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Photon scattering errors during stimulated Raman transitions in trapped-ion qubits

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We study photon scattering errors in stimulated Raman driven quantum logic gates. For certain parameter regimes, we find that previous, simplified models of the process significantly overestimate the gate error rate due to photon scattering. This overestimate is shown to be due to previous models neglecting the detuning dependence of the scattered photon frequency and Lamb-Dicke parameter, a second scattering process, interference effects on scattering rates to metastable manifolds, and the counter-rotating contribution to the Raman transition rate. The resulting improved model shows that there is no fundamental limit on gate error due to photon scattering for electronic ground state qubits in commonly-used trapped-ion species when the Raman laser beams are red detuned from the main optical transition. Additionally, photon scattering errors are studied for qubits encoded in metastable $D_{5/2}$ manifold, showing that gate errors below 10^{-4} are achievable for all commonly-used trapped ions.

I. INTRODUCTION

Stimulated Raman transitions are often used to implement trapped-ion quantum logic gates. In such setups, a pair of laser beams drives a two-photon stimulated transition between qubit states $|0\rangle$ and $|1\rangle$ through one or more intermediate states. Photon scattering during this process is unavoidable and understanding the fundamental limit it places on achievable gate fidelity is potentially crucial for the viability of trapped-ion quantum computing.

Although photon scattering errors have been previously studied [1, 2], we revisit the topic for two reasons. First, past work [1] produced a model of scattering that, while suitable for explaining moderate-detuning gate errors, is inaccurate at larger detunings. This inaccuracy is due to neglecting the detuning dependence of the scattered photon frequency and Lamb-Dicke parameter, contributions of a second scattering term (the ‘V’ scattering process in Fig. 2), interference effects in scattering to the metastable manifolds, and the contribution of the counter-rotating component of the laser electric field to the Raman transition rate. As shown below, including such effects changes the limiting behavior of the model, resulting in no lower bound on gate error, in contrast to the predictions of [1].

Second, the previous studies focused solely on qubits with either one or both states encoded in the $S_{1/2}$ ground manifold of trapped ions (‘ o ’ qubits and ‘ g ’ qubits, respectively). However, a recent proposal [3] has shown that encoding qubits in

metastable states of trapped ions (‘ m ’ qubits) has several important advantages over g and o type encodings. Therefore, we also study photon scattering errors for m qubits.

Below, after describing our qubit choices and defining our model, we calculate the photon-scattering-induced errors in single-qubit and two-qubit gates in m qubits and compare them to g qubits. There are many physical differences between m and g qubits, such as the dipole coupling between the qubit states and $P_{1/2}$ that exists for g qubits but is absent in m qubits, or certain scattering terms present in m qubits that are not present in g qubits. These physical differences exert influences of varying magnitudes on the quantitative scattering error, but on net, they tend to increase the detuning from resonance required for a certain error in m qubits relative to g qubits. As for the qualitative scattering behavior, we find that the main difference between the two schemes is the existence of a lower bound on two-qubit gate errors in m qubits which is absent in g qubits. However, for all trapped ions considered in this work, this lower bound is sufficiently small (less than 10^{-4}) that low-overhead error correction is possible for Raman gates in the m qubit scheme.

After describing the overall behavior of the two different qubit schemes in the large-detuning model, we discuss the contributions of higher-lying levels to the scattering rates of the m qubit model. Additionally, for both m and g qubits, we estimate the contribution to gate error from Rayleigh scattering.

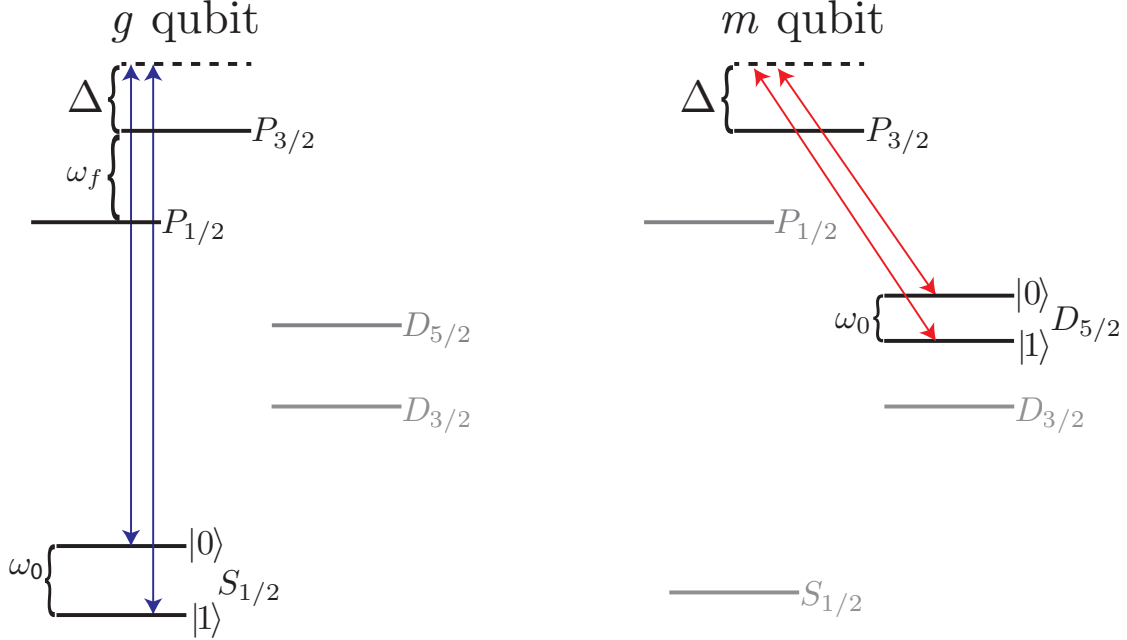


FIG. 1. The g qubits are encoded in hyperfine sublevels of the $S_{1/2}$ manifold with gates often performed via Raman transitions; m qubits are encoded in the hyperfine sublevels of the $D_{5/2}$ level with gates performed in the same way. Manifolds that do not participate in qubit operations in the respective schemes are shown in gray.

II. CHOICE OF QUBIT

In what follows, we consider a number of common trapped-ion qubit species: ${}^9\text{Be}^+$, ${}^{25}\text{Mg}^+$, ${}^{43}\text{Ca}^+$, ${}^{87}\text{Sr}^+$, ${}^{133}\text{Ba}^+$, ${}^{135}\text{Ba}^+$, ${}^{137}\text{Ba}^+$, ${}^{171}\text{Yb}^+$, and ${}^{173}\text{Yb}^+$. We present results for g qubits in all species. For m qubits, we perform calculations only for the species with a sufficiently long-lived ($\gtrsim 1$ second lifetime) $D_{5/2}$ manifold: ${}^{43}\text{Ca}^+$, ${}^{87}\text{Sr}^+$, ${}^{133}\text{Ba}^+$, ${}^{135}\text{Ba}^+$, and ${}^{137}\text{Ba}^+$ (see [4] for further discussion). The attributes of all these ion species are given in Table II.

For our qubit choices, we use hyperfine ‘clock’ qubits for both m and g qubits, due to their insensitivity to magnetic field noise and corresponding suppression of Rayleigh dephasing [5] (see [6] for the definition of ‘clock qubit’). We give our qubit choices (along with other qubit details) explicitly in Table II in terms of F (hyperfine angular momentum quantum number) and m_F (corresponding angular momentum projection quantum number). Since g qubits are already well-established, we simply follow [1] in their encoding scheme. As for m qubits, we choose states that should be relatively easy to prepare and readout, as well as give consistent behavior across all m qubit species for studying the scattering errors the states $|0\rangle = |D_{5/2}, F = I + \frac{5}{2}, m_F = \pm(I + \frac{5}{2} - 1)\rangle$ and

$|1\rangle = |D_{5/2}, F = I + \frac{5}{2} - 1, m_F = \pm(I + \frac{5}{2} - 1)\rangle$ (depending on the sign of relevant hyperfine splittings), where F is the hyperfine angular momentum quantum number, m_F is the corresponding angular momentum projection quantum number, and I is the nuclear spin (for further discussion of this choice, see [7]). Details about the qubits in the various species are given in Table II.

III. SCATTERING PROBABILITY

We assume that gates are performed on m and g qubits using stimulated Raman transitions (Fig. 1). These are coherent two-photon processes where population is transferred between $|0\rangle$ and $|1\rangle$ virtually through higher energy intermediate states $|k\rangle$. During this process, there is a chance of spontaneous photon scattering, changing the ion’s state or qubit phase and causing an error. The scattering probability is therefore important for characterizing the fidelity achievable by logic gates, as it sets an upper bound.

The rate of ‘ Λ and V scattering’ (the processes with upward pointing laser beam arrows in Fig. 2) from qubit states $|i\rangle$ to final state $|f\rangle$ can be calculated from (see Appendix A for derivation)

	⁹ Be ⁺	²⁵ Mg ⁺	⁴³ Ca ⁺	⁸⁷ Sr ⁺	¹³³ Ba ⁺	¹³⁵ Ba ⁺	¹³⁷ Ba ⁺	¹⁷¹ Yb ⁺	¹⁷³ Yb ⁺
I	3/2	5/2	7/2	9/2	1/2	3/2	3/2	1/2	5/2
$\tau_{D_{5/2}}$ (s)	-	-	1.110	0.357	29.856			0.0072	
$ 0\rangle (m)$	-	-	$ 6, +5\rangle$	$ 6, -6\rangle$	$ 2, +2\rangle$	$ 4, -3\rangle$	$ 4, -3\rangle$	-	-
$ 1\rangle (m)$	-	-	$ 5, +5\rangle$	$ 7, -6\rangle$	$ 3, +2\rangle$	$ 3, -3\rangle$	$ 3, -3\rangle$	-	-
$ 0\rangle (g)$	$ 2, 0\rangle$	$ 3, 0\rangle$	$ 4, 0\rangle$	$ 5, 0\rangle$	$ 1, 0\rangle$	$ 2, 0\rangle$	$ 2, 0\rangle$	$ 0, 0\rangle$	$ 2, 0\rangle$
$ 1\rangle (g)$	$ 1, 0\rangle$	$ 2, 0\rangle$	$ 3, 0\rangle$	$ 4, 0\rangle$	$ 0, 0\rangle$	$ 1, 0\rangle$	$ 1, 0\rangle$	$ 1, 0\rangle$	$ 3, 0\rangle$
Clock point (m, G)	-	-	2.54	6.49	33.0	1.79	0.0720	-	-
$\omega_0/2\pi (m, \text{GHz})$	-	-	0.025	0.036	0.062	0.012	0.00047	-	-
$\omega_0/2\pi (g, \text{GHz})$	1.3	1.8	3.2	5.0	9.9	7.2	8.0	12.6	10.5
$d^2(\omega_0/2\pi)/dB^2 (m, \text{kHz/G}^2)$	-	-	55.9	36.7	10.6	119	1.72	-	-
$d^2(\omega_0/2\pi)/dB^2 (g, \text{kHz/G}^2)$	3.13	2.19	1.21	0.783	0.395	0.545	0.487	0.309	0.373
$\gamma_{P_{3/2}}/2\pi (\text{MHz})$	19.4	41.8	23.2	24.0	25.2			25.9	
$\alpha_{S_{1/2}}$	1	1	0.9350	0.9406	0.7417			0.9875	
$\alpha_{D_{3/2}}$	-	-	0.0063	0.0063	0.02803			0.0017	
$\alpha_{D_{5/2}}$	-	-	0.0587	0.0531	0.2303			0.0108	
$\omega_f/2\pi (\text{THz})$	0.198	2.75	6.68	24.0	57.2			99.8	

TABLE I. Characteristics of the qubits and ion species we consider. Throughout the table, m and g denote values for m or g qubits. I is the nuclear spin; $\tau_{D_{5/2}}$ is the $D_{5/2}$ lifetime; $|0\rangle$ and $|1\rangle$ are the qubit states in the notation $|F, m_F\rangle$; ω_0 is the qubit frequency; $d^2\omega_0/dB^2$ is the second-order B -field-dependence of the qubit frequency; $\gamma_{P_{3/2}}$ is the decay rate of the $P_{3/2}$ manifold; α_M is the branching ratio of $P_{3/2}$ to manifold M ; and $\omega_f/2\pi$ is the fine-structure separation of the P manifolds. Lifetimes taken from [8, 9]; $\gamma_{P_{3/2}}$ values were taken/calculated from [10–15]; α_M values taken from [16–19]; ω_f values taken from [1]; other values calculated from atomic parameters.

$$\Gamma_{f,\text{AV}} = \sum_{i,j,q} \frac{e^2 E_j^2 \mu_{P_i}^2}{4\hbar^2} \xi_i \gamma_{P_f} \left| \sum_k \left(\frac{\langle f|\vec{r}\cdot\hat{\mathbf{e}}_q|k\rangle\langle k|\vec{r}\cdot\hat{\mathbf{e}}_j|i\rangle}{\mu_{P_f}\mu_{P_i}(\omega_{kP}-\Delta)} + \frac{\langle f|\vec{r}\cdot\hat{\mathbf{e}}_j|k\rangle\langle k|\vec{r}\cdot\hat{\mathbf{e}}_q|i\rangle}{\mu_{P_f}\mu_{P_i}(\omega_{ki}+\omega_{kf}+\Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{P_f}} \right)^3, \quad (1)$$

where ξ_i is the occupation probability of qubit state $|i\rangle$ (on average $\frac{1}{2}$ for each qubit state in the gates we consider), \vec{r} is the position operator of the electron relative to the atomic core (note that the matrix elements of the electric dipole operator can be tricky to calculate; see [20] for a simple introduction to computing them while avoiding pitfalls), γ_{P_f} is the decay rate from $P_{3/2}$ to the manifold containing f , the various ω_{nm} are transition frequencies with n and m corresponding to states or levels (see [21] for more thorough definition of ω_{nm}), Δ is the detuning measured from the $P_{3/2}$ manifold (positive

for detuning above this manifold, and neglecting the energy spread of the manifold's sublevels), E_j and $\hat{\mathbf{e}}_j$ are the electric field and polarization direction of beam j respectively, and $\hat{\mathbf{e}}_q$ is the scattered photon polarization. The parameters μ_{P_i} (corresponding to transitions between the $P_{3/2}$ manifold and the manifold containing state i) and μ_{P_f} (corresponding to transitions between the manifolds containing k and f) are the transition dipole matrix elements of the spin-orbital coupling. Their general form can be derived by invoking the Wigner-Eckart theorem [22], giving

$$\mu_{ul} = \left| \langle L_l|\vec{r}|L_u\rangle \sqrt{(2J_l+1)(2L_l+1)} \left\{ \begin{matrix} L_l & L_u & 1 \\ J_u & J_l & S \end{matrix} \right\} \right|, \quad (2)$$

where $\langle L_u|\vec{r}|L_l\rangle$ is the reduced dipole matrix element for transitions between upper level u and lower level l , S is the spin, L is the orbital angular momen-

tum, J is the total angular momentum ($L+S$), and the bracketed term is the Wigner 6j-symbol. The matrix element μ_{ul} can also be related to the de-

cay rate γ_{ul} from the J_u, L_u level to the J_l, L_l level by [23]

$$\gamma_{ul} = \frac{e^2 \omega_{ul}^3}{3\pi \epsilon_0 \hbar c^3} \mu_{ul}^2, \quad (3)$$

where ω_{ul} is the transition frequency, and e is

the charge of the electron. In m qubits (but not g qubits), ‘ladder scattering’ (the processes with downward pointing laser beam arrows in Fig. 2) can also contribute to the error. The ladder scattering rate is given by

$$\Gamma_{f,\text{lad}} = \frac{e^2 E_j^2 \mu_{P_i}^2}{4\hbar^2} \sum_{i,j,q} \xi_i \gamma_{P_f} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_j^* | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{kP} + \Delta + 2\omega_{PD})} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_j^* | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{kP} - \Delta + \omega_{Df})} \right) \right|^2 \left(1 - \frac{2\omega_{PD} + \Delta}{\omega_{P_f}} \right)^3. \quad (4)$$

Note that in Eqn. 1, the Λ scattering term (first term in the sum within the modulus) dominates over V scattering; similarly, in Eqn. 4, the first term in the modulus dominates. In both equations, these terms correspond to a two-photon scattering process where the laser photon is first absorbed or emitted, while the other weaker terms (what we call the counter-rotating contributions to the scattering rate) correspond to processes where the scattered photon is emitted first (Fig. 2).

We can rewrite Eqns. 1 and 4 in terms of g_{ul} given in [1] as

$$g_{ul} = \frac{eE\mu_{ul}}{2\hbar}. \quad (5)$$

Note that in the following calculations, for m qubits, g_{P_i} and μ_{P_i} will correspond to transitions between the $D_{5/2}$ and $P_{3/2}$ manifolds, *i.e.*, u and l in Eqns. 2 and 5 will correspond to $P_{3/2}$ and $D_{5/2}$, respectively. For g qubits, g_{P_i} and μ_{P_i} will correspond to transitions between $S_{1/2}$ and $P_{3/2}$. Assuming that g_{P_i} is the same for both Raman beams, we can now write Eqns. 1 and 4 as

$$\Gamma_{f,\text{AV}} = g_{P_i}^2 \sum_{i,j,q} \xi_i \gamma_{P_f} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_j | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{kP} - \Delta)} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_j | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{ki} + \omega_{kf} + \Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{P_f}} \right)^3 \quad (6)$$

and

$$\Gamma_{f,\text{lad}} = g_{P_i}^2 \sum_{i,j,q} \xi_i \gamma_{P_f} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_j^* | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{kP} + \Delta + 2\omega_{PD})} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_j^* | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{kP} - \Delta + \omega_{Df})} \right) \right|^2 \left(1 - \frac{2\omega_{PD} + \Delta}{\omega_{P_f}} \right)^3, \quad (7)$$

where ω_{Df} is the frequency of the transition between $D_{5/2}$ and the manifold containing the state f . Eqn. 6 gives what we will call the ‘full model’ for g qubits, and the sum of Eqns. 6 and 7 gives the corresponding full model for m qubits. In m qubits, we include contributions to the gate error from some of the closest higher energy intermediate states, but we neglect

such contributions in g qubits for reasons given in section III C below.

We will also define what we call our ‘simplified model’, in which we neglect the detuning dependence of the scattered photon frequency (*i.e.*, we assume $(1 + \Delta/\omega_{P_f})^3 \approx 1$), neglect the contributions of the higher energy intermediate manifolds, and as-

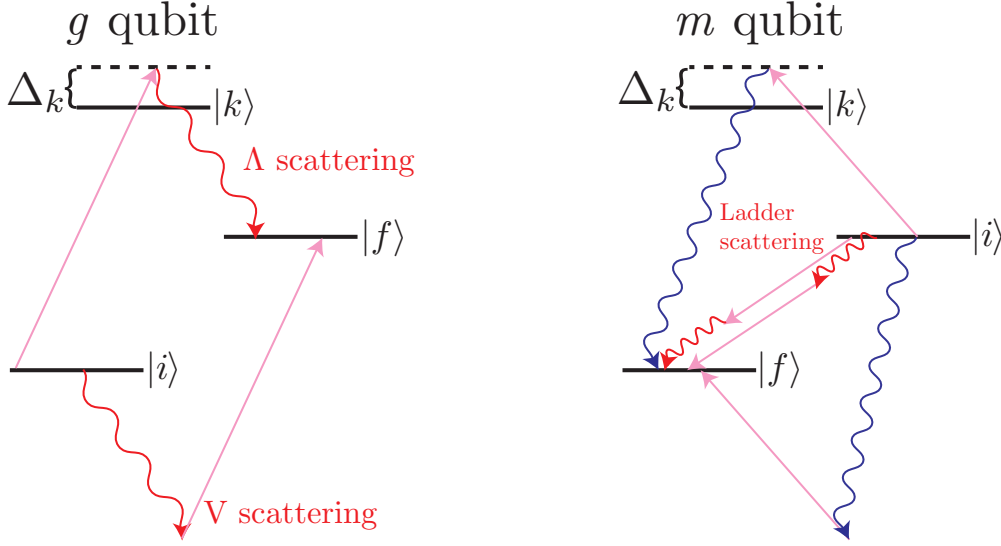


FIG. 2. All two-photon scattering pathways from state $|i\rangle$ to $|f\rangle$ via excited state $|k\rangle$ in g qubits and m qubits. Raman laser beam shown in pink, scattered light shown in red and blue. Processes in which the Raman beam points up in the diagram are Λ or V scattering events. Processes in which the Raman beam points down are ladder scattering events (note that ladder scattering events are allowed only in m qubits).

sume that only the first term in the squared modulus of Eqn. 6 appreciably contributes to the scattering rate in both m qubits and g qubits. This model results in a simpler version of Eqn. 6 [24],

$$\Gamma_f \approx g_{P_i}^2 \sum_{i,j,q} \xi_i \gamma_{P_f} \left| \sum_k \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_j | i \rangle}{\mu_{P_f} \mu_{P_i} (\omega_{kP} - \Delta)} \right|^2. \quad (8)$$

These assumptions have also been made in studies of g qubits (*e.g.*, [1]) where the 10^{-4} gate error threshold required detunings on the order of 10 THz. However, as we will show, even at these modest detunings, our model can give large corrections to this simplified model. Despite this, the simplified model in m qubits provides an intuitive illustration of the scattering behavior for a large range of detunings, so we therefore elect to present both models of m qubits throughout this paper.

A. Single-qubit gates

We follow [1] in their choice of a representative single-qubit gate: a π -rotation around the x -axis of the equivalent Bloch sphere, a $\hat{\sigma}_x$ gate. We will assume the gates are driven by two laser beams that induce two-photon stimulated Raman transitions. In the case of m qubits, we assume both beams are purely π -polarized because we found that they minimize the scattering probability and power requirements. In g qubits, we assume each beam has equal parts σ^+ and σ^- polarization. For this gate, the required time is given by [1]

$$\tau_{1q} = \frac{\pi}{2|\Omega_R|}, \quad (9)$$

where the Rabi frequency Ω_R is calculated according to

$$\Omega_R = g_{P_i}^2 \sum_k \left(\frac{\langle 1 | \vec{r} \cdot \hat{\mathbf{e}}_r^* | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_b | 0 \rangle}{\mu_{k_i}^2 (\omega_{kP} - \Delta)} + \frac{\langle 1 | \vec{r} \cdot \hat{\mathbf{e}}_r | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_b^* | 0 \rangle}{\mu_{k_i}^2 (\omega_{k_i} + \omega_{P_i} + \Delta)} \right), \quad (10)$$

where r and b (for red and blue) label the two Raman beams, $|0\rangle$ and $|1\rangle$ are the two qubit states and k indexes the available intermediate states. Since Raman scattering (scattering events for which

$|i\rangle \neq |f\rangle$) is the dominant source of errors for most species, we will, for the moment, neglect errors caused by Rayleigh scattering (scattering events for which $|i\rangle = |f\rangle$); Section III D below gives a discus-

sion of these errors. We can now calculate the general Raman scattering error as

$$P_{\text{Ram}} = \tau_{1q} \Gamma_{\text{Ram}} = \frac{\pi \Gamma_{\text{Ram}}}{2|\Omega_R|}, \quad (11)$$

where Γ_{Ram} is given by the sum of the scattering rates to all possible final states $|f\rangle$ for which $|f\rangle \neq |i\rangle$. For g qubits, this will be the sum of Eqn. 6 over all relevant final states; for m qubits, it will be the sum of both Eqns. 6 and 7 over all relevant final states. This scattering error is plotted for g and m qubits in Fig. 3.

In the simplified model of m qubits, we neglect intermediate manifolds aside from the lowest energy $P_{3/2}$ manifold, the detuning dependence of the scattered photon frequency, ladder decay, and the counter-rotating field contribution to the scattering rate and Rabi frequency (the second terms in the sum of Eqn. 6 and Eqn. 10). In this case, the sum over k has the same form for all ion species considered, giving an expression for the m qubit simplified model's Rabi frequency for each ion species the form

$$\Omega_R = -\frac{2}{15} \frac{g_{P_i}^2}{\Delta}. \quad (12)$$

and a Raman scattering rate of the form

$$\Gamma_{\text{Ram}} = \rho \frac{4}{15} \frac{g_{P_i}^2 \gamma}{\Delta^2} = 2\rho \left| \Omega_R \frac{\gamma}{\Delta} \right|, \quad (13)$$

where ρ is the Raman-only fraction of the total scattering rate (the value of ρ can be inferred from the nearly constant ratio in the low-detuning regime of Fig. 5) and γ is the decay rate of the $P_{3/2}$ manifold. For the m qubit's simplified model, this results in a gate error of

$$P_{\text{Ram}} = \rho \frac{\pi \gamma}{|\Delta|}. \quad (14)$$

This simplified model of m qubit scattering gives an especially simple form for P_{Ram} , since only one manifold ($P_{3/2}$) appreciably contributes to the scattering. The m qubit's simplified model behavior is also shown in Fig. 3 as the dashed lines.

From Fig. 3, we can see that the m qubits and g qubits each, as groups, have markedly similar behavior because their $P_{3/2}$ decay rates are all within ~ 5 MHz (Table II) of each other. As expected, the full model results deviate from the initial linear regime as the detuning becomes large. In m qubits, the full model yields a lower scattering probability than the simplified model for red-detuning, and a higher scattering probability for blue-detuning. The deviations from the m simplified model can be observed in the lower plot of Fig. 3

B. Two-qubit gates

Consider now a two-qubit Mølmer-Sørensen gate, driven by three Raman beams [5, 25]. We will suppose that the three beams are comprised of one pair of co-propagating beams of power \mathcal{P} and one beam counter-propagating with this pair with intensity $2\mathcal{P}$, since this distribution of power minimizes the scattering error for this beam geometry. For this gate, the duration is

$$\tau_{2q} = \frac{\pi}{2\sqrt{2}|\Omega_R|} \frac{\sqrt{K}}{\eta}. \quad (15)$$

This is the gate time for the two-qubit gate in [1] but reduced by a factor of $1/\sqrt{2}$ because of the unequal distribution of intensities in this setup. In this equation, K is the number of loops the $|01\rangle$ and $|10\rangle$ states trace out in phase space (we set $K = 1$ for our calculations) and η is the Lamb-Dicke parameter, which for counter-propagating Raman beams is given [1] by

$$\eta = \Delta k z_0 b_p^{(i)} = 2k_L z_0 \frac{1}{\sqrt{2}} = \sqrt{2} \frac{\omega_L}{c} z_0, \quad (16)$$

where Δk is the magnitude of the difference between the two Raman beams' wavevectors (this difference wavevector being aligned to the mode of interest), k_L and ω_L are respectively the wavenumber and frequency of the Raman beams, and $b_p^{(i)}$ is the mode participation factor (equal to $1/\sqrt{2}$ here). Note that the simplified model of [1] considers perpendicular beams which would increase the Lamb-Dicke parameter by a factor of $\sqrt{2}$. The root-mean-square spatial spread of the ground state wavefunction z_0 is given by

$$z_0 = \sqrt{\hbar/2M\omega_{\text{trap}}}, \quad (17)$$

where M is the mass of each ion and ω_{trap} is the frequency of the driven motional mode.

The scattering probability for this gate is the single-qubit gate scattering probability scaled by a factor of 4, as well as the extra gate time factor [1]. The factor of 4 comes from two considerations: first, that the two-qubit gate uses three beams with a total power twice as great as in the total beam power used in the single-qubit gate, and second that there are two ions. Both of these differences scales the gate error by a factor of 2 to generate an overall increase of 4. This gives a general form for the two-qubit gate Raman scattering error,

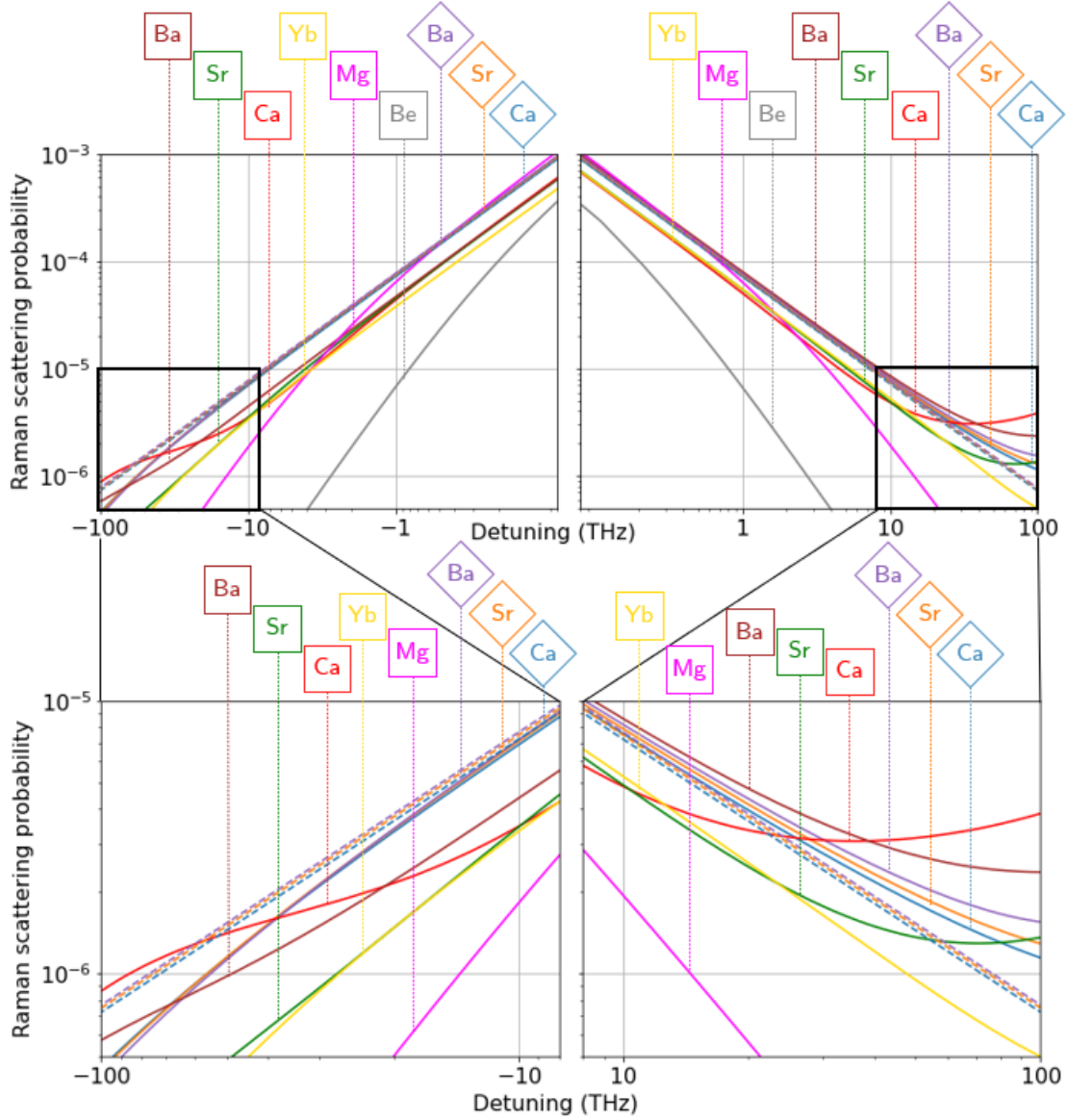


FIG. 3. Raman scattering probability for m qubits and g qubits during single-qubit $\hat{\sigma}_x$ gate for large detuning. The m qubit ions are labeled by diamonds and the g qubit ions are labeled by squares. Detuning is measured relative to $P_{3/2}$ in m qubits; for g qubits, red detuning is measured relative to $P_{1/2}$, and blue detuning is measured relative to $P_{3/2}$. The lower plot shows zoomed-in regions of the upper plot with the simplified model behavior shown for m qubits as dashed lines. Note that the left side of the figure corresponds to the laser frequency approaching zero.

$$P_{R2q} = \frac{\pi\Gamma_{\text{Ram}}}{2\sqrt{2}|\Omega_R|} \frac{4\sqrt{K}}{\eta}. \quad (18)$$

For the simplified model in m qubits, this results in

$$P_{R2q,simp} = \rho \frac{\pi\gamma}{|\Delta|} \frac{4\sqrt{K}}{\sqrt{2}\eta}. \quad (19)$$

Because η is proportional to the laser frequency, it

will in general depend on detuning. For the simplified model, we neglect this detuning dependence, so the error probability again exhibits linear behavior. We do, however, include this dependence in the full model, as the size of the effect is too large to reasonably ignore. In the full model for both g and m qubits, we can see that the Lamb-Dicke parameter leads to a further detuning dependence of the form

$$P_{R2q} \propto \frac{\omega_{P_i}}{|\Delta(\omega_{P_i} + \Delta)|}, \quad (20)$$

where ω_{P_i} denotes the frequency of the transition between the $P_{3/2}$ manifold and the manifold containing the qubit states. The Raman scattering error of a 1-loop two-qubit Mølmer-Sørensen gate is plotted in Fig. 4; the full models of g and m qubits are shown by the solid curves, and the simplified m qubit model is shown by the dashed curves.

The qualitative behavior of the sharply-increasing red-detuned m qubit scattering probability in Fig. 4 can be understood by observing that as the laser frequency approaches zero, the gate time goes to infinity (because the Lamb-Dicke parameter is approaching zero); however, the frequency of photons scattered to, *e.g.*, $S_{1/2}$ approaches a non-zero value (the $S_{1/2} \leftrightarrow D_{5/2}$ transition frequency). The gate error, being the product of the scattering rate and gate time, therefore approaches infinity as the laser frequency goes to zero. This does not occur in g qubits, because the scattered photon frequency also goes to zero as the laser frequency approaches zero. Additionally, the scattering rate in g qubits is generally lower than in m qubits across the detuning range considered. However, scattering errors in m qubits are largely due to scattering to $S_{1/2}$, *i.e.*, outside the qubit manifold. Such errors are easier to detect and correct; indeed, one of us (WCC) recently applied some of the results presented here in an investigation of erasure conversion in quantum error correction with metastable states [26].

The form of the g scattering probability plotted in Figs. 3 and 4 differs in several ways from that given in [1]. First, we compute D -manifold scattering rates directly, whereas [1] calculates them by multiplying the total scattering rate by the branching ratio to the D -manifolds. This overestimates the D -scattering rate, as it neglects interference between the $P_{1/2}$ and $P_{3/2}$ manifolds. Second, we include the V scattering process and the counter-rotating electric field component contribution to the Rabi frequency (*i.e.*, the contribution to Rabi flopping that is neglected in the rotating-wave approximation). Third, we include the detuning dependence of the Lamb-Dicke parameter and scattered photon frequency (while [1] used a single value of η and ω_{sc} for the entire detuning range). This latter effect makes the scattering rate proportional to ω_{sc}^3 , the cube of the scattered photon frequency (Appendix A). Note that if a detuning is chosen such that $\Delta < -\omega_{P_f}$ the contribution to the scattering probability due to scattering to level f becomes precisely zero. This is because the proportionality to ω_{sc}^3 in such a condition renders the f scattering rate negative, which is

non-physical. Another way to see that such scattering is not permitted is to note that it violates energy conservation.

Finally, it is notable that by neglecting the above effects, the simplified model of [1] implied a lower bound on Raman scattering-induced gate error in g qubits (1.06×10^{-4} , 0.50×10^{-4} , 1.46×10^{-4} , and 0.007×10^{-4} for Ca^+ , Sr^+ , Ba^+ , and Yb^+ respectively). This bound appears because the D -manifold scattering error in that model approaches a non-zero value at large red detunings. However, in our model, this lower bound does not exist. The Raman scattering error approaches zero at large red detunings. This is as it should be, due to considerations of energy conservation.

C. Higher levels

The above models have incorporated scattering probability contributions from higher levels in m qubits, but not g qubits (a disparity we will justify in this section). Higher levels contribute to the scattering probability in two ways: through increasing the overall scattering rate, and increasing or decreasing the Rabi frequency. Because the Rabi frequency and scattering rate have different scaling in detuning, the inclusion of higher levels can actually result in a net decrease in scattering probability for some parameter regimes.

There are two factors which clearly attenuate the magnitude of contributions from the higher levels to the Raman scattering error: the larger frequency denominators of the scattering rates and the smaller radial overlap with higher levels' wavefunctions. To numerically estimate the contribution of the higher levels, we calculated it for the $P_{3/2}$, $F_{5/2}$, and $F_{7/2}$ manifolds available in the University of Delaware database [27] in Ca^+ , Sr^+ , and Ba^+ .

The contributions from higher levels differ substantially in m qubits and g qubits. Destructive interference in the Rabi frequency due to higher levels causes m qubit behavior to change noticeably at large red- and blue-detunings. Interference effects are largely absent in g qubits, however, since the contributions of higher $P_{1/2}$ and $P_{3/2}$ essentially cancel. The reason for this cancellation is that Raman scattering flips the electron spin via the spin-orbit coupling $\mathbf{L} \cdot \mathbf{S}$ in the excited state. However, the dipole matrix element for this process is equal in magnitude but opposite in sign for the two fine-structure manifolds $P_{1/2}$ and $P_{3/2}$ [1, 28]. Since we are considering detunings far from the resonant excitation of the higher levels, these matrix elements largely cancel out their contributions; deviations from the g qubit model neglecting higher levels do not exceed

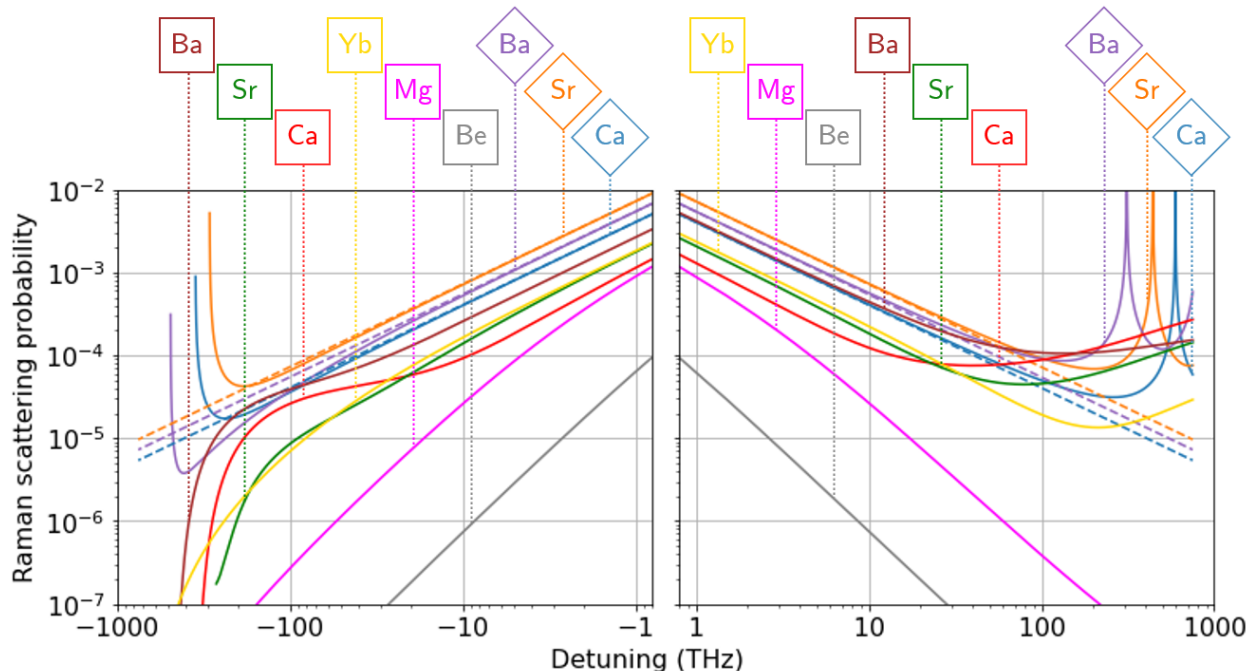


FIG. 4. Raman scattering probability from a 1-loop two-qubit Mølmer-Sørensen gate on m and g qubits. The m qubit ions are labeled by diamonds and the g qubit ions are labeled by squares. The simple model for m qubits is shown in the plot as dashed lines. Detuning is measured relative to $P_{3/2}$ in m qubits; for g qubits, red detuning is measured relative to $P_{1/2}$, and blue detuning is measured relative to $P_{3/2}$. Note that the left side of the figure corresponds to the laser frequency approaching zero.

more than a few percent until about 1 PHz detuning, where the laser frequency reaches a resonance with a higher P manifold.

Including the higher levels in the m qubit model lowers scattering probability at the 10^{-4} error level by a small amount for red detunings. The corrections for blue detuning, however, can be much larger at the 10^{-4} error level; inclusion of the higher levels *increases* the gate error for blue detuning. These corrections are large enough that when higher levels are included, Sr^+ and Ba^+ can no longer get below the 10^{-4} error level for blue detuning.

D. Rayleigh scattering errors

So far, we have neglected Rayleigh scattering-induced errors. Previous discussions of scattering in the literature (*e.g.* [1]) have characterized the infidelity contribution of Rayleigh scattering-induced dephasing as being proportional to the difference in elastic scattering rates; however, as is shown in [29], the dephasing rate due to Rayleigh scattering must be computed by including interference between the Rayleigh scattering amplitudes. This can lead to Rayleigh scattering becoming the dominant source

of error for certain parameter regimes. However, since we are considering clock qubits in both m qubits and g qubits, the Rayleigh-scattering-induced decoherence will be negligible. [5]

Rayleigh scattering can however cause errors during two-qubit gates in two other ways: recoil from the momentum kick during Rayleigh scattering and nonlinearities in the two-qubit gate. Both effects were studied in [1] and recoil was found to yield a gate error of the form

$$\epsilon_{\text{Ray}} = P_{E2q} \frac{\langle |\beta|^2 \rangle}{2K}, \quad (21)$$

where P_{E2q} is the probability of an elastic Rayleigh scattering event during the two-qubit gate, β the recoil displacement in phase space due to the scattering event, and K is the number of loops traced out in phase space (again, $K = 1$ for our gate). The expected value of $|\beta|^2$ depends on the polarization choice, but we can determine an upper bound on the reduction in fidelity by taking the recoil displacement to equal its maximum value. For our choice of laser beam geometry, the squared magnitude of the recoil displacement is given by [1]

$$|\beta|^2 = \frac{\eta^2}{2} \left(\frac{1}{\sqrt{2}} + \cos\theta \right)^2, \quad (22)$$

where η is the Lamb-Dicke parameter and θ is the angle of the recoil direction from the axis along the motional mode to which we are coupling. The maximum value is at $\theta = 0$, so

$$\langle |\beta|^2 \rangle \leq \eta^2 \left(\frac{3}{4} + \frac{1}{\sqrt{2}} \right) \quad (23)$$

implying

$$\epsilon_{Ray} \leq P_{E2q} \eta^2 \left(\frac{3}{8} + \frac{1}{2\sqrt{2}} \right). \quad (24)$$

The ratio of this upper bound on Rayleigh recoil error to the Raman scattering error during a two-qubit gate is plotted in Fig. 5 for m qubits and g qubits. The two lightest species, Be^+ and Mg^+ , have the largest Rayleigh scattering errors precisely because they are so light; their low masses increase their sensitivity to photon recoil. Additionally, they have higher frequency Raman transitions which makes their Lamb-Dicke parameters larger and therefore further increases their sensitivity to Rayleigh scattering events. In the other species, for most of the detuning range, the Rayleigh recoil errors are small. For m qubits, the magnitude of this contribution to the error is 5 or 6 orders of magnitude lower at the 10^{-4} error level compared to Raman scattering contribution in the various species of ions we consider. The g Rayleigh recoil error is roughly 1 to 3 orders of magnitude smaller than the Raman scattering error for most of the detuning range shown (again, except for Be^+ and Mg^+). The m qubits fare better than g qubits with respect to the Rayleigh recoil error in large part because m qubits have lower branching ratios to the qubit manifold, which leads to a

lower probability of elastic scattering events. Furthermore, due to their lower frequency transitions, they have lower Lamb-Dicke parameters, which further suppresses the Rayleigh recoil error.

The infidelity contribution of gate nonlinearities due to recoil momentum displacement is even smaller. As noted in [1], the error due to such nonlinearities is proportional to η^4 . This is negligible for most g qubits and even less important in m qubits, due to their smaller Lamb-Dicke parameters. In Be^+ and Mg^+ , this error can still be larger than the Raman scattering error, but it is small compared to the Rayleigh recoil error.

IV. POWER REQUIREMENTS

Now we will calculate the power required to achieve a given gate error rate (recall that we are considering a two beam single-qubit gate and a three-beam two-qubit gate). We first rewrite Eqn. 3,

$$\rho_q \frac{\gamma}{g_{P_i}^2} = \frac{4\hbar\omega_{3/2}^3}{3\pi\epsilon_0 c^3 E^2}, \quad (25)$$

where ρ_q is the inelastic fraction of the scattering from $P_{3/2}$ to the qubit manifold, ω_{P_i} is the frequency of the transition from the qubit manifold to $P_{3/2}$, and E is the peak electric field strength. Note that this equation differs from Eqn. 15 of [1] only by the factor ρ_q . This is because g_{P_i} in [1] is defined for $S_{1/2} \leftrightarrow P_{3/2}$ transitions, and the branching ratio from $P_{3/2}$ to the lower D levels was considered small enough that the authors treated the decay rate to $S_{1/2}$ as the total decay rate. In m qubits, g_{P_i} is defined for transitions from $D_{5/2} \leftrightarrow P_{3/2}$, but ρ_q is too small to treat this decay rate as the total. The decay rate to an individual level must then be the total decay rate weighted by the branching ratio to that level.

By rewriting $g_{P_i}^2$ in Eqn. 25 in terms of Ω_R and its detuning dependence $r(\Delta)$, defined as

$$r(\Delta) = |\Omega_R(\Delta)|/g_{P_i}^2 = \left| \sum_k \left(\frac{\langle 1|\vec{r} \cdot \hat{\epsilon}_r^*|k\rangle \langle k|\vec{r} \cdot \hat{\epsilon}_b|0\rangle}{\mu_{ki}^2(\omega_{kP} - \Delta)} + \frac{\langle 1|\vec{r} \cdot \hat{\epsilon}_r|k\rangle \langle k|\vec{r} \cdot \hat{\epsilon}_b^*|0\rangle}{\mu_{ki}^2(\omega_{ki} + \omega_{P_i} + \Delta)} \right) \right|, \quad (26)$$

we can write the power requirement as a function of detuning for single and two-qubit gates as (see Appendix B for derivation)

$$\mathcal{P}_{1q}(\Delta) = \frac{\hbar\omega_{P_i}^3 w_0^2}{3c^2 \rho_q \gamma} \frac{\pi}{\tau_{1q} r(\Delta)} \quad (27)$$

and

$$\mathcal{P}_{2q}(\Delta) = \frac{2\hbar\omega_{P_i}^3 w_0^2}{3\sqrt{2}c^2 \rho_q \gamma} \frac{\pi\sqrt{K}}{\tau_{2q} \eta(\Delta) r(\Delta)}, \quad (28)$$

where w_0 is the beam waist and $\eta(\Delta)$ is the Lamb-Dicke parameter (including detuning dependence). Because the gate error is also some function of Δ ,

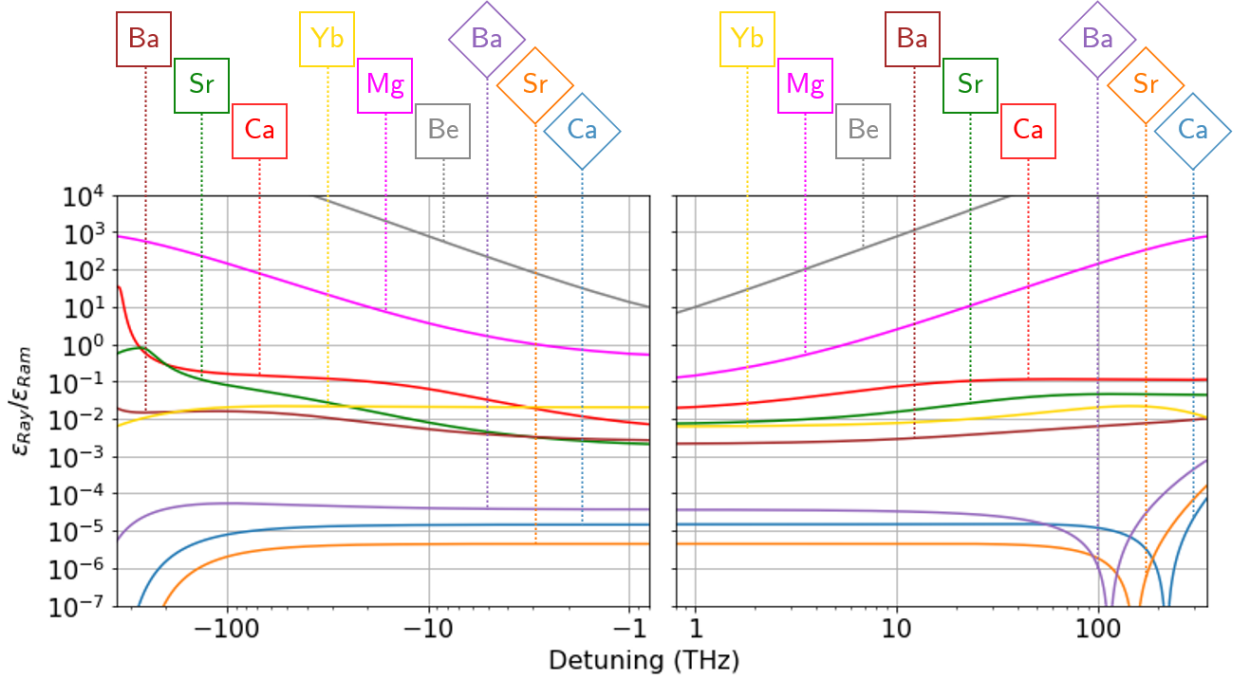


FIG. 5. Ratio of the upper bound on Rayleigh recoil error to Raman scattering error during a two-qubit gate for m qubits and g qubits. The m qubit ions are labeled by diamonds and the g qubit ions are labeled by squares. Note that, for the detunings plotted, the Rayleigh recoil error upper bound exceeds the Raman scattering error only for g qubits in Be^+ , Mg^+ , and Ca^+ . The steep declines in the ratio in m qubits at large blue detunings is due to destructive interference with the $F_{7/2}$ manifolds in the Rayleigh scattering rate.

$\epsilon(\Delta)$ (Sections III A and III B), we can visualize the power and error requirements by plotting the set of points $(\epsilon(\Delta), \mathcal{P}(\Delta))$, shown in Figs. 6 and 7.

For the simplified model, $r(\Delta)$ can be directly related to the gate error ϵ , allowing us to write the power as a function of error as (Appendix B)

$$\mathcal{P}_{1q}(\epsilon_{1q}) = \rho \frac{5\pi\hbar\omega_{P_i}^3 w_0^2}{2c^2\epsilon_{1q}} \frac{\pi}{\tau_{1q}} \quad (29)$$

and

$$\mathcal{P}_{2q}(\epsilon_{2q}) = \rho \frac{10\pi\hbar\omega_{P_i}^3 w_0^2}{c^2\epsilon_{2q}} \frac{\pi}{\tau_{2q}} \frac{K}{\eta^2}, \quad (30)$$

for single and two-qubit gates, respectively.

As can be seen in Figs. 6 and 7, the power requirements for g qubits are, for a large range of detunings, about one order of magnitude smaller than the required power for m qubits at the same error probability; this is mostly because of the small branching ratio to $D_{5/2}$ qubit manifold. However, we note that high power lasers are readily available at the large red detunings anticipated for m qubits, so these higher power requirements may not be a limiting factor.

From Fig. 7, we can also see the discrepancies between the simplified model and the full model reappearing. The full model power curves bend backwards past the minima of $\epsilon(\Delta)$, entering a regime of diminishing returns where the gates both take longer and have higher errors. For a fixed gate time, this farther detuning from resonance means more power is required to drive the gate.

V. CONCLUSION

We constructed a model of two-photon scattering errors during stimulated Raman transitions in trapped-ion qubits which incorporates all two-photon scattering processes, as well as the detuning dependencies of all system parameters involved. We also estimated the contribution to the scattering error from higher energy levels in m qubits and computed an upper bound on Rayleigh scattering error in both m and g qubits, finding this latter error to be negligible in all but the lightest ion species for most of the detuning range considered. We found that including all the above effects produced non-negligible corrections to simpler models of the systems, in particular that the more complete model implies there

is no lower bound on Raman scattering-induced infidelity of g qubits as suggested by past models of such errors [1].

Additionally, the results of the calculations (summarized in Table II) show that, although m qubits have a lower bound on gate infidelity and the detunings and powers required for 10^{-4} error in m qubits are larger than in g qubits, low errors should still be experimentally achievable in m qubits because high power lasers are more readily available at the required wavelengths for the ion species considered. In sum, low Raman scattering errors are achievable in stimulated Raman-driven gates for both m and g trapped-ion qubits at sufficiently large red detunings.

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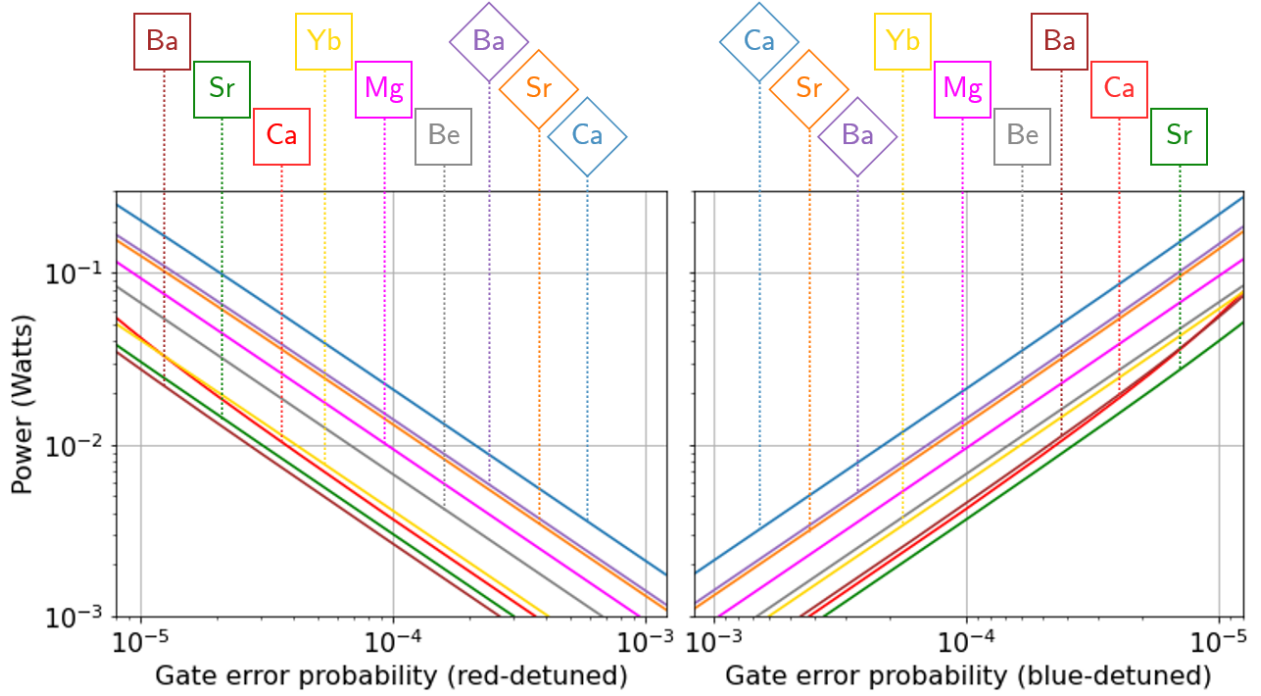


FIG. 6. Total power required to achieve a given error rate during a single-qubit π -rotation gate in the full model (assumes $20 \mu\text{m}$ beam waist and $1 \mu\text{s}$ gate time). The m qubit ions are labeled by diamonds and the g qubit ions are labeled by squares. Errors for red-detuned g qubits here are calculated from detunings below $P_{1/2}$ only.

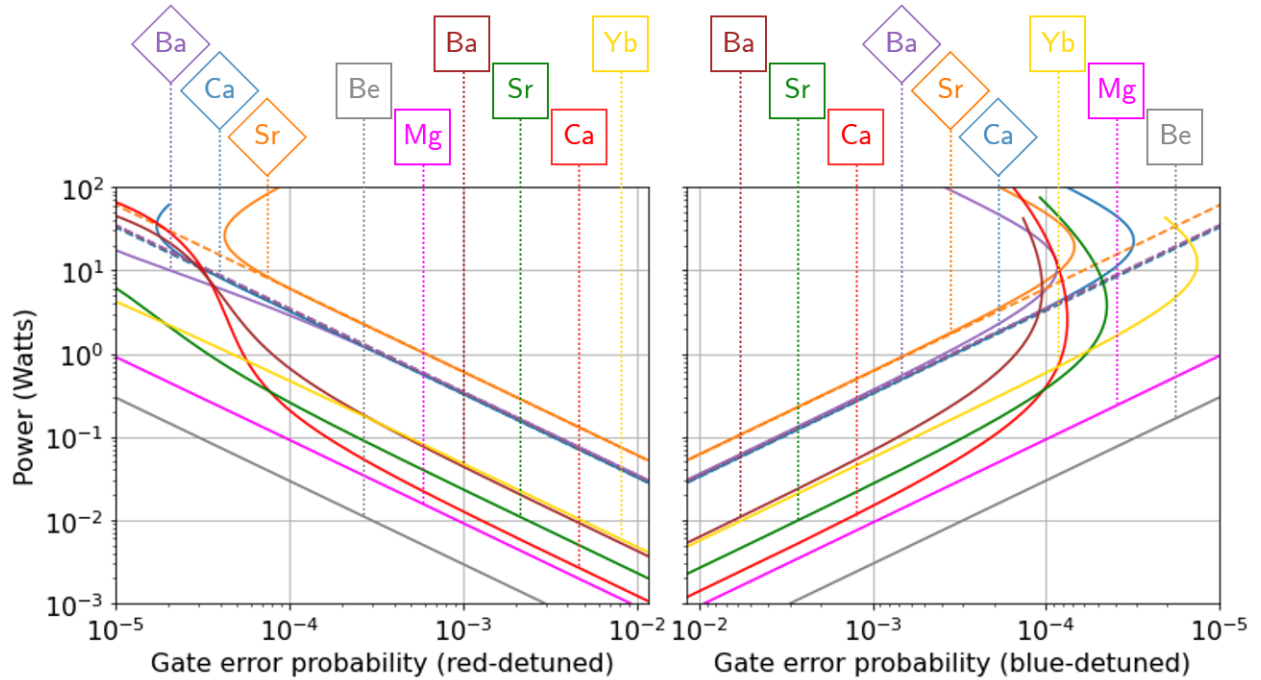


FIG. 7. Total power required to achieve a given error rate during a two-qubit Mølmer-Sørensen gate (assumes $20 \mu\text{m}$ beam waist, $10 \mu\text{s}$ gate time, and a 5MHz axial trap frequency). The m qubit ions are labeled by diamonds and the g qubit ions are labeled by squares. Solid lines correspond to full model, dashed lines correspond to the simplified model. Errors for red-detuned g qubits here are calculated from detunings below $P_{1/2}$ only.

		${}^9\text{Be}^+$	${}^{25}\text{Mg}^+$	${}^{43}\text{Ca}^+$	${}^{87}\text{Sr}^+$	${}^{133}\text{Ba}^+$	${}^{135}\text{Ba}^+$	${}^{137}\text{Ba}^+$	${}^{171}\text{Yb}^+$	${}^{173}\text{Yb}^+$
$\omega_0/2\pi$ (GHz)	m	-	-	0.025	0.036	0.062	0.012	0.00047	-	-
	g	1.3	1.8	3.2	5.0	9.9	7.2	8.0	12.6	10.5
η	m	-	-	0.036	0.021	0.028	0.028	0.028	-	-
	g	0.213	0.143	0.077	0.053	0.038	0.038	0.038	0.046	0.047
$\Delta/2\pi$ (THz)	m	-	-	-40.0 (963 nm)	-66.0 (1335 nm)	-45.3 (676 nm)	-45.6 (677 nm)	-45.9 (677 nm)	-	-
	g	-1.00 (313 nm)	-4.55 (281 nm)	-9.05 (402 nm)	-13.0 (429 nm)	-26.4 (515 nm)	-26.6 (516 nm)	-26.9 (516 nm)	-15.3 (376 nm)	-15.4 (376 nm)
Power (W)	m	-	-	4.9	9.1	4.4	4.4	4.5	-	-
	g	0.067	0.13	0.30	0.37	0.94	0.96	0.98	0.67	0.67

TABLE II. Comparison of g and m qubit gate characteristics. The qubit frequency for m and g qubits is given in the first two rows. The Lamb-Dicke parameter η is given for a 5 MHz trap frequency and counter-propagating beams at the $P_{3/2}$ resonances frequencies (resonance with $D_{5/2}$ and $S_{1/2}$ in m and g qubits, respectively). The detuning Δ corresponds to the detuning (from $P_{3/2}$ in m qubits and from $P_{1/2}$ in g qubits) required for 10^{-4} error, and the corresponding laser wavelength is given parenthetically below each detuning. Total power requirements are given for the 10^{-4} error threshold of the two-qubit Mølmer-Sørensen gate of Section III B, driven by 3 Raman beams.

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Appendices

Appendix A: Derivation of scattering rate

To derive the general Raman ΛV scattering formula (Eqn. 6), begin with Eqn. 8.7.3 of [30],

$$\Gamma_{i \rightarrow f, \Lambda V} = \sum_{\mathbf{k}_{sc}, \lambda} \frac{\pi e^4 \omega \omega_{sc} n}{2 \epsilon_0^2 \hbar^2 V^2} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\epsilon}_\lambda^* | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon} | i \rangle}{\omega_k - \omega} + \frac{\langle f | \vec{r} \cdot \hat{\epsilon} | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon}_\lambda^* | i \rangle}{\omega_k + \omega_{sc}} \right) \right|^2 \delta(\omega_{fi} + \omega_{sc} - \omega). \quad (\text{A1})$$

This equation describes the ΛV scattering rate to state f during virtual transitions from state i through the manifold containing the states indexed by k (where all these states are hyperfine sublevels). The transitions are driven by an n -photon laser beam of frequency ω and polarization $\hat{\epsilon}$, scattering a photon with frequency ω_{sc} and polarization $\hat{\epsilon}_\lambda^*$, where λ indexes the two independent polarizations in the chosen basis. The rate is calculated by summing contributions from all scattering modes \mathbf{k}_{sc} , λ allowed in the quantization volume V . The frequencies can be understood by the energy level diagram of Fig. 8.

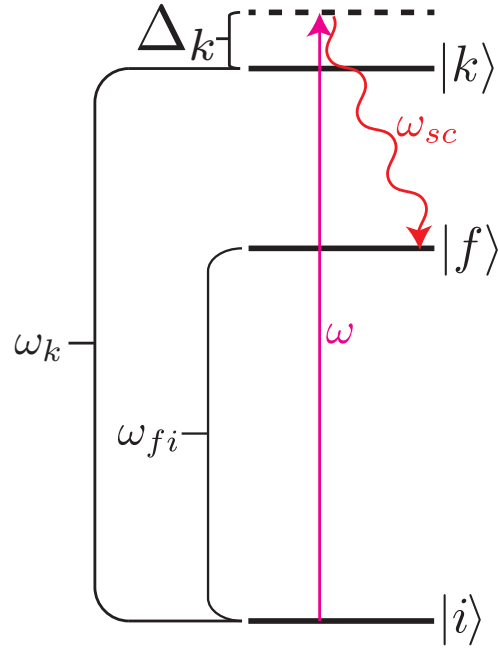


FIG. 8. Relevant frequency definitions for Eqn. A1. The transitions are driven by a laser beam of frequency ω . Scattered photon frequency is given by $\omega_{sc} = \omega - \omega_{fi}$.

Rearranging this equation, we find

$$\Gamma_{i \rightarrow f, \Lambda V} = \sum_{\mathbf{k}_{sc}, \lambda} \frac{\hbar \omega n}{\epsilon_0 V} \frac{\pi e^4 \omega_{sc}}{2 \epsilon_0 \hbar^3 V} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\epsilon}_\lambda^* | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon} | i \rangle}{\omega_k - \omega} + \frac{\langle f | \vec{r} \cdot \hat{\epsilon} | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon}_\lambda^* | i \rangle}{\omega_k + \omega_{sc}} \right) \right|^2 \delta(\omega_{fi} + \omega_{sc} - \omega). \quad (\text{A2})$$

Now, if we model the laser field as a classical electric field plane wave with amplitude E ,

$$\mathbf{E}_{\text{las}}(\mathbf{r}, t) = \frac{E}{2} \left(\hat{\epsilon} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + \hat{\epsilon}^* e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega t)} \right), \quad (\text{A3})$$

we can calculate the temporal and spatial average of

$$\langle \mathbf{E}_{\text{las}}^*(\mathbf{r}, t) \cdot \mathbf{E}_{\text{las}}(\mathbf{r}, t) \rangle = \frac{E^2}{2}. \quad (\text{A4})$$

Comparing this to the quantized result in an n photon Fock state, $\langle n | \hat{E}^\dagger \hat{E} | n \rangle = \frac{\hbar \omega}{\epsilon_0 V} (n + \frac{1}{2})$ allows us

to make the replacement, assuming $n \gg 1$,

$$\frac{2\hbar\omega}{\epsilon_0 V} n \approx E^2 \quad (\text{A5})$$

to get

$$\Gamma_{i \rightarrow f, \Lambda V} = \sum_{\mathbf{k}_{sc}, \lambda} \frac{E^2 \pi e^4 \omega_{sc}}{4 \epsilon_0 \hbar^3 V} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\epsilon}_\lambda^* | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon} | i \rangle}{\omega_k - \omega} + \frac{\langle f | \vec{r} \cdot \hat{\epsilon} | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon}_\lambda^* | i \rangle}{\omega_k + \omega_{sc}} \right) \right|^2 \delta(\omega_{fi} + \omega_{sc} - \omega). \quad (\text{A6})$$

Next, we can take the limit of large quantization

volume V ,

$$\sum_{\mathbf{k}_{sc}} \rightarrow \frac{V}{(2\pi)^3} \iint d\omega_{sc} d\Omega \frac{\omega_{sc}^2}{c^3}, \quad (\text{A7})$$

enabling us to write the spontaneous scattering rate in the form

$$\Gamma_{i \rightarrow f, \Lambda V} = \sum_\lambda \iint d\omega_{sc} d\Omega \frac{E^2 e^4 \omega_{sc}^3}{32 \pi^2 c^3 \epsilon_0 \hbar^3} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\epsilon}_\lambda^* | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon} | i \rangle}{\omega_k - \omega} + \frac{\langle f | \vec{r} \cdot \hat{\epsilon} | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon}_\lambda^* | i \rangle}{\omega_k + \omega_{sc}} \right) \right|^2 \delta(\omega_{fi} + \omega_{sc} - \omega). \quad (\text{A8})$$

Now we wish to perform the integral over $d\Omega \equiv d\phi d\theta \sin(\theta)$, the direction of the scattered photon's k -vector, \hat{k}_{sc} . For this, it is conceptually help-

ful to gather terms into an expression for the vector transition dipole matrix element for spontaneous scattering,

$$\vec{r}_{sc}(\omega_{sc}) \equiv \frac{Ee}{2\hbar} \sum_k \left(\frac{\langle f | \vec{r} | k \rangle \langle k | \vec{r} \cdot \hat{\epsilon} | i \rangle}{\omega_k - \omega} + \frac{\langle f | \vec{r} \cdot \hat{\epsilon} | k \rangle \langle k | \vec{r} | i \rangle}{\omega_k + \omega_{sc}} \right) \quad (\text{A9})$$

which allows us to write Eq. (A8) in the form

$$\Gamma_{i \rightarrow f, \Lambda V} = \sum_\lambda \iint d\omega_{sc} d\Omega \frac{e^2 \omega_{sc}^3}{8 \pi^2 c^3 \epsilon_0 \hbar} |\vec{r}_{sc} \cdot \hat{\epsilon}_\lambda^*|^2 \delta(\omega_{fi} + \omega_{sc} - \omega). \quad (\text{A10})$$

Without loss of generality, we will choose a polarization basis such that one of the basis polarization vectors lies in the plane of \vec{r}_{sc} and \hat{k}_{sc} ; we will call this vector $\hat{\epsilon}_{sc}$ (note that the other basis vector will not contribute to the scattering, as it will necessarily be perpendicular to \vec{r}_{sc}). Choosing θ to be the angle between \vec{r}_{sc} and \vec{k}_{sc} , the angle between \vec{r}_{sc} and $\hat{\epsilon}_{sc}$ is $\frac{\pi}{2} - \theta$, which implies

$$\begin{aligned} |\vec{r}_{sc} \cdot \hat{\epsilon}_{sc}|^2 &= |\vec{r}_{sc}|^2 \cos^2\left(\frac{\pi}{2} - \theta\right) \\ &= |\vec{r}_{sc}|^2 \sin^2(\theta), \end{aligned} \quad (\text{A11})$$

allowing us to carry out the integral over the direction of the spontaneously emitted photon to get

$$\begin{aligned}
\Gamma_{i \rightarrow f, \Delta V} &= \int d\omega_{sc} \frac{e^2 \omega_{sc}^3}{3\pi c^3 \epsilon_0 \hbar} |\vec{r}_{sc}(\omega_{sc})|^2 \delta(\omega_{fi} + \omega_{sc} - \omega) \\
&= \frac{e^2 (\omega - \omega_{fi})^3}{3\pi c^3 \epsilon_0 \hbar} |\vec{r}_{sc}(\omega - \omega_{fi})|^2 \\
&= \frac{e^2 (\omega - \omega_{fi})^3}{3\pi c^3 \epsilon_0 \hbar} (|\vec{r}_{sc}(\omega - \omega_{fi})|)^2 \\
&= \frac{e^2 (\omega - \omega_{fi})^3}{3\pi c^3 \epsilon_0 \hbar} \left(\sqrt{\sum_q |\vec{r}_{sc}(\omega - \omega_{fi}) \cdot \hat{\mathbf{e}}_q|^2} \right)^2 \\
&= \frac{e^2 (\omega - \omega_{fi})^3}{3\pi c^3 \epsilon_0 \hbar} \sum_q |\vec{r}_{sc}(\omega - \omega_{fi}) \cdot \hat{\mathbf{e}}_q|^2 \\
&= \frac{E^2 e^4 (\omega - \omega_{fi})^3}{12\pi c^3 \epsilon_0 \hbar^3} \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\omega_k - \omega} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\omega_k + \omega - \omega_{fi}} \right) \right|^2
\end{aligned} \tag{A12}$$

where $\hat{\mathbf{e}}_q$ is a polarization vector corresponding to π , σ^+ , or σ^- , with q indexing these possibilities.

Now, if $\Delta_k \equiv \omega - \omega_k$ is the detuning relative to some level k , we see that we can write $\omega - \omega_{fi}$ as $\omega_k - \omega_{fi} + \Delta_k$. Since we choose to measure the detuning relative to the $P_{3/2}$ manifold, we define $\Delta = \omega - \omega_{Pi}$, where ω_{Pi} is the frequency of the

transition between the manifold containing $|i\rangle$ and $P_{3/2}$. This allows us to rewrite $\omega - \omega_{fi}$ as $\omega_{Pf} + \Delta$, where ω_{Pf} is the frequency of the transition between the manifold containing state f and $P_{3/2}$; additionally, we may rewrite $\omega_k - \omega$ as $\omega_k - \omega_{Pi} - \Delta$. Rewriting $\omega - \omega_{fi}$ as such and rearranging gives us

$$\begin{aligned}
\Gamma_{i \rightarrow f, \Delta V} &= \frac{E^2 e^4 \omega_{Pf}^3}{12\pi c^3 \epsilon_0 \hbar^3} \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\omega_k - \omega_{Pi} - \Delta} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\omega_k + \omega_{Pf} + \Delta} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3 \\
&= \frac{e^4 \omega_{Pf}^3}{3\pi c^3 \epsilon_0 \hbar} \frac{E^2}{4\hbar^2} \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\omega_k - \omega_{Pi} - \Delta} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\omega_k + \omega_{Pf} + \Delta} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3.
\end{aligned} \tag{A13}$$

We begin the next step by noting Eqn. 3,

$$\frac{e^2 \omega_{Pf}^3}{3\pi \epsilon_0 \hbar c^3} \mu_{Pf}^2 = \frac{1}{\tau_{fl}} \equiv \gamma_{Pf}$$

where γ_{Pf} is the decay rate from the $P_{3/2}$ manifold

to the manifold containing state f ; this is given by $\alpha_f \gamma$, where α_f is the branching ratio into the manifold containing state f and γ is the total spontaneous decay rate for the manifold containing state l . With this, we can write

$$\begin{aligned}
\Gamma_{i \rightarrow f, \Delta V} &= \frac{\gamma_{Pf}}{\mu_{Pf}^2} \frac{e^2 E^2}{4\hbar^2} \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\omega_k - \omega_{Pi} - \Delta} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\omega_k + \omega_{Pf} + \Delta} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3 \\
&= \gamma_{Pf} \frac{e^2 E^2}{4\hbar^2} \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\mu_{Pf} (\omega_k - \omega_{Pi} - \Delta)} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{Pf} (\omega_k + \omega_{Pf} + \Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3.
\end{aligned} \tag{A14}$$

Now we divide and multiply by μ_{Pi} , *i.e.*, the ma-

trix element of Eqn. 2 between the $P_{3/2}$ and the

manifold containing i .

$$\begin{aligned}
\Gamma_{i \rightarrow f, \Delta V} &= \gamma_{Pf} \frac{e^2 E^2}{4\hbar^2} \sum_q \left| \sum_k \mu_{Pi} \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k - \omega_{Pi} - \Delta)} \right. \right. \\
&\quad \left. \left. + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k + \omega_{Pf} + \Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3 \\
&= \gamma_{Pf} \frac{e^2 E^2 \mu_{Pi}^2}{4\hbar^2} \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k - \omega_{Pi} - \Delta)} \right. \right. \\
&\quad \left. \left. + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k + \omega_{Pf} + \Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3. \tag{A15}
\end{aligned}$$

Noting the definition of g_{Pi} (Eqn. 5), this becomes

$$\Gamma_{i \rightarrow f, \Delta V} = \gamma_{Pf} g_{Pi}^2 \sum_q \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k - \omega_{Pi} - \Delta)} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k + \omega_{Pf} + \Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3. \tag{A16}$$

Finally, we make our result applicable to gates; we do so by averaging over the undisturbed state $|i\rangle$ during the course of the gate. If we consider a $\hat{\sigma}_x$ gate, then an ion initially in the state $|0\rangle$ will get mapped to $|1\rangle$. The ion's state $|i\rangle$ then has a

time dependence given by $|i(t)\rangle = \cos(2\pi t/\tau)|0\rangle + \sin(2\pi t/\tau)|1\rangle$, where τ is the gate time. Considering this time dependence and averaging Eqn. A16 over the gate time gives

$$\Gamma_{f, \Delta V} = \gamma_{Pf} \frac{g_{Pi}^2}{2} \sum_{i,q} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}} | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k - \omega_{Pi} - \Delta)} + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}} | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_k + \omega_{Pf} + \Delta)} \right) \right|^2 \left(1 + \frac{\Delta}{\omega_{Pf}} \right)^3, \tag{A17}$$

where $|i\rangle$ now indexes the two qubit states, $|0\rangle$ and $|1\rangle$.

By the same reasoning, we can get the ladder scattering rate (which contributes in m qubits but not g qubits),

$$\begin{aligned}
\Gamma_{f, \text{lad}} &= \gamma_{Pf} \frac{g_{Pi}^2}{2} \sum_{i,q} \left| \sum_k \left(\frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}_q | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}^* | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_{kP} + \Delta + 2\omega_{PD})} \right. \right. \\
&\quad \left. \left. + \frac{\langle f | \vec{r} \cdot \hat{\mathbf{e}}^* | k \rangle \langle k | \vec{r} \cdot \hat{\mathbf{e}}_q | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_{kP} - \Delta + \omega_{Df})} \right) \right|^2 \left(1 - \frac{2\omega_{PD} + \Delta}{\omega_{Pf}} \right)^3. \tag{A18}
\end{aligned}$$

To generate the full model scattering rate, we sum

over all scattering events $i \rightarrow f$ except for $i \rightarrow i$ scat-

tering events, since we are ignoring Rayleigh scattering.

We can get the simplified model's scattering rate equation by neglecting the V scattering term in Eqn. A17 and assuming $(1 + \Delta/\omega_{Pf})^3 \approx 1$

$$\Gamma_f \approx \gamma_{Pf} \frac{g_{Pi}^2}{2} \sum_i \left| \sum_{k,q} \frac{\langle f | \vec{r} \cdot \hat{e}_q | k \rangle \langle k | \vec{r} \cdot \hat{e} | i \rangle}{\mu_{Pf} \mu_{Pi} (\omega_{kP} - \Delta)} \right|^2. \quad (\text{A19})$$

We can again obtain the Raman scattering rate by summing over all scattering events except for $i \rightarrow i$.

Appendix B: Power requirements derivation

Here we derive the expressions given in Section IV. For a Gaussian laser beam of power \mathcal{P} we consider [31]:

$$E^2 = \frac{4\mathcal{P}}{\pi w_0^2 c \epsilon_0}, \quad (\text{B1})$$

$$\frac{\gamma}{g_{P_i}^2} = \frac{\hbar \omega_{P_i}^3 w_0^2}{3c^2 \mathcal{P} \alpha_q}, \quad (\text{B2})$$

where w_0 is the laser beam waist and α_q is the branching ratio for transitions between $P_{3/2}$ and the qubit manifold. Note that in general $|\Omega_R| = g_{P_i}^2 r(\Delta)$ for some $r(\Delta)$, so we can replace $g_{P_i}^2$ with $|\Omega_R|/r(\Delta)$. This means that the power can be written as

$$\mathcal{P} = \frac{\hbar \omega_{P_i}^3 w_0^2 |\Omega_R|}{3c^2 \alpha_q \gamma r(\Delta)}. \quad (\text{B3})$$

For a single-qubit gate, $|\Omega_R| = \pi/2\tau_{1q}$ where τ_{1q} is the gate time; for a two-qubit gate, $|\Omega_R| = \pi\sqrt{K}/2\sqrt{2}\tau_{2q}\eta(\Delta)$. This allows us to write the total power required for each as a function of Δ :

$$\mathcal{P}_{1q}(\Delta) = 2 \frac{\hbar \omega_{P_i}^3 w_0^2}{6c^2 \alpha_q \gamma} \frac{\pi}{\tau_{1q} r(\Delta)} \quad (\text{B4})$$

and

$$\mathcal{P}_{2q}(\Delta) = 4 \frac{\hbar \omega_{P_i}^3 w_0^2}{6\sqrt{2}c^2 \alpha_q \gamma} \frac{\pi\sqrt{K}}{\tau_{2q}\eta(\Delta)r(\Delta)}, \quad (\text{B5})$$

where the factor of 2 in front of Eqn. B4 is due to the use of two beams of equal power, and the factor of 4 in front of Eqn. B5 is due to the use of two beams of equal power along with one beam with double the power.

If, as we did in Section III, we neglect the effects of large detuning, we can write power as a function of the gate error directly. We will start by rewriting $r(\Delta) = 2/15|\Delta|$ in terms of the single-qubit gate error ϵ_{1q} ,

$$\epsilon_{1q} = \rho \frac{\pi\gamma}{|\Delta|} = \rho \frac{15\pi\gamma r(\Delta)}{2} \implies r(\Delta) = \frac{2\epsilon_{1q}}{15\rho\pi\gamma}, \quad (\text{B6})$$

or for a two-qubit gate,

$$\begin{aligned} \epsilon_{2q} &= \rho \frac{\pi\gamma}{|\Delta|} \frac{4\sqrt{K}}{\sqrt{2}\eta} = \rho \frac{15\pi\gamma r(\Delta)}{2} \frac{4\sqrt{K}}{\sqrt{2}\eta} \\ \implies r(\Delta) &= \frac{2\epsilon_{2q}}{15\rho\pi\gamma} \frac{\sqrt{2}\eta}{4\sqrt{K}}. \end{aligned} \quad (\text{B7})$$

Substituting into Eqns. B4 and B5, we get

$$\mathcal{P}_{1q}(\epsilon_{1q}) = \rho \frac{5\pi\hbar\omega_{P_i}^3 w_0^2}{2c^2 \epsilon_{1q} \alpha_q} \frac{\pi}{\tau_{1q}} \quad (\text{B8})$$

and

$$\mathcal{P}_{2q}(\epsilon_{2q}) = \rho \frac{10\pi\hbar\omega_{P_i}^3 w_0^2}{c^2 \epsilon_{2q} \alpha_q} \frac{\pi}{\tau_{2q}} \frac{K}{\eta^2}. \quad (\text{B9})$$

Appendix C: Recovering classical limits

We can test the completeness of our model by seeing if it is able to recover classical elastic scattering

$$\Gamma = E^2 \frac{e^4 \omega_L^3}{12\pi c^3 \epsilon_0 \hbar^3} \left| \sum_k |\langle i|\vec{r}|k\rangle|^2 \left(\frac{1}{\omega_{ki} - \omega_L} + \frac{1}{\omega_{ki} + \omega_L} \right) \right|^2. \quad (\text{C1})$$

For Raman beams with intensity I , we have $E^2 = 2I/\epsilon_0 c$; additionally, we can replace the scat-

tering rate in the limit of large detuning. Considering only elastic scattering excludes ladder scattering processes (since they are inelastic). This means we can write the elastic scattering rate as

$$\sigma = \alpha^2 \frac{8\pi}{3} \frac{\omega_L^4}{c^2} \left| \sum_k |\langle i|\vec{r}|k\rangle|^2 \left(\frac{1}{\omega_{ki} - \omega_L} + \frac{1}{\omega_{ki} + \omega_L} \right) \right|^2, \quad (\text{C2})$$

where $\alpha = e^2/4\pi\epsilon_0\hbar c$ is the fine structure constant.

Now we can calculate the limits. For large blue detuning ($\omega_L \gg \omega_{ki}$ for every k), we have

$$\left(\frac{1}{\omega_{ki} - \omega_L} + \frac{1}{\omega_{ki} + \omega_L} \right) \approx -2 \frac{\omega_{ki}}{\omega_L^2}, \quad (\text{C3})$$

allowing us to rewrite Eqn. C2 as

$$\sigma_{\text{blue}} = \alpha^2 \frac{32\pi}{3} \frac{1}{c^2} \left| \sum_k \omega_{ki} |\langle i|\vec{r}|k\rangle|^2 \right|^2. \quad (\text{C4})$$

Using the Thomas-Reiche-Kuhn sum rule (for the single valence electron only, and neglecting recoil and the ion's monopole charge), the sum can be evaluated to

$$\sum_k \omega_{ki} |\langle i|\vec{r}|k\rangle|^2 = \frac{\hbar}{2m_e}. \quad (\text{C5})$$

This gives

$$\sigma_{\text{blue}} = \alpha^2 \frac{32\pi}{3} \frac{1}{c^2} \frac{\hbar^2}{4m_e^2} = \frac{8\pi}{3} \alpha^4 a_0^2, \quad (\text{C6})$$

where a_0 is the Bohr radius and $\alpha^2 a_0$ is the classical electron radius. So for large blue detuning, the

model recovers the Thomson cross-section of the valence electron.

For red-detuning, ($\omega_L \ll \omega_{ki}$ for every k), we have

$$\left(\frac{1}{\omega_{ki} - \omega_L} + \frac{1}{\omega_{ki} + \omega_L} \right) \approx \frac{2}{\omega_{ki}}, \quad (\text{C7})$$

giving

$$\sigma_{\text{red}} = \alpha^2 \frac{8\pi}{3} \frac{\omega_L^4}{c^2} \left| \sum_k \frac{2|\langle i|\vec{r}|k\rangle|^2}{\omega_{ki}} \right|^2. \quad (\text{C8})$$

The DC polarizability of the ion can be written as

$$\alpha^{(0)} \equiv e^2 \sum_{k \neq i} \frac{\langle i|\vec{r}|k\rangle \langle k|\vec{r}|i\rangle + \langle k|\vec{r}|i\rangle \langle i|\vec{r}|k\rangle}{E_k - E_i}, \quad (\text{C9})$$

allowing us to rewrite Eqn. (C8) as

$$\begin{aligned} \sigma_{\text{red}} &= \frac{8\pi}{3} \alpha^2 \frac{\omega_L^4}{c^2 e^4} \hbar^2 |\alpha^{(0)}|^2 \\ &= \frac{8\pi}{3} \left(\frac{\omega_L}{c} \right)^4 \hbar^2 \left| \frac{\alpha^{(0)}}{4\pi\epsilon_0} \right|^2 \\ &= \frac{8\pi}{3} k_L^4 \hbar^2 |\alpha^{(0)}|^2. \end{aligned} \quad (\text{C10})$$

We can compare to the classical result by using the Clausius-Mossotti relation,

$$|\alpha^{(0)}|^2 = \left(\frac{3\epsilon_0}{N}\right)^2 \left(\frac{\epsilon_r - 1}{\epsilon_r + 2}\right) = \left(\frac{3\epsilon_0}{N}\right)^2 \left(\frac{n^2 - 1}{n^2 + 2}\right) \approx \left(\frac{3\epsilon_0}{N}\right)^2 \frac{4}{9}(n - 1)^2, \quad (\text{C11})$$

where N is the number density of particles in the material, ϵ_r is the dielectric constant, and n is the refractive index. The second equality is true for non-magnetic media, and the third, approximate equality holds when $n \approx 1$. Applying this to Eqn. (C10), we find

$$\sigma_{\text{red}} \approx \frac{2k_L^4}{3\pi N^2} |n - 1|^2, \quad (\text{C12})$$

which is the expression for classical Rayleigh scattering (*e.g.* Jackson [32]). Note that if we had ignored the V scattering process, *i.e.*, the $1/(\omega_{ki} + \omega_L)$ term, we would not have recovered the correct limits.