiments, each tweezer site is either empty or occupied by one particle. At each site, the local Hilbert space $\mathcal{H}$ consists of $|\emptyset\rangle$, the state corresponding to an empty site, and the set of all internal states [Fig. 6(a)]. For a particular application, only a target subset of these states $\mathcal{T}$ is used. For example, for state preparation, one state in $\mathcal{T}$ is used; for encoding a qubit, at least two states from $\mathcal{T}$ are needed. Note that while we ignore the motional degree of freedom of a particle within a tweezer, the same framework can be extended to include motional states [30].

For error detection, one encodes the presence of an error as occupation in a set of detectable internal states $\mathcal{F}$ (bright

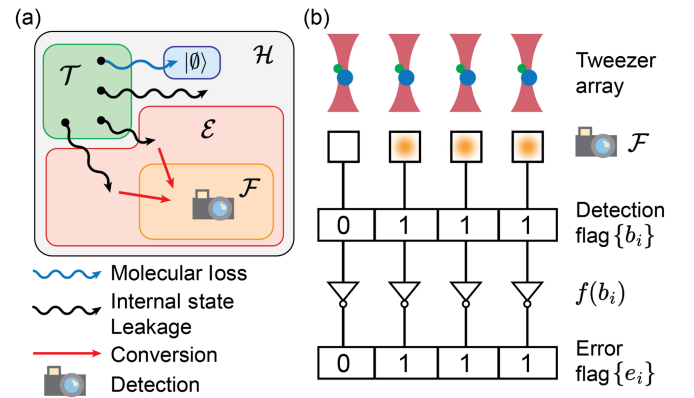


FIG. 6. General framework for site-resolved error detection. In panel (a), $\mathcal{H}$ is the Hilbert space on a tweezer site, $\mathcal{T}$ is the subspace where error-free molecules reside, and $\mathcal{F}$ is the set of detectable states used to encode errors. Leakage errors transfer population from $\mathcal{T}$ to $\mathcal{F}$, which can subsequently be converted to $\mathcal{F}$ for detection. The loss of a particle results in an empty tweezer ($|\emptyset\rangle$). (b) Detection of total population in $\mathcal{F}$ is binarized into a bitstring $\{b_i\}$, from which the error flag $\{e_i\}$ is obtained.

states). The detection outcome population in $\mathcal{F}$ is binarized into bitstrings $\{b_i\}$ and converted into an error flag $\{e_i = f(b_i)\}$, where $e_i = 1$ ($e_i = 0$) indicates an error (error-free) site [Fig. 6(b)]. Note that $\{e_i\}$ could be used to remove errors midsequence or to postselect for error-free instances by excising error-flagged data.

When at least one bit of information is encoded in $\mathcal{T}$, errors with a known location are called erasures [33,34]. In particular, population that leaks out of $\mathcal{T}$ into other internal states $\mathcal{E}$ can be transferred to the error flag states $\mathcal{F}$ and subsequently detected. This process is known as erasure conversion [35].

For site-resolved error detection to be practically useful, it must not affect error-free sites. Specifically, for quantum erasure detection, all populations and coherences in $\mathcal{T}$ should be minimally affected by detection. We will reserve the term midcircuit for situations when $\mathcal{T}$ encodes a qubit and is minimally affected by conversion or detection.

In the following, we explicitly show how this framework applies to the following three tasks: (1) preparation of defect-free arrays, (2) measurement-enhanced state preparation, and (3) erasure conversion and detection.

For the task of preparing defect-free arrays, the target space $\mathcal{T}$ consists of all detectable bright states. In our case, they are the 12 hyperfine states within the rotational manifold that permits optical cycling [$\mathcal{T} = X^2\Sigma(v=0, N=1)$]. Because the $\mathcal{T}$ are the only laser-coolable states, any site is either in $\mathcal{T}$ or in the empty state $|\emptyset\rangle$. Error detection involves determining empty states, i.e., population in $|\emptyset\rangle$, without substantially affecting occupied sites (in states $\mathcal{T}$). This task is accomplished through nondestructive fluorescence detection, which returns a binarized outcome b_i , with 1 corresponding to a fluorescing site and 0 corresponding to a dark site. The error flag is given by $e_i = f(b_i) = \sim b_i$ since fluorescing sites are filled and hence error-free, while dark sites are empty and thus have occupation errors. Defect-free preparation is accomplished by rearranging error-free or occupied tweezers into the desired configuration.

For measurement-enhanced internal state preparation, the target space $\mathcal{T}$ consists of a specific dark state. In our specific case described in Sec. V, this state is $|- \rangle = |X^2\Sigma(v=0, N=0), F=1, m_f=-1\rangle$, with state preparation errors flagged as population in bright states $\mathcal{F} = X^2\Sigma(v=0, N=1)$. Error detection requires that error-free sites, which have population in $|- \rangle$, remain unaffected by fluorescence detection of error flags $\mathcal{F}$. Thus, the detection scheme should not cause leakage into other internal states or loss, which in this framework can be thought of as leakage into $|\emptyset\rangle$. The error detection outcome again consists of bitstrings $\{b_i\}$, with 1 for sites with fluorescence and 0 for dark sites. Here, the mapping to errors is simply $e_i = f(b_i) = b_i$. Internal state preparation errors can then be identified and optionally removed via tweezer rearrangement.

The above two tasks can be viewed as a form of classical generalized erasure detection. For the final case of detecting qubit leakage errors (quantum erasures), the target space $\mathcal{T}$ contains at least two states. In our case, these are the dark states $|1\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=1, m_F=0\rangle$ and $|0\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=0, m_F=0\rangle$, as described in Sec. VI A of the main text. Similar to measurement-enhanced state preparation, errors are encoded in the bright states $\mathcal{F} = X^2\Sigma(v=0, N=1)$ and detected. The same mapping of error detection bitstrings $\{b_i\}$ to error flags $\{e_i\}$ applies [$f(b_i) = b_i$]. In our demonstration of erasure conversion of blackbody-induced leakage errors described in Sec. VI C of the main text, blackbody excitation causes leakage into the states $\mathcal{E} = X^2\Sigma(v=1, N=1)$, which are converted into the bright states $\mathcal{F} = X^2\Sigma(v=1, N=0)$ with a vibrational repumping laser.

APPENDIX B: EXPERIMENTAL APPARATUS AND OPTICAL TWEezer LOADING PROCEDURE

A detailed description of our molecular tweezer apparatus and tweezer loading procedures can be found in Refs. [19,24,66]. In brief, CaF molecules are produced in a cryogenic buffer gas source [44], slowed via chirped slowing [45,67], and loaded into a dc magneto-optical trap (MOT) [46,47,68,69]. Subsequently, the MOT is magnetically compressed, Λ cooled in free space [48], and transferred into a Λ -enhanced blue-detuned MOT (Λ -BDM) [49]. The molecules are subsequently transferred into an attractive 1064-nm ODT and transported to the focal plane of the optical tweezer traps. The optical tweezers are then switched on, and Λ cooling is applied to load molecules stochastically into tweezer sites [17]. Following loading, the molecules are in the laser-coolable manifold $\mathcal{F} = X^2\Sigma(v=0, N=1)$.

The optical tweezer array is one dimensional and formed by $\lambda = 781$ nm light. A single laser beam passes through an acousto-optical deflector (AOD), which is driven by an arbitrary waveform generator (AWG). The diffracted orders then pass through a microscope objective (NA = 0.65), which focuses them into spots with Gaussian waists of $w_0 = 730$ nm. For initial loading, the traps are separated by $4.20(6)$ μm .

APPENDIX C: DETECTION PROCEDURES

For all detection procedures, only molecules in the optically cyclable rovibrational manifold $X^2\Sigma(v=0, N=1) = \mathcal{F}$ appear bright on the camera and can be detected; molecules in other states appear dark and are indistinguishable from unoccupied sites.

1. Initial detection procedure to determine tweezer occupation

For all data used for this work, we first determine the initial molecular occupation at each tweezer site following

stochastic loading by taking a short (4-ms) nondestructive fluorescence image using a variant of Λ imaging [17,24,48]. The image is nondestructive in the sense that molecules are not lost and that they remain in the laser-coolable manifold $\mathcal{F} = X^2\Sigma(v=0, N=1)$.

The detailed procedure, along with how this procedure can be combined with rearrangement, can be found in Ref. [24].

2. Rapid resonant imaging procedure for midsequence error detection

For the rapid resonant images used for improving state preparation and for erasure detection, we use the newly developed, rapid (3-ms) resonant imaging scheme described in detail in Sec. IV of the main text. Molecular fluorescence is collected through the objective and imaged onto an EMCCD camera.

To image molecules in $\mathcal{F}$, we apply light resonant with all resolved hyperfine transitions of the $X^2\Sigma(v=0, N=1) - A^2\Pi_{1/2}(v=0, J=1/2, +)$ transition. Laser light addressing the $X^2\Sigma(v=1, 2, 3, N=1) \rightarrow A^2\Pi_{1/2}(v'=v-1, J=1/2, +)$ transitions acting as vibrational repumpers is also applied. We additionally apply light addressing the $X^2\Sigma(v=0, N=3) \rightarrow B^2\Sigma(v'=0, N=2)$ transition, which acts as a rotational repumper [19]. These beams propagate perpendicular to the imaging axis and cause no observable background for typical imaging durations. Therefore, the false-positive probability ϵ_{01} , which determines the data rate, is independent of imaging parameters and only depends on camera noise.

An additional consideration for error detection is crosstalk, where population leaks between $\mathcal{F}$ and $\mathcal{T}$. In our scheme, crosstalk is forbidden by selection rules. Because the optical transitions used are all E1 transitions, and $\mathcal{F} = X^2\Sigma(v=0, N=1)$ and $\mathcal{T} \subset X^2\Sigma(v=0, N=0)$ are of opposite parity, no population leakage occurs even in the presence of off-resonant scattering from imaging light.

3. Final detection to identify tweezer occupation

For final readout, we perform a long (30-ms) destructive fluorescence image using a variant of Λ imaging [17,24,48]. Details, including the thresholding procedure, can be found in Ref. [19].

4. Binarizing fluorescence images

We employ up to four different imaging configurations during our investigations: a 4-ms nondestructive Λ image, a 3-ms midsequence image (rapid resonant imaging) for internal state error detection, a 3-ms midsequence image (rapid resonant imaging) for blackbody error detection, and a 30-ms destructive readout image. Note that the two rapid resonant imaging configurations use slightly different magnetic field magnitudes and directions. We treat these two cases separately and determine their identification

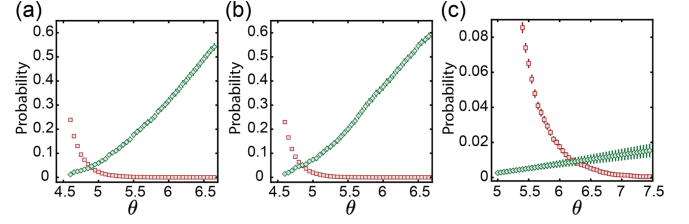


FIG. 7. Imaging misidentification probabilities. The false-positive probability ϵ_{01} (red squares) and false-negative probability ϵ_{10} (green diamonds) are shown versus image classification threshold θ for (a) the internal state error rapid resonant image, (b) the blackbody error rapid resonant image, and (c) the final destructive readout image. Note that for panel (c), the background due to imaging light is non-negligible and contributes to ϵ_{01} . The error bars represent a ± 1 standard deviation and are obtained using a bootstrapping procedure [19].

errors independently since their corresponding photon scattering rates could differ slightly.

For each imaging configuration, we use a threshold θ in summed camera counts for each tweezer site to classify the presence (1) or absence (0) of a molecule in $X^2\Sigma(v=0, N=1)$ [19]. Depending on the experimental sequence, more than one rapid resonant image can be applied. For example, a sequence could use rapid resonant imaging both to flag initial internal state errors and to detect subsequent blackbody errors.

For all investigations, the identification threshold of the initial nondestructive image is sufficiently high such that the imaging survival (approximately 98%) dominates the nondestructive detection fidelity (f_{ND}), which is the probability that a tweezer identified as occupied remains occupied following imaging.

To characterize the performance of the rapid resonant images and the final readout image, we follow the procedure in Ref. [19] to obtain the false-positive probabilities ϵ_{01} and false-negative probabilities ϵ_{10} . The results are shown in Fig. 7. Table I provides the thresholds used, along with ϵ_{01} and ϵ_{10} for each imaging configuration at these thresholds.

5. Measurement correction

We perform measurement correction on identification errors [19,24,70]. Briefly, the probabilities of detecting a bright tweezer site and a dark tweezer site can be expressed as $\mathbf{P}_{\text{det}} = \{P_{1,\text{det}}, P_{0,\text{det}}\}$. In the presence of identification

TABLE I. False-negative and false-positive probabilities for imaging.

	θ	$\epsilon_{01}(\theta)(\%)$	$\epsilon_{10}(\theta)(\%)$
Internal state error imaging	4.8	7.2(2)	3.3(4)
Blackbody error imaging	4.8	6.3(2)	4.5(5)
Final readout	6.2	0.99(10)	0.88(20)

errors, these are related to the true probabilities $\mathbf{P} = \{P_1, P_0\}$ via $\mathbf{P}_{\text{det}} = \mathbf{M} \cdot \mathbf{P}$, where

$$\mathbf{M} = \begin{pmatrix} \bar{\epsilon}_{10} & \epsilon_{01} \\ \epsilon_{10} & \bar{\epsilon}_{01} \end{pmatrix}, \quad (\text{C1})$$

and $\bar{\epsilon} = 1 - \epsilon$. Inverting this equation gives $\mathbf{P} = \mathbf{M}^{-1} \cdot \mathbf{P}_{\text{det}}$.

In addition to identification errors, we also correct for population loss errors due to the final destructive imaging procedure. For the final destructive image, the bias magnetic field is ramped to zero, and the tweezer depth is ramped to the imaging depth. This process results in a population survival of $s_r = 0.9977(4)$. Population in $X^2\Sigma(v=0, N=1)$ prior to the magnetic field and tweezer ramp, denoted $P_{m,1}$, is calculated via $P_{m,1} = P_1/s_r$, which is the measurement-corrected population used for all analyses.

APPENDIX D: INTERNAL STATE PREPARATION PROCEDURE

In this appendix, we provide details on the internal state preparation procedure, which includes optical pumping and microwave transfer.

1. Optical pumping procedure

Our procedure for optical pumping into $|D'\rangle = |X^2\Sigma(v=0, N=1), J=3/2, F=2, m_F=-2\rangle$ involves using light resonant with the $X^2\Sigma(v=0, N=1) \rightarrow A^2\Pi_{1/2}(v=0, J=1/2, +)$ transition. It involves several steps, with a crucial final step where $|D'\rangle$ is the only hyperfine state in $X^2\Sigma(v=0, N=1)$ that is dark. In brief, the final optical pumping step uses a single beam containing minimal σ^+ projection onto the quantization axis set by the magnetic field, which is oriented along the tweezer polarization axis and has a magnitude of $B = 3.9$ G. The beam contains four frequencies addressing the four hyperfine manifolds in $X^2\Sigma(v=0, N=1)$ and has a total power of $P = 6.4$ μW . The tweezer depth is kept at $U = k_B \times 930(20)$ μK during optical pumping.

2. Microwave transfer procedure

Two population transfers via microwave pulses are used to implement and verify the internal state preparation procedure. Each pulse is performed at a tweezer depth of $U = k_B \times 130(3)$ μK in a magnetic field of $B = 4.4$ G oriented at $\theta = 53^\circ$ from the tweezer polarization axis. The first pulse transfers population from $|D'\rangle$ to $|-\rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=1, m_F=-1\rangle$ and the second from $|-\rangle$ to $|\uparrow\rangle$. Each utilizes a composite $90_y 180_x 90_y$ pulse sequence that increases the robustness to microwave power fluctuations. We also shape the amplitude of each pulse using a Gaussian envelope that is truncated at $\pm 3\sqrt{2}\sigma$, where σ is the Gaussian width.

The peak Rabi frequency during the first (second) composite pulse is $\Omega \approx 2\pi \times 900$ kHz ($\Omega \approx 2\pi \times 300$ kHz), with a total pulse duration of 3.7 μs (11.0 μs).

APPENDIX E: RAPID IMAGING, COMPOSITE ERASURE DETECTION, AND THEIR CHARACTERIZATION

1. Rapid imaging procedure

All rapid imaging configurations use light containing four frequency components resonant with the four resolved hyperfine transitions between the $X^2\Sigma(v=0, N=1)$ and $A^2\Pi_{1/2}(v=0, J=1/2, +)$ manifolds. Vibrational repumpers for $X^2\Sigma(v=1, 2, 3, N=1)$ and a rotational repumper for $X^2\Sigma(v=0, N=3)$ are simultaneously applied. The resonant X - A light is delivered by two retroreflected beams with a total intensity of $I = 4.5$ mW/cm^2 and a hyperfine power ratio of 5:2.7:1.1:3.8. The $v=1$ and $N=3$ repumpers are delivered along the same path as the X - A light and are retroreflected; the $v=2, 3$ repumping light is delivered with a single beam.

The internal state error detection sequence (rapid resonant imaging) is composed of a 1-ms tweezer depth ramp from $U = k_B \times 130(3)$ μK to $U = k_B \times 930(20)$ μK , a rapid resonant image (3 ms), and a 1-ms tweezer ramp back to $U = k_B \times 130(3)$ μK . This image occurs at ($B = 4.4$ G, $\theta = 53^\circ$).

The blackbody error detection and composite erasure image sequences are composed of a 1-ms tweezer depth ramp from $U = k_B \times 39(1)$ μK to $U = k_B \times 930(20)$ μK , a rapid resonant image (3 ms), and a 1-ms tweezer ramp back to $U = k_B \times 39(1)$ μK . Halfway through the erasure image, a 40- μs X - B Raman π pulse realizes a spin echo in the hyperfine qubit manifold. Composite erasure imaging occurs at ($B = 2.0$ G, $\theta = 90^\circ$), which is also a pseudo-magic configuration for the rotational qubit.

2. Photon scattering rate

For rapid resonant imaging, the X - A photon scattering rate is determined from the number of photons collected by the camera. Image histograms are constructed by post-selecting for molecules that survive the rapid resonant image; this ensures that the imaging lifetime, which is comparable to the image duration, does not affect the scattering rate measurement. We find that approximately 24 photons are collected by the camera during the 3-ms rapid resonant image. Incorporating the microscope objective collection efficiency and optical losses along the imaging path gives a scattering rate of $R_{\text{scat}} \approx 1.6 \times 10^5$ s^{-1} .

We have compared this scattering rate with that obtained using a Bloch equation simulation using the measured hyperfine balance and detunings [67,71]. Ignoring ac Stark shifts, our simulations give a scattering rate of $R_{\text{scat}} \approx 2.8 \times 10^5$ s^{-1} , about a factor of 2 larger than observed. We attribute the discrepancy to ac Stark shifts or

a slight miscalibration of the resonant frequency. Our free space simulations show that a 3-MHz overall detuning to the blue results in a factor of approximately 2 decrease in the simulated scattering rate. The differential ac Stark shifts between the various hyperfine states in $\mathcal{F}$ are approximately 10% of the trap depth (rms deviation) at our tweezer wavelength. This finding corresponds to a shift of approximately 2 MHz, compared to an excited state linewidth of approximately 8 MHz.

3. Off-resonant photon scattering rate

We measure the lifetime of the target internal state $|- \rangle = |X^2\Sigma(v=0, N=0), J=1/2, F=1, m_F=-1\rangle$ in the rapid imaging configuration ($U = k_B \times 930(20) \mu\text{K}$, $B = 4.4 \text{ G}$, and $\theta = 53^\circ$) with and without the resonant imaging light applied. For all lifetime measurements, a 5-ms rapid resonant imaging pulse is applied at the end of the variable hold time to remove any population shelved in $X^2\Sigma(v=1, N=1)$ states via blackbody pathways. From these measurements, we measure a contribution of $\gamma = 0.76(11) \text{ s}^{-1}$ due to rapid resonant imaging.

We note that the population loss is due to off-resonant scattering and primarily results in population leakage to $X^2\Sigma(v=0, N=2)$ hyperfine states (20 states in total) and the other three hyperfine states in the $X^2\Sigma(v=0, N=0)$ manifold. We note that these leakage errors could, in principle, be detected in a more sophisticated conversion scheme utilizing more repumpers and microwave pulses, and they are therefore not a fundamental limitation.

To a good approximation, the observed loss rate is given by the off-resonant photon scattering rate. We therefore compare the observed population loss rate with the predicted off-resonant photon scattering rate by $|- \rangle$ due to the rapid imaging light. Because the off-resonant photon scattering rate causes leakage and is predicted to be low, the photon scattering rate can simply be calculated using Fermi's golden rule. The off-resonant photon scattering rate is primarily due to excitation to $A^2\Pi_{1/2}(v=0, J=1/2, -)$ and $A^2\Pi_{1/2}(v=0, J=3/2, -)$ and is given by

$$\Gamma_{\text{sc}} = \frac{\pi c^2}{2\hbar\omega^3} \Gamma^2 I \left(\frac{2}{3} \frac{1}{\Delta_{1/2}^2} + \frac{1}{3} \frac{1}{\Delta_{3/2}^2} \right), \quad (\text{E1})$$

where I is the intensity of the imaging light and $\Delta_{1/2} \approx 2\pi \times 20 \text{ GHz}$ ($\Delta_{3/2} \approx 2\pi \times 50 \text{ GHz}$) is the detuning from the $X^2\Sigma(v=0, N=0) \rightarrow A^2\Pi_{1/2}(v=0, J=1/2, -)$ ($X^2\Sigma(v=0, N=0) \rightarrow A^2\Pi_{1/2}(v=0, J=3/2, -)$) transition. The predicted off-resonant scattering rate is $\Gamma_{\text{sc}} = 0.24 \text{ s}^{-1}$, a factor of 3 lower than the observed population loss. This prediction suggests the possible presence of small amounts of spectral impurities in the imaging light that could be filtered out using an optical cavity.

APPENDIX F: ANALYSIS PROCEDURE AND LOSS MODEL FOR OPTICAL PUMPING AND TWEEZER PREPARATION FIDELITIES

In this appendix, we provide details about extracting the bare optical pumping fidelity f_{OP} , the error-excised internal state purity p_E , and the overall error-excised tweezer preparation fidelity f_E , which includes both occupation and internal state errors.

1. Determining optical pumping fidelity versus optical pumping time

The bare signal $\tilde{p}_E$ is the measured probability that a site initially identified to be occupied by the initial nondestructive image and identified to be error-free in its internal state by a midsequence rapid image is measured to be in $\mathcal{F}$ (bright) in the final destructive image. We first construct an expression for $\tilde{p}_E$ based on the various sources of imperfections. These include (1) the survival probability s of the initial nondestructive image, (2) the survival probability $s_{E,1}$ of detectable molecules following rapid resonant imaging, (3) the survival probability $s_{E,0}$ of the target state $|- \rangle$ following rapid resonant imaging, and (4) the microwave transfer fidelity s_{MW} from the target state $|- \rangle$ to the detectable state $|\uparrow\rangle$ for final imaging.

We measure an initial nondestructive survival probability of $s = 0.952(3)$, which includes all losses arising from the initial nondestructive image, F pumping, m_F pumping, and hold time. We measure $s_{E,1} = 0.13(1)$ and $s_{E,0} = 0.9962(3)$. The microwave transfer fidelity $s_{\text{MW}} = 0.9931(8)$ is measured by tracking population depolarization versus the number of microwave sweeps. For notational convenience, we also define $s_{\text{all}} = s_{E,0} \cdot s_{\text{MW}}$.

In terms of the above fidelities, $\tilde{p}_E$ can be related to the bare optical pumping fidelity f_{OP} through

$$\tilde{p}_E = \frac{s[f_{\text{OP}}s_{\text{all}}\bar{\epsilon}_{01} + s_{E,1}(1 - f_{\text{OP}})\epsilon_{01}]}{(1 - f_{\text{OP}})s\epsilon_{10} + (sf_{\text{OP}} + (1 - s))\bar{\epsilon}_{01}}. \quad (\text{F1})$$

Explicitly, the numerator is the fraction of correctly prepared occupied tweezers that are bright during the final destructive image *and also* identified to be dark in the error detection image (see Fig. 8). The first term corresponds to tweezers that were identified to be dark in the error detection image and that should be bright in the verification image. The second term corresponds to tweezers that were incorrectly identified to be dark in the error detection image and that should be bright in the verification image. The denominator is the total number of tweezers initially identified to be occupied and also identified to be dark during the error detection image (column 3 in Fig. 8 multiplied by identification errors). Note that measurement correction of the final image is not included in the expression.

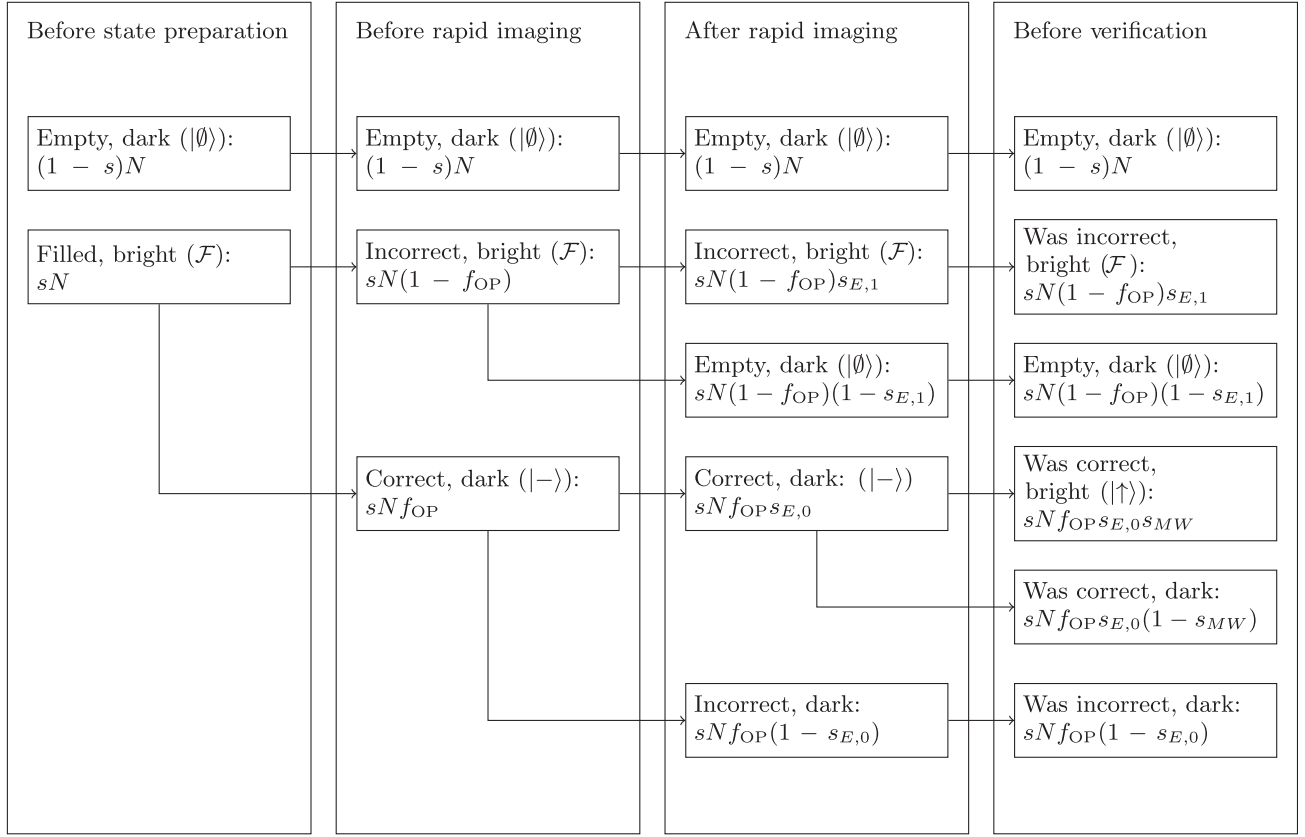


FIG. 8. Populations during state preparation error detection process. The initial number of occupied tweezers is N , and “correct”(“incorrect”) refers to molecules that are prepared in the target internal state (in other states). The listed populations do not take into account identification errors, which are threshold dependent and are encoded by the parameters ϵ_{01} and ϵ_{10} . Postinitial detection includes loss during additional hold time following the nondestructive image. Note that we measurement correct for identification errors in the final verification image, but we do not measurement correct identification errors for the midsequence image. This approach allows a fair indication of the improvement that can be achieved via midsequence tweezer rearrangement.

In the limit of no error detection [setting $\theta_E = \infty$, $\epsilon_{10}(\theta_E) = 1$ and $\epsilon_{01}(\theta_E) = 0$], the above expression reduces to

$$\tilde{p}_{E\infty} = s[f_{OP}s_{all} + s_{E,1}(1 - f_{OP})], \quad (F2)$$

which can be solved for the optical pumping fidelity f_{OP} ,

$$f_{OP} = \frac{\tilde{p}_{E\infty}/s - s_{E,1}}{s_{all} - s_{E,1}}. \quad (F3)$$

This process allows us to determine the achieved bare optical pumping fidelity f_{OP} from $\tilde{p}_E$ for a given optical pumping duration t . We note that this procedure makes no assumptions nor does it incorporate any information about the identification errors of the rapid imaging sequence, except that $\epsilon_{10}(\infty) = 1$ and $\epsilon_{01}(\infty) = 0$. With the relationship between optical pumping fidelity f_{OP} and the experimentally controlled optical pumping duration t established, we can use the above model to *predict* $\tilde{p}_E$ as a function of error detection threshold θ_E because the

false-positive probabilities $\epsilon_{01}(\theta)$ and false-negative probabilities $\epsilon_{10}(\theta)$ versus threshold θ are already known. Thus, we can verify the measured infidelities and the above model. As shown in Fig. 9, these predicted curves agree well with the experimentally measured values of $\tilde{p}_E$, confirming the validity of our model and the measured values of s , $s_{E,0}$, $s_{E,1}$, and s_{MW} .

2. Obtaining error-excised internal state purity

The error-excised internal state purity p_E , which is the probability that a site with a molecule identified to be free of internal state errors via midsequence rapid imaging is indeed in the correct internal state, is given by

$$\begin{aligned} p_E &= \frac{s f_{OP} s_{E,0} \bar{\epsilon}_{01}}{s(1 - f_{OP})\epsilon_{10}s_{E,1} + s f_{OP} \bar{\epsilon}_{01}} \\ &= \frac{f_{OP} s_{E,0} \bar{\epsilon}_{01}}{(1 - f_{OP})\epsilon_{10}s_{E,1} + f_{OP} \bar{\epsilon}_{01}}. \end{aligned} \quad (F4)$$

Explicitly, the denominator is the fraction of tweezers that are occupied with a molecule when conditioned on being

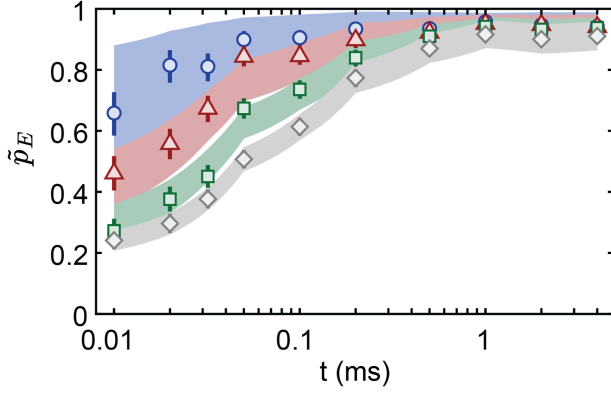


FIG. 9. Error-excised survival following state preparation. The detection-conditioned probability $\tilde{p}_E$ of a molecule appearing in the final image versus optical pumping duration t is used to control the bare optical pumping fidelity f_{OP} . Here, $\tilde{p}_E$ is shown for various error-detection image classification thresholds $\theta_E = (4.6, 5.6, 6.6, \infty)$ (blue circles, red triangles, green squares, gray diamonds). Colored bands show the corresponding theoretical predictions. The error bars are 68% Wilson-score intervals.

identified to be occupied in the initial image and dark in the error detection image. The numerator is the fraction of tweezers that are in fact occupied with a molecule in the correct internal state when conditioned on being identified to be occupied in the initial image and dark in the error detection image (see Fig. 8).

In particular, we point out that if one can selectively remove the incorrectly prepared molecules perfectly ($s_{E,1} = 0$), the internal state purity is then given by $s_{E,0}$, the survival probability of a correctly prepared molecule. Note, however, that without knowing the location of the internal state error, it is converted into an occupation error, and the tweezer preparation fidelity is not enhanced.

3. Obtaining the Tweezer preparation fidelity

The tweezer preparation fidelity, which is the probability that a site initially identified to be occupied and free of internal state errors during midsequence error detection is indeed so, is the practically important metric for tweezer initialization in quantum simulation and quantum information processing applications. It can be expressed in terms of the optical pumping fidelity f_{OP} and the initial occupation fidelity s as

$$f_E = \frac{s f_{\text{OP}} s_{E,0} \bar{\epsilon}_{01}}{(1 - f_{\text{OP}}) s \epsilon_{10} + (s f_{\text{OP}} + (1 - s)) \bar{\epsilon}_{01}}. \quad (\text{F5})$$

Explicitly, the numerator is given by the number of correctly prepared occupied tweezers that are identified to be dark during the error detection image (column 3 in Fig. 8 multiplied by the identification error), and the denominator is the total number of tweezers identified to be dark during the error detection image. In Fig. 2(c) of the

main text, f_E is plotted versus the bare tweezer preparation fidelity $f = s f_{\text{OP}}$.

Because s is limited by the performance of the initial nondestructive image, whose optimization and improvement are separate topics from the current work, we provide a separate metric that allows isolating the limitations due to our internal state error detection scheme. In the inset of Fig. 2(c) in the main text, we provide f_E^0 versus f_{OP} , where f_E^0 is the tweezer preparation fidelity assuming no initial tweezer occupation errors ($s = 1$) given by

$$f_E^0 = \frac{f_{\text{OP}} s_{E,0} \bar{\epsilon}_{01}}{(1 - f_{\text{OP}}) \epsilon_{10} + f_{\text{OP}} \bar{\epsilon}_{01}}. \quad (\text{F6})$$

APPENDIX G: GENERATING DEFECT-FREE AND INTERNALLY PURE ARRAYS VIA REARRANGEMENT

In this appendix, we describe our protocol for preparing defect-free and internally pure arrays of molecules and the procedure to determine the array preparation fidelities.

Array preparation involves five steps. (1) A nondestructive image of a stochastically filled array of molecules is taken. Empty tweezers are turned off, and occupied tweezers are rearranged into a uniformly spaced defect-free (occupation) array, as described in our earlier work [24]. (2) A second nondestructive image is taken to verify successful rearrangement. This image is used to identify occupied tweezers. (3) Our measurement-enhanced internal state preparation procedure is applied, producing an error image. This image is used to identify tweezer instances with state preparation errors. (4) Another round of rearrangement is applied, replacing tweezer instances flagged as having an error with error-free and occupied tweezers. For each shot, the target array length equals the number of tweezers identified to be occupied and error-free. (5) Population is transferred from the target internal state $\mathcal{T}$ to $\mathcal{F}$ for destructive readout.

Next, we extract the raw success probability $P_{n,\text{raw}}$ of generating a defect-free and internally pure array of at least n molecules starting from the leftmost tweezer. This probability is conditioned on identifying at least n occupied and error-free tweezers. Since defect-free arrays can be shortened deterministically and with 100% probability by simply dropping tweezers, $P_{n,\text{raw}}$ gives the fidelity of preparing a defect-free, internally pure array of n molecules.

We next correct for measurement errors. The threshold used for the final destructive image keeps the false-positive probability negligible ($\epsilon_{01} < 0.1\%$) and the false-negative probability small ($\epsilon_{10} = 1.2\%$). In this limit, the measurement-corrected success rate is

$$P_n = \frac{P_{n,\text{raw}}}{(1 - \epsilon_{10})^n}. \quad (\text{G1})$$

In Fig. 2(d) of the main text, we show the measurement-corrected success probability as a function of array length. An exponential fit gives the single-particle rearrangement fidelity $f_R = 0.951(1)$, in good agreement with the error-excised state preparation fidelity presented in the main text.

APPENDIX H: MEASURING AND MODELING LIFETIME OF $X^2\Sigma(v=0, N=1,3)$ MOLECULES

Since molecules in $\mathcal{F} = X^2\Sigma(v=0, N=1)$ serve as error flags, it is important to quantify their population lifetime, which we measure in the pseudomagic configuration [$U = k_B \times 39(1)$ μ K, $B = 2.0$ G]. As discussed in the main text, molecules in the detection manifold $\mathcal{F}$ that leak to $X^2\Sigma(v=0, N=3)$ because of blackbody excitation can be repumped back into $\mathcal{F}$ using a rotational repumper, which is applied during the final destructive image.

We model population loss in $\mathcal{F}$ using a rate equation model that includes $X^2\Sigma(v=0)$ rotational states with odd N for $N \leq 13$, $X^2\Sigma(v=1)$ rotational states with even N for $N \leq 13$, and vacuum loss. We further assume that the population is initially distributed uniformly across all hyperfine states in $\mathcal{F}$ and all blackbody excitations and that decays keep populations uniformly distributed within each N manifold.

Figure 10 shows the measured $X^2\Sigma(v=0, N=1,3)$ total population decay and inferred populations in various rovibrational manifolds. Fitting our rate equation model to the data yields a vacuum loss rate of $\gamma_{\text{vac}} = 0.047(3)$ s^{-1} .

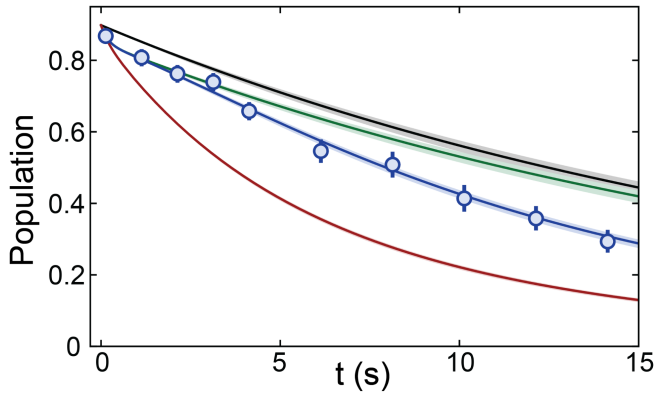


FIG. 10. Population decay of $X^2\Sigma(v=0, N=1)$ molecules. Blue circles show the measured total population in $X^2\Sigma(v=0, N=1,3)$ versus hold time t . The blue line shows a fit using our rate equation model, which incorporates blackbody excitation, spontaneous vibrational decay, and vacuum loss. Using our rate equation model and the vacuum loss rate determined from fitting the data, we infer various molecular populations. Simulation results (solid lines) and $\pm 1\sigma$ error bands (shaded bands) are shown for the total population (black), the $X^2\Sigma(v=0, N=1)$ population (red), the $X^2\Sigma(v=0, N=1,3)$ population (blue), and the $X^2\Sigma(v=0)$ population (green). The error bars are 68% Wilson-score intervals.

Our model, along with the fitted vacuum loss rate, allows us to infer populations in various rovibrational manifolds (Fig. 10) and attribute losses. At the measured lifetime of $\tau = 13.5(5)$ s, our model predicts of the molecules that survive vacuum loss, 32% of population is missed in the final readout image, of which 5% is due to equilibration with states in $v=1$ and 27% is due to a multistep excitation and decay process that populates $v=0, N > 5$ states.

1. Measuring population loss due to Raman light

In this section, we describe the procedures used to measure population errors in our hyperfine qubit when subject to Raman light. We characterize population loss due to single-photon scattering by applying a single Raman frequency component and measuring population loss. We measure a single-photon lifetime per Raman component of $\tau_{1\gamma} = 1.06(5)$ s and a population lifetime of $\tau_d = 4.3(2)$ s in the absence of Raman light. These lifetimes, combined with the π time of $t_\pi = 29$ μ s, allow us to estimate a single-photon limited population error per Raman π pulse of $\epsilon_p = 4.1(3) \times 10^{-5}$.

2. Estimating off-resonant scattering rates due to Raman light

To quantify the effect of off-resonant scattering by Raman light, we define the quality factor $Q_\pi = \Omega_R / (\pi\Gamma_{\text{sc}})$, which describes the ratio of the two-photon Rabi frequency Ω_R to the off-resonant scattering rate Γ_{sc} . For Raman light detuned near the $X - A^2\Pi_{1/2}$ and $X - A^2\Pi_{3/2}$ transitions, and assuming each Raman leg has equal intensity and σ^+ projections onto the quantization axis, the π -pulse quality factor Q_π for our choice of states is given by

$$Q_\pi = \frac{1}{\pi\Gamma} \frac{\left| \frac{1}{\Delta^2} - \frac{1}{(\Delta - \delta_{\text{rot}})^2} - \frac{1}{(\Delta - \delta_{\text{rot}} - \delta_{FS})^2} \right|}{\left(\frac{1}{\Delta^2} + \frac{1}{(\Delta - \delta_{\text{rot}})^2} + \frac{1}{(\Delta - \delta_{\text{rot}} - \delta_{FS})^2} \right)}, \quad (\text{H1})$$

where Δ is the single-photon detuning from the $X^2\Sigma(v=0, N=0) \rightarrow A^2\Pi_{1/2}(v=0, J=1/2, -)$ transition, $\delta_{\text{rot}} = 2\pi \times 33$ GHz is the angular frequency difference between $A^2\Pi_{1/2}(v=0, J=1/2, -)$ and $A^2\Pi_{1/2}(v=0, J=3/2, -)$, and $\delta_{FS} = 2\pi \times 2120$ GHz is the angular frequency difference between $A^2\Pi_{1/2}(v=0, J=3/2, -)$ and $A^2\Pi_{3/2}(v=0, J=3/2, -)$.

Figure 11 shows Q_π versus the single-photon detuning Δ for X-A Raman light. In a near-detuned X-A Raman configuration ($\Delta = -2\pi \times 19.6$ GHz), we predict $Q_\pi = 630$. From single-photon scattering rate measurements, we find $Q_\pi = 374(29)$, which is close to but lower than the theoretical value by a factor of 1.7(1). At the optimal single-photon detuning, we predict $Q_\pi = 20.3 \times 10^3$, in good agreement with the measured $Q_\pi = 24(2) \times 10^3$.

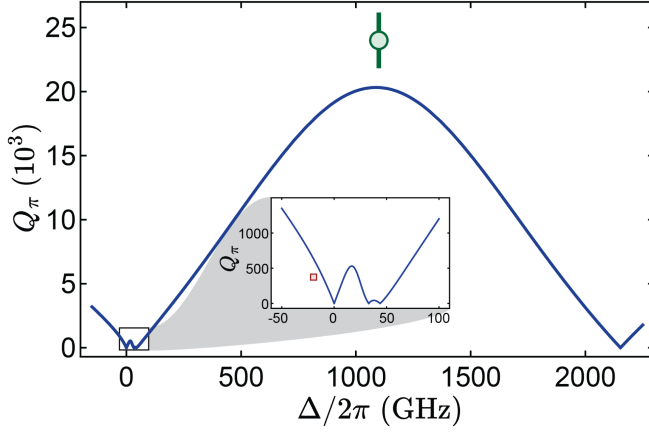


FIG. 11. Raman π -pulse quality factor. The theoretical Raman π -pulse quality factor $Q_\pi = \Omega_R/(\pi\Gamma_{sc})$ gives an upper bound on the number of Raman π pulses that can be applied with $1/e$ of population remaining. The quality factor (blue curve) is plotted vs single-photon detuning $\Delta/2\pi$ from the $X^2\Sigma(N=0) \rightarrow A^2\Pi_{1/2}(J=1/2, -)$ transition. The empirically measured quality factor at $\Delta \approx 1.1$ THz (green circle) matches the theoretical upper bound. In the inset, we show that the quality factor at $\Delta = -2\pi \times 19.6$ GHz (red square) is a factor of 1.7(1) lower than theory. The error bars are computed from 1σ fitting errors obtained when determining Ω_R and Γ_{sc} .

APPENDIX I: QUBIT TRANSFER BETWEEN ROTATIONAL AND HYPERFINE ENCODINGS

In this appendix, we provide supplemental information and data showing compatibility between our hyperfine qubit encoding used for erasure conversion and the previously used rotational encoding where a two-qubit gate has been demonstrated.

The building blocks for transferring a qubit between the rotational encoding and the hyperfine encoding are π pulses along the Raman and microwave transitions [Fig. 12(a)]. For these data, due to technical limitations, Raman light detuned from the $X-B$ transition was used. Specifically, the Raman light is detuned halfway between the $X^2\Sigma(N=0) \rightarrow B^2\Sigma(N=1, J=1/2)$ and $X^2\Sigma(N=0) \rightarrow B^2\Sigma(N=1, J=3/2)$ transitions. This process results in a single-photon detuning of 500 MHz for optical transitions resonant with the hyperfine qubit states. For this scheme, Q_π is measured to be 37(3).

We characterize the ability to transfer arbitrary states between the rotational ($|\uparrow\rangle - |\downarrow\rangle$) and hyperfine encoding ($|\downarrow\rangle - |0\rangle$) by comparing single-particle coherence after four different Ramsey sequences: two in which no transfer occurs and two in which a single transfer occurs [Fig. 12(b)]. We find that the decrease in single-particle coherence is consistent with zero after one round of transfer [Figs. 12(c) and 12(d)]. To more precisely estimate the transfer efficiency, we measure single-particle coherence as a function of repeated transfers between the two different qubit encodings [Fig. 12(e)]. From these measurements, we

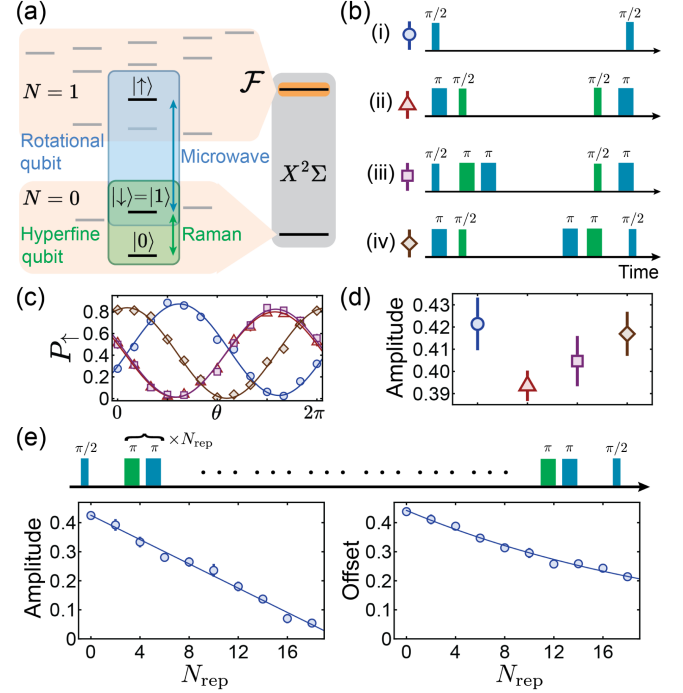


FIG. 12. Coherent qubit transfer between rotational and hyperfine encodings. (a) Rotational and hyperfine qubit encodings. (b) Four Ramsey sequences (i)–(iv) used to probe various coherences between the three states involved in the two qubit encodings. Blue (green) boxes depict microwave pulses on the rotational encoding $|\uparrow\rangle - |\downarrow\rangle$ (hyperfine encoding $|1\rangle - |0\rangle$). Sequences (i)–(ii) probe rotational coherence and hyperfine coherence, respectively. Sequences (iii) and (iv) verify coherences between all three internal states. (c) $P_\uparrow$ versus the phase θ of the final $\pi/2$ pulse for the four Ramsey sequences. The corresponding markers are indicated in panel (b). (d) Amplitude of $P_\uparrow$ Ramsey fringes for the four Ramsey sequences. The corresponding markers are indicated in panel (b). (e) Characterizing efficiency of switching qubit encodings. A superposition initially created in the rotational encoding is repeatedly transferred to and from the hyperfine encoding using a Raman and microwave π pulse. Amplitude and offset of Ramsey fringes versus the number of transfers N_{rep} . The error bars in panels (c) and (e) are 68% Wilson-score intervals. The error bars in panel (d) are 1σ fitting errors.

find a transfer fidelity of $f = 0.951(2)$, limited by off-resonant scattering of the Raman light. We find that off-resonant scattering accounts for 89(9)% of the infidelity, with the remaining amount consistent with the separately measured microwave transfer infidelity. We therefore expect the transfer fidelity to improve to $f \approx 0.995$ when using optimally detuned Raman light on the $X-A$ transition.

APPENDIX J: MODELING BLACKBODY LEAKAGE ERRORS

In this appendix, we describe a model that we use to capture population dynamics due to blackbody vibrational

excitation and subsequent decay. This model allows us to extract an erasure conversion efficiency from the measured data.

Blackbody errors occur when population $p_{\mathcal{T}}$ from the target qubit manifold ($\mathcal{T} = \{|0\rangle, |1\rangle\}$) is excited to $X^2\Sigma(v=1, N=1)$ via absorption of a thermal blackbody photon, which occurs at a rate of γ_{BB} . This population is converted to population $p_{\mathcal{F}}$ in the detectable states [$\mathcal{F} = X^2\Sigma(v=0, N=1)$] by the $v=1$ vibrational repumper that is applied continuously midsequence in our blackbody erasure conversion scheme. However, if the rate of vibrational repumping is too slow or if molecules are lost in the repumping process, part of the population can end up in states outside of $\mathcal{T}$, which cannot be detected in our scheme. These decay events reduce the erasure conversion efficiency η .

In the limit where the conversion is much faster than all other timescales, the populations are described by the rate equation model:

$$\begin{aligned}\dot{p}_{\mathcal{T}} &= -(\gamma_{\text{vac}} + \gamma_{BB})p_{\mathcal{T}}, \\ \dot{p}_{\mathcal{F}} &= \eta\gamma_{BB}p_{\mathcal{T}} - \gamma_{\text{vac}}p_{\mathcal{F}},\end{aligned}\quad (\text{J1})$$

with initial conditions of $p_{\mathcal{T}}(0) = s$ and $p_{\mathcal{F}}(0) = 0$, because the molecules are initially in the hyperfine qubit manifold.

The population that is successfully pumped to $\mathcal{F} = X^2\Sigma(v=0, N=1)$ is subsequently detected via rapid resonant imaging, with a true positive probability of $\bar{\epsilon}_{10} = 1 - \epsilon_{10}$. The erasure-excised probability is then

$$p_{E,\mathcal{T}}(t) = \frac{p_{\mathcal{T}}(t)}{1 - p_{\mathcal{F}}(t)\bar{\epsilon}_{10}}. \quad (\text{J2})$$

In the experiment, $p_{\mathcal{T}}(t)$ and $p_{\mathcal{F}}(t)$ are directly measured. In addition, $p_{E,\mathcal{T}}$ is available via postselection. These probabilities can be used to extract parameters or to provide verification of our loss model. For example, using the measured decay of $p_{\mathcal{T}}(t)$ along with the value of γ_{vac} extracted from the $X^2\Sigma(v=0, N=1, 3)$ lifetime data, we obtain a blackbody rate of $\gamma_{BB,\text{exp}} = 4.0(2) \text{ s}^{-1}$, comparable to the blackbody theory prediction of $\gamma_{BB,\text{th}} = 3.4 \text{ s}^{-1}$ [56].

1. Early-time dynamics

At short times, the populations evolve linearly versus time, and we obtain

$$\begin{aligned}p_{\mathcal{T}}(t) &= s[1 - (\gamma_{\text{vac}} + \gamma_{BB})t], \\ p_{\mathcal{F}}(t) &= s\eta\gamma_{BB}t,\end{aligned}\quad (\text{J3})$$

where s is the initial nondestructive survival probability. To first order in t , we obtain

$$p_{E,\mathcal{T}}(t) = s[1 - (\gamma_{\text{vac}} + \gamma_{BB})t + s\eta\gamma_{BB}\bar{\epsilon}_{10}t], \quad (\text{J4})$$

giving an erasure-excised population decay rate of

$$\gamma_{p,E} = -\frac{1}{s} \frac{dp_{E,\mathcal{T}}(t)}{dt} = \gamma_{\text{vac}} + \gamma_{BB}(1 - s\eta\bar{\epsilon}_{10}). \quad (\text{J5})$$

Note that $\gamma_{p,E}$, γ_{vac} , γ_{BB} , s , and $\bar{\epsilon}_{10}$ are all separately measured. Additional losses and imperfections can be lumped into η . We can therefore combine all imperfections such as imperfect identification ($\bar{\epsilon}_{10}$), imperfect erasure conversion, and blackbody equilibration into a single parameter η_{eff} and write

$$\gamma_{p,E} = -\frac{1}{s} \frac{dp_{E,\mathcal{T}}(t)}{dt} = \gamma_{\text{vac}} + \gamma_{BB}(1 - s\eta_{\text{eff}}). \quad (\text{J6})$$

Fitting to our measured erasure-excised population decay data, we obtain $\eta_{\text{eff}} = 0.78(8)$ when using our measured value of $\gamma_{BB,\text{exp}} = 4.0(2) \text{ s}^{-1}$. Using the theoretical value of $\gamma_{BB,\text{th}} = 3.4 \text{ s}^{-1}$ gives a similar value of $\eta_{\text{eff}} = 0.84(4)$.

2. Intermediate-time dynamics

At intermediate times, blackbody vibrational excitations from $X^2\Sigma(v=0)$ to $X^2\Sigma(v=1)$ and spontaneous radiative decays back to $X^2\Sigma(v=0)$ lead to a steady-state $X^2\Sigma(v=1)$ fraction of

$$f_{\text{term}} = \frac{e^{-\hbar\omega_{01}/k_B T}}{1 + e^{-\hbar\omega_{01}/k_B T}}, \quad (\text{J7})$$

where $\omega_{01} = 2\pi \times 17.5 \text{ THz}$ is the vibrational splitting between $X^2\Sigma(v=0)$ and $X^2\Sigma(v=1)$ and $T = 295 \text{ K}$ is the ambient lab temperature. Numerically, the $v=1$ fraction is $f_{\text{therm}} = 5.5\%$, and equilibration occurs at a rate of

$$\gamma_{\text{therm}} = 2\gamma_{BB} + \Gamma_{1,\text{rad}}, \quad (\text{J8})$$

where $\Gamma_{1,\text{rad}}$ is the $v=1$ to $v=0$ decay rate. Together, we obtain $\gamma_{\text{therm}} = 5.3 \text{ s}^{-1}$, corresponding to a characteristic timescale of $\tau_{\text{therm}} = 1/\gamma_{\text{therm}} \approx 190 \text{ ms}$.

Hence, for lifetime measurements that include wait times much longer than τ_{therm} , such as those presented in the main text out to $t = 2 \text{ s}$, equilibration dynamics could be present. However, they do not result in a substantial change in the estimated decay rates, as we will show in the next section.

3. Comparison with measurements

In this section, we discuss comparisons of our model with the measured population dynamics. Note that $\gamma_{p,E}$, γ_{vac} , γ_{BB} , s , and $\bar{\epsilon}_{10}$ are all separately measured, and η is assumed to be unity since the vibrational repump is continuously applied.

TABLE II. Erasure-corrected $N = 0$ population lifetime. Predictions for the erasure-corrected $N = 0$ population lifetime are given in the presence (Yes) or absence (No) of immediate blackbody population equilibration between $v = 0$ and $v = 1$ (f_{therm}), and using the theoretical (Th) or experimentally measured (Exp) blackbody excitation rate (γ_{BB}).

f_{therm}	γ_{BB}	$\gamma_{\text{p,E}}$
Yes	Th	0.118(4)
Yes	Exp	0.107(5)
No	Th	0.105(4)
No	Exp	0.096(4)

We perform four estimates of $\gamma_{\text{p,E}}$ that either assume immediate equilibration of $p_{v=1}$ to $f_{\text{therm}} = 5.5\%$ or no equilibration ($f_{\text{therm}} = 0$) at all, along with either the theoretical value or experimentally extracted value for γ_{BB} . Specifically, we use the measured vacuum lifetime γ_{vac} , erasure identification error $\bar{\epsilon}_{10}$, and initial overall survival s , but we use the theoretical value of $\eta = 1$.

The results are summarized in Table II and range from 0.096 s^{-1} to 0.118 s^{-1} with error bars of approximately 0.05 s^{-1} . These values are in tension with the measured erasure-corrected population lifetime of $\gamma_{\text{p,E}} = 0.14(1) \text{ s}^{-1}$, suggesting some degree of loss during vibrational repumping, which reduces the overall conversion efficiency to $\eta \approx 85\%$.

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