# Supplementary Information: Dipole-phonon quantum logic with alkaline-earth monoxide and monosulfide cations

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## 1. EFFECT OF ELECTRIC-FIELD NOISE ON INTERNAL STATES OF MOLECULAR IONS

There are many sources of electric-field noise in ion traps [1]: fluctuating electrical fields from injected circuits, varying patch potentials from the trap surface, Johnson noise from the resistance of trap electrodes, and so on. In the DPQL procedure, these noise sources could directly couple to the dipole moments of molecular ions and thus cause transitions between $\Lambda$-doublet splittings and rotational states as well as via ion motion. Here we consider two major forms of electric-field noise[2]: one is a fluctuating, uniform electrical field $\zeta(t)$; the other is a fluctuating quadrupole field, which perturbs the spring constant of the trap by $\Delta K = \epsilon(t) M \omega_z^2$, where $M$ is ion mass and $\omega_z$ is the axial secular frequency. The Hamiltonian under the above noise model can then be written as:

$$H = \frac{P^2}{2M} + \frac{1}{2} M(1 + \epsilon(t)) \omega_z^2 z^2 + \zeta(t) q z. \tag{S1}$$

Here $P$ is the ion momentum and $q$ is the charge of ion. Using first-order time-dependent perturbation theory, we can derive the heating rate near the motional ground state[3, 4]:

$$\frac{d}{dt}\langle n \rangle = \frac{\pi \omega_z^2}{4} S_\epsilon(2\omega_z) + \frac{q^2}{2M\hbar\omega_z} S_\zeta(\omega_z), \tag{S2}$$

where $S_j(\omega) = \int_{-\infty}^{\infty} d\tau \langle j(t)j(t+\tau)\rangle e^{i\omega\tau}$ (with $j = \epsilon, \zeta$) is the power spectral density of the fluctuating $\epsilon(t)$ and $\zeta(t)$ respectively and $|n\rangle$ is the motional state. Here, we neglect the higher order term $\langle \epsilon^2(t)\zeta^2(t)\rangle$. The first term in eqn (S2) is from the quadrupole field fluctuation. For a molecular ion that is sympathetically cooled and well compensated from excess micromotion, the coupling between the quadrupole field fluctuation and the molecular ion dipole is small since the electric field noise is close to zero at the RF null. The second term couples the molecular ion dipole through $S_\zeta(\omega)$, which results in a spectral energy density $\rho_H(\omega)$

$$\rho_H(\omega) = \epsilon_0 S_\zeta(\omega) = \frac{2M\hbar\epsilon_0\omega_z\Gamma^*}{q^2}, \tag{S3}$$

where $\epsilon_0$ is the permittivity of free space and $\Gamma^*$ is the heating rate caused by $\zeta(t)$. We model the frequency dependence of the electric field noise as a single power law[5], $S_\zeta(\omega) \propto f^{-0.6}$ over the $\Lambda$-doublet splitting range ($\omega < 2\pi \times 100$ MHz) and broadband noise for higher frequencies. Here we consider the co-trapped $Ca^+$ and $CaO^+$ to have an axial COM frequency of $\omega_z = 2\pi \cdot 450$ kHz and the COM heating rate to be $\Gamma^* = 300$ quanta/s. For $\Lambda$-doublet of $J = 7/2$, electric-field noise results in a transition rate $B_{ef}\rho_H(\omega) \sim 0.001$ s⁻¹, where $B_{ef}$ is the Einstein B coefficient for the transition. This transition rate is negligible compared to the thermalization process from blackbody radiation, in which the blackbody-limited life time is 4 s. This result indicates that electric-field noise could affect the DPQL process via motional heating with effective dipole moment of order 10 $e\cdot$nm, but not via directly coupling with dipole moment of molecular ion which is on the order of 0.1 $e\cdot$nm. We also calculate the effect of electric-field noise on the rotation state distribution under room temperature using the rate equation model. The results shows that the blackbody-limited lifetime of the states is not affected.

## 2. NUMERICAL SIMULATION

To demonstrate some two-molecule techniques, we present the results of a numerical simulation of the Hamiltonian

$$H = \sum_{q,i} \left( \frac{\omega_{\text{mol}}^{(i)}}{2} \sigma_z^{(i)} + \omega_q \left( a_q^\dagger a_q + \frac{1}{2} \right) + \frac{g_q^{(i)}}{2} \sigma_x^{(i)} \left( a_q^\dagger + a_q \right) \right) \tag{S4}$$

where $\hbar = 1$, $\omega_{\text{mol}}^{(i)}$ is the energy splitting of molecule $(i)$, $\omega_q$ is the secular frequency of the normal mode $q$ along the trap axis, $\sigma_x^{(i)} = \left| e^{(i)} \right\rangle \left\langle f^{(i)} \right| + \left| f^{(i)} \right\rangle \left\langle e^{(i)} \right|$, and $\sigma_z^{(i)} = \left| f^{(i)} \right\rangle \left\langle f^{(i)} \right| - \left| e^{(i)} \right\rangle \left\langle e^{(i)} \right|$. Here, $g_q^{(i)} \equiv d\mathcal{E}_{0,q}^{(i)} = \frac{d}{e} \sqrt{2m^{(i)}\omega_q^3} b_q^{(i)}$ is the vacuum Rabi frequency of the interaction where $\mathcal{E}_{0,q}^{(i)}$ is the electric field amplitude at the position of ion $(i)$ due to a single phonon in normal mode $q$, $m^{(i)}$ is the mass of ion $(i)$, $e$ is the elementary charge, and $b_q^{(i)}$ is the component of the eigenvector of normal mode $q$ at ion $(i)$. For simplicity, the effect of motional heating is not included in these simulations.

We present the results of numerical simulations of a three-ion chain with two CaO$^+$ molecular ions in the $X\,^2\Pi_{3/2}$ state, with $J = 7/2$ on the outside ends of the chain and a single Ca$^+$ atomic ion at the center of the chain. We present simulations of various techniques for state preparation and for demonstrating the virtual-phonon-mediated dipole-dipole interaction. To demonstrate the results, we plot the "average excitation" of each molecule $(i)$ and each normal mode $q$. We define the "average excitation" of molecule $(i)$ to be the probability of measuring the molecule in the $\left| f^{(i)} \right\rangle$ state $\left| \left\langle f^{(i)} \middle| \psi(t) \right\rangle \right|^2$. We define the "average excitation" of normal mode $q$ to be the average phonon number $\langle n_q \rangle = \langle \psi(t) | a_q^\dagger a_q | \psi(t) \rangle$.

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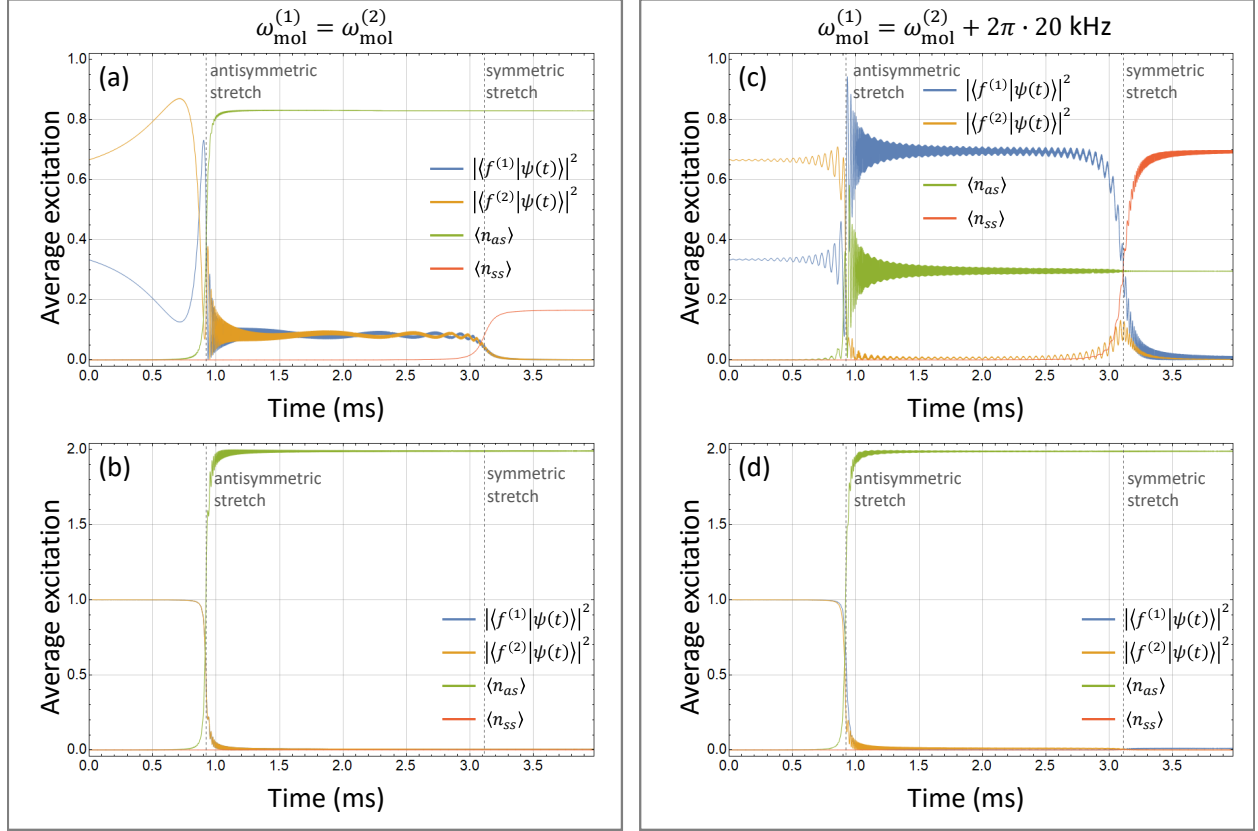
**Fig. S1.** Simulation of a frequency sweep to prepare the $|ee\rangle$ state. We begin with all normal mode frequencies below $\omega_{mol}^{(1)}$ and $\omega_{mol}^{(2)}$ in the motional ground state for all normal modes. We then increase the single-molecule trap frequency $\omega_0$ linearly at a rate of $\dot{\omega}_0 = 3.0 \times 10^9$ rad/s$^2$ until both the antisymmetric and symmetric stretch mode frequencies pass through $\omega_{mol}^{(1)}$ and $\omega_{mol}^{(2)}$. (a) With equal splittings ($\omega_{mol}^{(1)} = \omega_{mol}^{(2)}$), we select an arbitrary single-molecular-excitation initial state $|\psi_{mol}(0)\rangle = \sqrt{\frac{2}{3}} |ef\rangle + \sqrt{\frac{1}{3}} e^{i\pi/4} |fe\rangle$. (b) With equal splittings, we start with $|\psi_{mol}(0)\rangle = |ff\rangle$. In this plot, the average excitations for molecules (1) and (2) are overlapping within the thickness of the line. (c) $\omega_{mol}^{(1)} = \omega_{mol}^{(2)} + 2\pi \cdot 20$ kHz and $|\psi_{mol}(0)\rangle = \sqrt{\frac{2}{3}} |ef\rangle + \sqrt{\frac{1}{3}} e^{i\pi/4} |fe\rangle$. Far away from resonance, $\left|\omega_{mol}^{(1)} - \omega_{mol}^{(2)}\right| \gg |J_{12}|$, and the amplitude of oscillation between $|fe\rangle$ and $|ef\rangle$ is small. As one of the normal mode frequencies approaches resonance, the coupling strength $J_{12}$ increases, and the previously stated limit is no longer valid. In this regime, there are nontrivial oscillations between $|fe\rangle$ and $|ef\rangle$. (d) $\omega_{mol}^{(1)} = \omega_{mol}^{(2)} + 2\pi \cdot 20$ kHz and $|\psi_{mol}(0)\rangle = |ff\rangle$. Again, the average excitations for molecules (1) and (2) are overlapping within the thickness of the line. In every case shown, sweeping the frequency such that the antisymmetric and symmetric stretch mode frequencies pass through $\omega_{mol}^{(1)}$ and $\omega_{mol}^{(2)}$ removes any molecular excitation and prepares the molecular state in $|ee\rangle$. Additionally, if the initial molecular state is $|\psi_{mol}(0)\rangle = |ff\rangle$, such a sweep will yield two phonons in the antisymmetric stretch mode—as this is the first mode to sweep through resonance—regardless of the difference in molecular splittings.
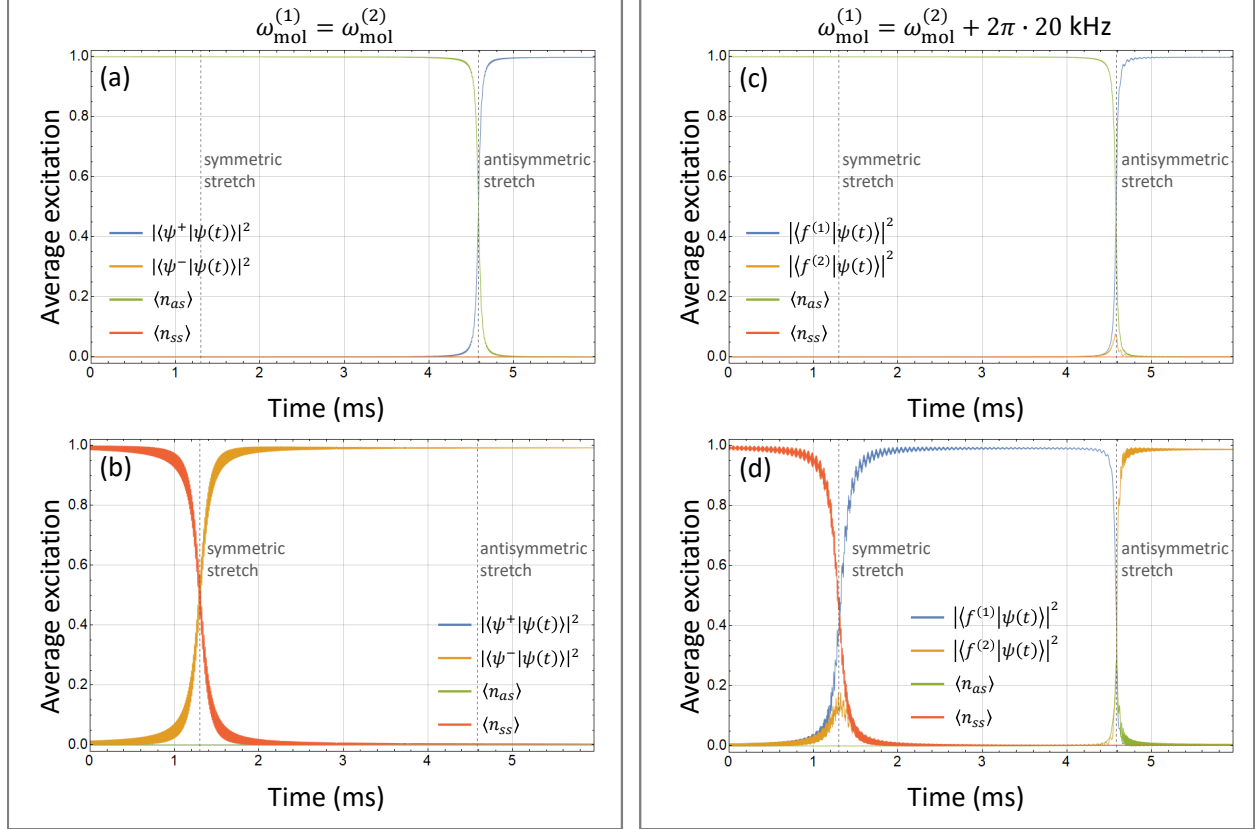
**Fig. S2.** Simulation of molecular state preparation techniques. For each simulation, the initial molecular state is $|ee\rangle$ with a single phonon in either the symmetric or antisymmetric stretch mode, and the initial single-molecule trap frequency is tuned such that the symmetric and antisymmetric stretch mode frequencies are above $\omega_{mol}^{(1)}$ and $\omega_{mol}^{(2)}$. The single-molecule trap frequency is decreased at a rate of $\dot{\omega}_0 = -2.0 \times 10^9$ rad/s² until the antisymmetric and symmetric stretch mode frequencies are below $\omega_{mol}^{(1)}$ and $\omega_{mol}^{(2)}$. (a) With $\omega_{mol}^{(1)} = \omega_{mol}^{(2)}$, starting with a single phonon in the antisymmetric stretch mode yields the Bell state $|\psi^+\rangle = (|fe\rangle + |ef\rangle)/\sqrt{2}$. (b) With $\omega_{mol}^{(1)} = \omega_{mol}^{(2)}$, starting with a single phonon in the symmetric stretch mode yields the Bell state $|\psi^-\rangle = (|fe\rangle - |ef\rangle)/\sqrt{2}$. (c) With $\omega_{mol}^{(1)} = \omega_{mol}^{(2)} + 2\pi \cdot 20$ kHz, starting with a single phonon in the antisymmetric stretch mode yields $|fe\rangle$. (d) With $\omega_{mol}^{(1)} = \omega_{mol}^{(2)} + 2\pi \cdot 20$ kHz, starting with a single phonon in the symmetric stretch mode yields $|ef\rangle$.
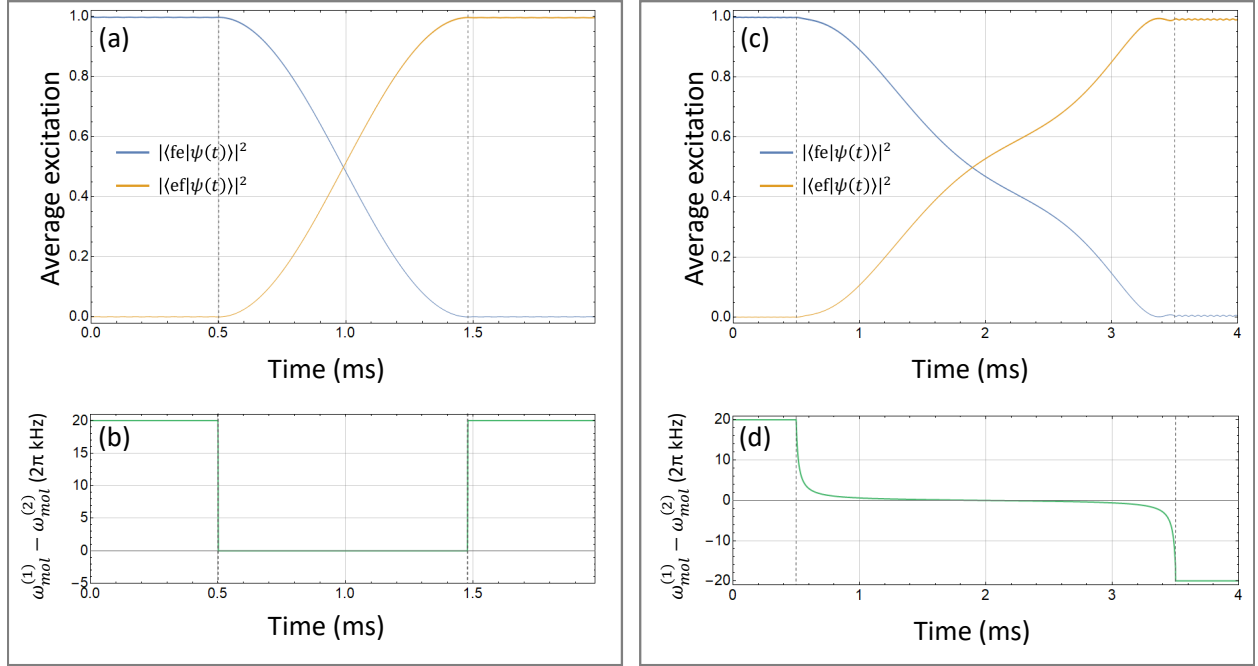
**Fig. S3.** Two different methods to transfer the population from $|fe\rangle$ to $|ef\rangle$. In the first method, depicted in (a) and (b), we start with the molecular splittings different by $2\pi \cdot 20$ kHz. To begin the transfer, we quickly set this difference to zero, allowing the population to coherently oscillate between $|fe\rangle$ and $|ef\rangle$. After waiting $t_\pi = \frac{\pi}{2J_{12}}$, where $J_{12}$ is the virtual-phonon-mediated dipole-dipole interaction strength, we restore the difference in molecular splittings to stop this oscillation. The average molecular excitation of molecules (1) and (2) are shown in (a) and the difference in molecular splittings as a function of time is shown in (b). For the second method, shown in (c) and (d), we adiabatically vary the difference in molecular splitting. To achieve a high probability transfer with a linear sweep of the molecular splitting difference, it is necessary for the sweep of the molecular splitting difference $|\dot\Delta| \ll J_{12}^2$. To dramatically decrease the time needed to perform an adiabatic sweep, we change the molecular splitting difference in a nonlinear fashion, with ramping speed $\dot\Delta(t)$. The ramp is faster for larger differences in molecular splittings. For all times, we maintain the limit $|\dot\Delta(t)| \ll J_{12}^2 + \left(\frac{1}{2}(\omega_{\text{mol}}^{(1)} - \omega_{\text{mol}}^{(2)})\right)^2$.