

the excited $^2P_{1/2}$ state and performing the rotating-wave approximation, the resonant Rabi frequency of Raman transitions between the qubit states is given by a sum over all spectral components of the comb teeth as indicated in Fig. 1 ($\hbar = 1$):

$$\Omega = \frac{|\mu|^2 \sum_l E_l E_{l-q}}{\Delta} \approx \Omega_0 \left(\frac{\omega_0 \tau}{e^{\omega_0 \tau/2} - e^{-\omega_0 \tau/2}} \right), \quad (2)$$

where μ is the dipole matrix element between the ground and excited electronic states, $E_k \equiv \nu_R \tilde{f}(2\pi k \nu_R)$, and q is an integer. In the approximate expression above, the sum is replaced by an integral and each pulse is described by $f(t) = \sqrt{\pi/2} E_0 \text{sech}(\pi t/\tau)$ with $\tau \ll T$, where $\Omega_0 = (\nu_R \tau) |\mu E_0|^2 / \Delta = s \gamma^2 / 2\Delta$ is the time-averaged resonant Rabi frequency of the pulse train and $s = \bar{I}/I_{\text{sat}}$ is the average intensity $\bar{I} = \nu_R c \epsilon_0 / 2 \int dt |f(t)|^2$ scaled to the $^2S_{1/2} \leftrightarrow ^2P_{1/2}$ saturation intensity. Note the net transition rate is suppressed unless the single-pulse bandwidth is large compared to the hyperfine frequency ($\omega_0 \tau \ll 1$), in which case $\Omega \approx \Omega_0$. In our experiments, $\omega_0 \tau \approx 0.08$.

For $I_{\text{sat}} = 0.15 \text{ W/cm}^2$, the data shown in Fig. 3 is consistent with an average intensity $\bar{I} \approx 500 \text{ W/cm}^2$.

In order to entangle multiple ions, we first address the motion of the ion by resolving motional sideband transitions. As depicted in Fig. 2, the pulse train is split into two perpendicular beams with wavevector difference k along the x -direction of motion. Their polarizations are mutually orthogonal to each other and to a weak magnetic field that defines the quantization axis [20]. We control the spectral beatnotes between the combs by sending both beams through AO modulators (driven at frequencies ν_1 and ν_2), imparting a net offset frequency of $\Delta\omega/2\pi = \nu_1 - \nu_2$ between the combs. For instance, in order to drive the first upper/lower sideband transition we set $|2\pi j \nu_R + \Delta\omega| = \omega_0 \pm \omega_t$, with j an integer and ω_t the trap frequency. In order to see how the sidebands are spectrally resolved, we consider the following Hamiltonian of a single ion and single mode of harmonic motion interacting with the Raman pulse train:

$$H_{\text{eff}} = \omega_t a^\dagger a + \frac{\omega_0}{2} \sigma_z + \frac{\theta_p}{2} \sum_n \delta(t - nT) \left(\sigma_+ e^{i(k\hat{x} + \Delta\omega t)} + \sigma_- e^{-i(k\hat{x} + \Delta\omega t)} \right), \quad (3)$$

where $\theta_p = \Omega T$ is the change in the Bloch angle due to a single pulse, σ_z is the Pauli-z operator, σ_\pm are raising and lowering operators, \hat{x} is the x -position operator of the trapped ion, a^\dagger and a are the raising and lowering operators of the x -mode of harmonic motion and the q parameter has been assumed to not be an integer or half-integer. In the interaction picture, the evolution operator after N pulses is given by V^N , where

$$V = \exp[-iH_0 T] \exp\left[\frac{-i\theta_p}{2} (\sigma_+ e^{ik\hat{x}} + \sigma_- e^{-ik\hat{x}})\right] \quad (4)$$

and $H_0 = \omega_t a^\dagger a + 1/2(\omega_0 + \Delta\omega)\sigma_z$. The time evolution operator is given by,

$$V^N = \exp[-iH_0 NT] \left(\hat{I} - i \frac{\theta_p}{2} \sum_{n=0}^{N-1} Q_n + \mathcal{O}(\theta_p^2) \right) \quad (5)$$

$$Q_n \equiv \sigma_+ e^{i(\omega_0 + \Delta\omega)nT} D(i\eta e^{i\omega_t nT}) + h.c., \quad (6)$$

where $D(\alpha) = \exp[\alpha a^\dagger - \alpha^* a]$ is the harmonic oscillator displacement operator in phase space, and $\eta = k\sqrt{\hbar/2m\omega_t}$ is the Lamb-Dicke parameter. In the Lamb-Dicke regime, $\eta\sqrt{\langle a^\dagger a \rangle + 1} \ll 1$, we can write $D(i\eta e^{i\omega_t nT}) \approx 1 + i\eta(e^{i\omega_t nT} a^\dagger + e^{-i\omega_t nT} a)$ turning the sum in Eq. (5) into a geometric series. If, for example, the offset frequency between the combs $\Delta\omega$ is tuned

to satisfy the resonance condition for the red sideband, $\vartheta_r \equiv (\omega_0 + \Delta\omega - \omega_t)T = 2\pi j$, where j is an integer, then the sum in Eq. (5) is approximately given by,

$$\sum_{n=0}^{N-1} Q_n \approx i\eta \frac{\sin N\vartheta_r/2}{\sin\vartheta_r/2} e^{i\vartheta_r(N-1)/2} \sigma_+ a + h.c. \quad (7)$$

The coefficient in Eq. (7) is the same as the field amplitude created by a diffraction grating of N slits, whose narrow peaks have an amplitude equal to N . In the limit $\omega_t T \ll 1$, the other terms in Eq. (5) that drive the carrier and other sideband transitions can be neglected when $N \gg (\omega_t T \eta)^{-1}$. This is analogous to the destructive interference of amplitudes away from the bright peaks in a diffraction grating. For $\omega_t/2\pi = 1.64 \text{ MHz}$, $T = 12.4 \text{ ns}$ and $\eta = 0.1$, the sidebands are well-resolved when $N \gg 80$.

For many applications in quantum information, the motional modes of the ion must be cooled and initialized to a nearly pure state. Fig. 4 shows that the pulsed laser can also be used to carry out the standard techniques of sideband cooling [20] to prepare the ion in the motional ground state with near unit fidelity. The set-up also easily lends itself to implementing a two-qubit entangling gate by applying two fields whose frequencies are symmetrically detuned from the red and blue sidebands [21, 22]. By simultaneously applying two modulation fre-