Notes on two photon scattering of the molecular state

November 24, 2019

1 Experiment observations

We have seen experimental evidences of an anomalous scattering / loss in the lifetime of the ground atomic / molecular states as well as broadening in excited state line width. We have measured power dependencies for some of the processes that is consistent with a two photon process from the ground state (or equivalently a one photon process from the excited state). The measurement we have done includes

1. Line width and decay time measurement of Raman process near $v = 0$ (1038nm)
   The result is consistent with a 300MHz to 600MHz excited state line width.
   We did not make a power dependency measurement.
   We measured carefully at multiple red detunings, which all shows similar results.
   We measured one point not as carefully for blue detuning (18GHz or 10GHz detuned depending on the excited state J) which shows similar result as the corresponding red detuning.

2. Off resonance PA rate (lifetime) in $|F^{\mathrm{Cs}} = 4, m_F^{\mathrm{Cs}} = 4; F^{\mathrm{Na}} = 2, m_F^{\mathrm{Na}} = 2\rangle$ near $v = 0$ PA line.
   The result is consistent with the Raman line width and decay time measurements above which gives a wide excited state line width.

3. Excited state line width for $v = 0$.
   The line width is narrow ($\leqslant$ 50MHz) when the tweezer is near 976nm but is wide ($\geqslant$ 500MHz) when the tweezer is near 1038nm red detuned from the $v = 0$ resonance by a few hundred GHz.
   We did not make a power dependency measurement.

4. Line width of a "mysterious" line
   Tweezer 100GHz red detuned.
   Line width $\approx$ 100MHz (determined to be narrow at the time but could have some broadening)
   We did not make a power dependency measurement.

5. $|F^{\mathrm{Cs}} = 3, m_F^{\mathrm{Cs}} = 3; F^{\mathrm{Na}} = 2, m_F^{\mathrm{Na}} = 2\rangle$ lifetime in 307366GHz tweezer.
   We measured a shorter than expected lifetime.
   The dependency of the loss rate to tweezer power is consistent with the prediction of a two photo process from the tweezer. In another word, it is consistent with a excited state broadened proportional to the tweezer power.

6. $|F^{\mathrm{Cs}} = 3, m_F^{\mathrm{Cs}} = 3; F^{\mathrm{Na}} = 2, m_F^{\mathrm{Na}} = 2\rangle$ lifetime in 306638GHz tweezer.
   We measured a shorter than expected lifetime.
   The power dependency may be consistent with a single photon process.

7. $v = 12$ line width.
   Narrow (25MHz) for tweezer at around 307366GHz (blue detuned $\approx$ 900GHz). (No tweezer power dependency taken.)
   Narrow (20MHz) for tweezer at 306612GHz (blue detuned $\approx$ 130GHz). (No tweezer power dependency taken.) We have also indirectly observed a Raman coupling to a deeper molecular bound state between the tweezer and the PA probe beam.
   Broad (250MHz) for tweezer at 306344.5GHz (red detuned $\approx$ 150GHz). Linearly depending on tweezer power.

8. $v = 14$ line width.
   Broad (200MHz) for tweezer at around 307366GHz (red detuned $\approx$ 1900GHz). Linearly depending on tweezer power.

9. Raman blue detuned from $v = 12$.
   Tweezer at around 306614GHz.
   No Rabi flopping observed.

2 Preliminary explanations for some of the results

The power dependencies we measured makes it clear that we very likely have some two photon (from ground state, one photon from excited state) process involved. We originally suspected a two photon process to a higher excited state but Olivier also suggested potentially a coupling to the ground state (unbound) motional continuum. This initially seems improbable due to the small matrix element with the ground state. However, I later learned that due to the mismatch in the excited state and ground state electronic potential, the excited state only have about 20% projection onto the ground bound state. In another word, even though the coupling to a particular ground trapped state is small, the total coupling to the ground state continuum is actually significant.
In terms of a two photon coupling (from ground state) to higher excited state, it is harder to calculate but it is easy to believe the existence of highly lossy state either from radiative or non-radiative (pre-dissociation) process.
In terms of a two photon coupling (from ground state) back to the ground electronic state, it should be easier to calculate (see below) and can more easily explain the apparent sensitivity to the detuning (red vs blue) of the excited state line width measurements. We have also directly measured a coupling to the ground bound state when we switched to blue detuning.

3 Caveat in the interpretation

There are two kinds of measurements above

1. Ground state scattering/loss rate
   This includes Raman decay time / line width and atomic state 2-body lifetime.

2. Excited state line width.

The first kind of measurement mainly involve a single laser frequency or two that are not very far apart ($\approx$ 300MHz). This single frequency is also detuned from any known excited state frequency by tens to hundreds of GHz (up to around 1THz). This is more directly related to the loss we care about. (FWIW, the loss during Raman process is one of this type of measurements).
The second kind of measurement usually involve two laser frequencies. [1] One of the two

[1] We've done measurements using the tweezer as PA without additional frequencies but those data are generally assumed to be power broadened since we cannot turn down the tweezer power too much during the measurement. Therefore, the tweezer PA measurement don't usually offer us much information on the excited state line width.

frequencies is detuned by few GHz to a THz and the other one is scanned around the resonance by up to a few GHz.
Since we usually express the difference in the ground state scattering rate in terms of a difference in the excited state line width (lifetime), it is tempting to compare this line width to the excited state line width direction measured at the same tweezer frequency. This is the main reason why we were measuring Raman transition using tweezer around 306614GHz after observing narrow line width for the nearby excited state.
However, the two are not directly comparable. When we use the directly measured excited state line width (Γ_e) to calculate the ground state scattering rate (γ) for a certain large detuning (Δ) as,

$$\gamma = \Gamma \frac{\Omega_1 \Omega_2}{\Delta^2}$$

(where Ω_1 and Ω_2 are the single photon Rabi frequencies), we have assumed that the loss of the excited state (i.e. the continuum the excited state is coupled to in order to decay) does not depend strongly on the final energy of the two photon process. Depending on the structure of the continuum we couple to via the two photon process, this may be a bad assumption.
This has subtly different implications for two photon couple up or two photon couple down.

3.1 Two photon coupling up

See figure 1A, the loss mechanism we suspect is a two photon coupling to a lossy continuum with a frequency dependent loss rate [2]. Generically, the loss rate is not flat which may give us a frequency dependent two photon loss rate. One way to see the difference between a detuned measurement and a on-resonance (with the singe photon excited state $|e\rangle$) measurement of the two photon decay rate is that the two process does not have the same two photon frequencies. For the on-resonance measurement using the same laser that couples $|e\rangle$ to the higher excited state (figure 1B), the loss rate experienced by the two photon resonance can be different. Since the loss rate can have an arbitrary dependency on frequency, one shouldn't expect the difference in the loss rate to be included in the detuning factor when calculating the scattering in the off-resonance case.

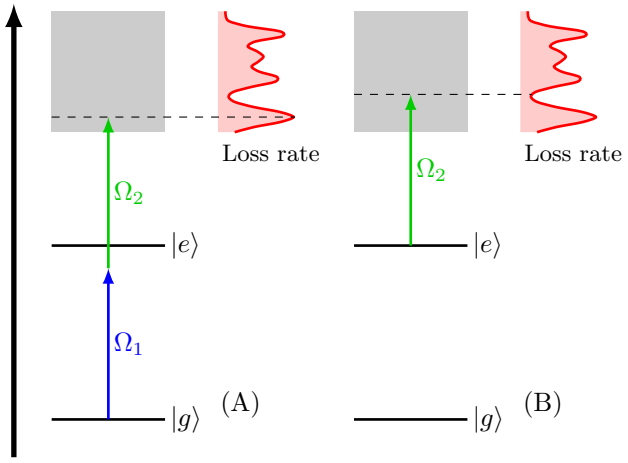

Figure 1: Two photon coupling to a (lossy) continuum. (A) Off resonance measurement, (B) On resonance measurement.

As a more concrete example, we'll consider a single two photon excited state with a stable single photon excited state.

[2] The loss rate depends on the laser power, i.e. Ω_1 and Ω_2. Here "loss rate" essentially mean the loss rate per power squared, which is proportional to the square of the coupling from $|e\rangle$ and the density of state. Also note here since the second beam is coupling to a continuum rather than a single state in the generic case, Ω_2 is not a single number but a function of frequency depending of the selection of normalization of the continuum.

See figure 2. The ground state $|g\rangle$ is coupled to $|e_1\rangle$, $|e_1\rangle$ is coupled to $|e_2\rangle$ and $|e_2\rangle$ can decay to another ground state $|g'\rangle$ with decay rate Γ and a 100% branching ratio. The energy of the two ground states $|g\rangle$ and $|g'\rangle$ are both 0, the energy of $|e_1\rangle$ is ω_0 and the energy of $|e_2\rangle$ is $\omega_0 + \omega$. The states are coupled by a single external laser with frequency ω. The laser drives the $|g\rangle \leftrightarrow |e_1\rangle$ transition with a Rabi frequency Ω_1 and the $|g\rangle \leftrightarrow |e_1\rangle$ transition with a Rabi frequency Ω_2. The single photon detuning on the $|e_1\rangle$ and the two photon detuning on $|e_2\rangle$ are both $-\Delta \equiv \omega - \omega_0$. For simplicity we have ignored the decay of the $|e_1\rangle$ state and we assume the laser frequency is the same as the $|e_1\rangle$ $|e_2\rangle$ energy splitting. We will also assume that $\Omega_{1,2} \ll \Delta \ll \omega < \omega_0$ as well as $\Gamma \ll \Delta$. Some of the conditions may not be true in the real experiment but this example should be enough to demonstrate the correct way to calculate the scattering rate. We'll calculate the scattering rate of the state $|g\rangle$ using a few different methods. [3]

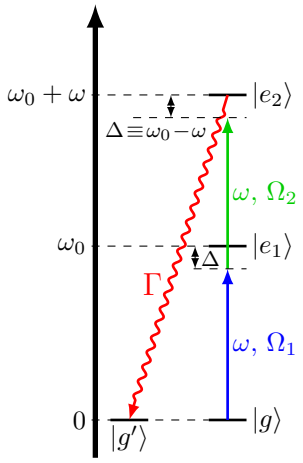

Figure 2: Four level model.

3.1.1 Off resonance Raman scattering

We can treat this as an off-resonance Raman scattering process. The Raman Rabi rate for the $|g\rangle \to |e_1\rangle \to |e_2\rangle$ is

$$\Omega_R = \frac{\Omega_1 \Omega_2}{2\Delta}$$

since $\Omega_R \ll \Omega_1 \ll \Delta$, the off resonant scattering rate is,

$$\begin{aligned}\gamma \approx & \frac{\Gamma}{4}\frac{\Omega_R^2}{\Delta^2} \\ = & \frac{\Gamma}{16}\frac{\Omega_1^2\Omega_2^2}{\Delta^4}\end{aligned}$$

3.1.2 Single photon off resonance scattering from coupled excited state

More directly related to the excited state line width measurement, we can first couple the two excited states together with Ω_2.
See figure 3A. The excited states becomes

$$|\pm\rangle = \frac{1}{\sqrt{2}}(|e_1\rangle \pm |e_2\rangle)$$

with energy $\omega_0 \pm \Omega_2/2$ and each with a lifetime of $\frac{\Gamma}{2}$. The Rabi frequency between the two excited states and the ground state is $\Omega_\pm = \frac{\Omega_1}{\sqrt{2}}$.

[3] All of the calculation below ignores the frequency dependency of the $|g'\rangle$ continuum $|e_2\rangle$ decays to. All the scattering rate should be scaled by a factor of $\left(\frac{2\omega}{\omega + \omega_0}\right)^3$.

The scattering from the two excited states are

$$\begin{aligned}\gamma_{\pm} \approx& \frac{\Gamma}{8}\frac{\Omega_{\pm}^2}{\left(\Delta \mp \Omega_2/2\right)^2}\\ \approx& \frac{\Gamma}{16}\frac{\Omega_1^2}{\Delta^2}\end{aligned}$$

and the total scattering rate

$$\begin{aligned}\gamma =& \gamma_- + \gamma_+\\ \approx& \frac{\Gamma}{8}\frac{\Omega_1^2}{\Delta^2}\end{aligned}$$

which clearly
doesn't agree with the result we calculated above.

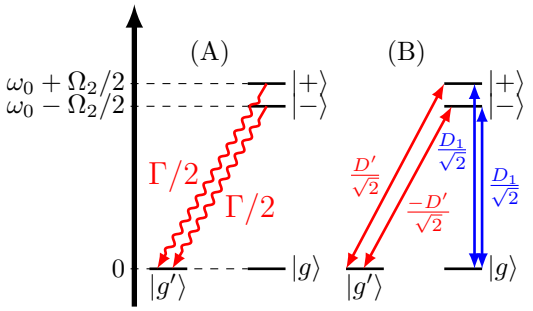

Figure 3: Coupled excited states. (B) Dipole moments between new states.

3.1.3 Single photon off resonance scattering from coupled excited state (the correct way)

The apparent disagreement between the two result above suggests that one of them must be wrong and it turns out that the second one is incorrect due to the wrong assumption it made. In this case, the issue manifests itself as an incorrect off resonant scattering rate from the excited state. There is not problem in coupling the excited state first to get $|\pm\rangle$ but the decay process from the two states are not independent which give rise to a different scattering rate away from resonance. (when $\Delta \gg \Omega_2$). Instead, the scattering from the two excited states have to be treated as interfering Raman processes and their amplitudes needs to be added, which will lead to cancellation as we'll see soon, before squared to get the scattering rate.
The correct way to calculate this requires using the dipole moment between the states [4]. For the purpose of this note, however, it is enough to only calculate the interference term and scales our previous result correspondingly.
Let the $D_1 = \langle g|\hat{d}|e_1\rangle$ and $D_2 = \langle g'|\hat{d}|e_2\rangle$ be the dipole moment coupling e_1 and e_2 to the corresponding ground states g and g'. The dipole moments between the new states are shown in figure 3B [5].
The wrong way to calculate the scattering gives a factor of

$$\begin{aligned}&\left(\frac{D'}{\sqrt{2}}\frac{D_1}{\sqrt{2}}\frac{1}{\Delta + \Omega_2/2}\right)^2 + \left(\frac{-D'}{\sqrt{2}}\frac{D_1}{\sqrt{2}}\frac{1}{\Delta - \Omega_2/2}\right)^2\\ \approx& 2\left(\frac{D'}{\sqrt{2}}\frac{D_1}{\sqrt{2}}\frac{1}{\Delta}\right)^2\\ =& \frac{D'^2 D_1^2}{2\Delta^2}\end{aligned}$$

[4] And the formula also correctly takes into account the density of state of the scattered photon and gives the correct $\left(\frac{2\omega}{\omega + \omega_0}\right)^3$ factor

The correct way gives,

$$\begin{aligned}
&\left(\frac{D'}{\sqrt{2}}\frac{D_1}{\sqrt{2}}\frac{1}{\Delta+\Omega_2/2}+\frac{-D'}{\sqrt{2}}\frac{D_1}{\sqrt{2}}\frac{1}{\Delta-\Omega_2/2}\right)^2\\
=&\frac{D'^2D_1^2}{4}\left(\frac{1}{\Delta+\Omega_2/2}-\frac{1}{\Delta-\Omega_2/2}\right)^2\\
=&\frac{D'^2D_1^2}{4}\left(\frac{\Delta-\Omega_2/2-\Delta-\Omega_2/2}{\Delta^2-\Omega_2^2/4}\right)^2\\
\approx&\frac{D'^2D_1^2\Omega_2^2}{4\Delta^4}
\end{aligned}$$

which means that we should scale the incorrect result above by

$$\frac{D'^2D_1^2\Omega_2^2}{4\Delta^4}\frac{2\Delta^2}{D'^2D_1^2}=\frac{\Omega_2^2}{2\Delta^2}$$

and the correct answer is

$$\begin{aligned}
\gamma=&\frac{\Gamma}{8}\frac{\Omega_1^2}{\Delta^2}\frac{\Omega_2^2}{2\Delta^2}\\
=&\frac{\Gamma}{16}\frac{\Omega_1^2\Omega_2^2}{\Delta^4}
\end{aligned}$$

which is exactly what we got above.

3.1.4 Summary for two photon coupling up

In the particular case (two photon excite to a single state) we calculated, the naive calculation using coupled excited state (i.e. direct result from PA measurement) gives the wrong result because of the interference from different scattering channels. More generally, to avoid this problem one need to realize,

1. The scattering is never a one photon process, one need to calculate the rate after considering all photon involved.

2. It is OK to couple some intermediate states first and use a dressed state picture to eliminate the number of photon/coupling/intermediate states involved. However, the coupled result may not be a single state and the scattering from the resulting states may not follow a simple Δ^{-2} rule.

3.2 Two photon coupling down

The potential second photon coupling down pathway is very similar. See figure 4A. The loss is caused by the ground state (atomic or molecular) coupling to the ground motional continuum. We can also directly apply the two points from the summary above.

1. In order to calculate ground state loss, we should consider the Raman process, which will likely have very different motional energies compared to the motional energy coupling down from the excited state.

2. The excited state is coupled to an atomic motional continuum and the resulting loss will have a very different profile from then Δ^{-2} scaling. Even if we see a broadening of the excited state due to this coupling, it does not mean much for the corresponding ground state loss rate.

The two kinds of ground state scattering measurements we have are atomic state PA rate and molecular state scattering (using Raman transition). Assuming the laser is clean and no other states are involved,

1. For atomic scattering rate (figure 4B), there is only one frequency so the Raman process can only come back down to the same energy as the initial state, which is not in an continuum.

2. For molecular scattering rate during transfer (figure 4C), similarly, the only energy the molecular state could couple to is the initial energy or a deeper bound state (2x the binding energy). Neither of these is a continuum either. The atomic state can indeed be coupled to the continuum but it should be much weaker than the molecular state (the up leg is much smaller) and we have not measured a decrease in atomic 2-body lifetime when the Raman beam is turned on.

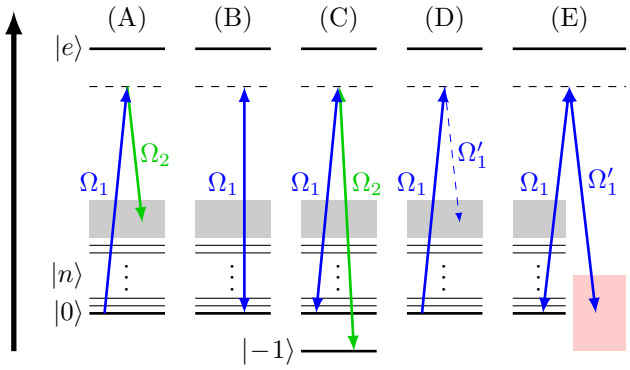

Figure 4: Coupling to ground state motional continuum. (A) Raman into motional continuum. (B) Single ground HF state, single laser frequency. (C) Single ground HF state, two frequencies for molecular Raman. (D) Unexpected sideband in laser. (E) Zero energy continuum.

Therefore, in order to generate any loss from this process for the ground state, we need to have either,

1. Additional frequencies in the laser (figure 4D) which can couple the state of interest to the motional continuum. Or,

2. Continuum that has zero energy (figure 4E) that the Raman beam can couple to. Candidate for this continuum includes the center of mass (COM) motional states and the motional continuum from other HF states.
The COM motional continuum actually also starts at a positive energy so it doesn't seems possible. The continuum from other HF states is much more promising especially since we know that the $|F^{\mathrm{Cs}}=3, m_F^{\mathrm{Cs}}=3; F^{\mathrm{Na}}=2, m_F^{\mathrm{Na}}=2\rangle$ state has a strong coupling to the molecular excited states. However, this cannot be applied to the $|F^{\mathrm{Cs}}=3, m_F^{\mathrm{Cs}}=3; F^{\mathrm{Na}}=2, m_F^{\mathrm{Na}}=2\rangle$ state lifetime since it is already the HF state with the lowest energy.

3.3 Summary for interpreting and connecting excited state and ground state results

From the discussion above, we see that no matter what kind of couple we have, the excited state line width may not be directly related to the life time in the tweezer of the corresponding frequency. For up coupling, the result is more closely related to the lifetime for tweezer of the average frequency, whereas for down coupling, the lifetime may not be related to the excited state line width at all.

4 Loss due to coupling to ground state motional continuum

Calculating the loss rate for coupling to the continuum requires knowledge of the continuum wavefunction. This is hard to do accurately for states with significant energy since the fine energy spacing will blow up the number of states involved. Fortunately, since we know the interaction potential is short range and much deeper than any of the motional energy scales, we can make certain approximations in order to get the coupling to the continuum based on the known coupling to the trapped atomic states in the tweezer.
First of all, we are going to ignore the COM motion and only consider the relative motion. This shouldn't affect our answer for coupling to the relative motional continuum too much but it does prevent us from calculating the coupling to the COM motional continuum. Since the COM motional continuum only exists at roughly the same energy levels as the relative motion, this should not be an issue (the coupling to the relative continuum at the same energy should always be stronger).
We'll also only consider $L = 0$ motional states since,

1. The selection rule means that we can only couple to at most a few low L states.
2. The coupling to $L \neq 0$ states are weaker due to smaller wave function amplitude for short distance.

We can then consider a small enough volume around the $r = 0$ of the relative coordinate. The coupling to the excited molecular state is entirely determined by the wavefunction inside. The wavefunction inside is determined by the boundary condition, which for $L = 0$[6] is a single number, and the energy of the state. Since the energy of the motional state is much smaller than the depth of the interaction potential, we can expect the shape of the wavefunction to be almost independent of the energy and the amplitude of it to be proportional to the boundary condition. In the weak interaction limit[7], we can assume that the boundary condition is given by the value of the non-interacting wavefunctions at $r = 0$.
In the ground motional state, the trapping frequencies are ω_i where $i = 1, 2, 3$ along the three axis. The value of the wavefunction at $r = 0$ is,

$$\begin{aligned}\psi_0 =& \prod_{i=1}^{3} \left(\frac{m\omega_i}{\pi\hbar}\right)^{1/4} \\ =& \left(\frac{m^3\omega_1\omega_2\omega_3}{\pi^3\hbar^3}\right)^{1/4}\end{aligned}$$

Assume the Raman Rabi frequency between the initial state and the ground motional state is Ω_R, the Raman Rabi frequency for another motional state would be,

$$\Omega_R' = \Omega_R\psi'(r=0)\left(\frac{\pi^3\hbar^3}{m^3\omega_1\omega_2\omega_3}\right)^{1/4}$$

In order to calculate the coupling to the continuum, we'll start with a discrete set of states and and then take the energy gap to 0 to calculate the result for true continuum. We'll use two different sets of states to calculate for cross-checking[8].

[6] Or for any particular L and m_L

[7] This should be true for most motional state with the exception of $|F^{\mathrm{Cs}}=3, m_F^{\mathrm{Cs}}=3; F^{\mathrm{Na}}=2, m_F^{\mathrm{Na}}=2\rangle$ state.

[8] Note that both calculations will be done under the same assumptions given above so their agreement does not necessarily mean that the assumption is valid.

4.1 Harmonic oscillator states

We can treat the continuum as the limit of a infinitely loose 3D harmonic oscillator state. Assuming a trapping frequency ω, we are interested in the value of the wavefunction at $r = 0$ for large n where n is the radial quantum number (energy gap of 2ω).
I'll calculate this value using two methods as well.

4.1.1 Using radial wavefunction $u = r\psi_r$

After doing the subsitution of $u = r\psi_r(r)$, the Schrödinger equation for u becomes that of a 1D harmonic oscillator with a hard wall at $r = 0$. The eigenstates are the odd harmonic oscillator states for the normal 1D potential and $\psi(r = 0)$ is given by $\left.\frac{1}{\sqrt{4\pi}}\frac{\mathrm{d}u}{\mathrm{d}r}\right|_{r=0}$.
The normalized wavefunction for u is,

$$
\begin{aligned}
u_n =& \frac{(-1)^n\sqrt{2}}{\sqrt{2^{2n+1}(2n+1)!}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{1/4}\exp\left(-\frac{m\omega r^2}{2\hbar}\right)H_{2n+1}\left(\sqrt{\frac{m\omega}{\hbar}}r\right)\\
\psi(r=0) =& \left.\frac{1}{\sqrt{4\pi}}\frac{\mathrm{d}u}{\mathrm{d}r}\right|_{r=0}\\
=& \frac{1}{\sqrt{2\pi}}\frac{(-1)^n}{\sqrt{2^{2n+1}(2n+1)!}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{1/4}\frac{\mathrm{d}}{\mathrm{d}r}H_{2n+1}\left(\sqrt{\frac{m\omega}{\hbar}}r\right)\Big|_{r=0}\\
=& \frac{1}{\sqrt{2\pi}}\frac{(-1)^n}{\sqrt{2^{2n+1}(2n+1)!}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{1/4}\sqrt{\frac{m\omega}{\hbar}}2(2n+1)H_{2n}(0)\\
=& \frac{(-1)^n}{\sqrt{2^{2n+2}(2n+1)!}}2(2n+1)(-2)^n(2n-1)!!\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
=& \frac{2^{n+1}(2n+1)(2n-1)!}{(n-1)!2^{n-1}\sqrt{2^{2n+2}(2n+1)!}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
=& \sqrt{\frac{(2n+1)!}{2^{2n}n^2}}\frac{1}{(n-1)!}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
=& \sqrt{\frac{\sqrt{2\pi(2n+1)}(2n+1)^{2n+1}}{2^{2n}n^2\mathrm{e}^{2n+1}}}\frac{\mathrm{e}^{n-1}}{\sqrt{2\pi(n-1)}(n-1)^{n-1}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
\approx& \sqrt{\frac{\sqrt{2\pi(2n+1)}(2n+1)^{2n+1}}{\pi\mathrm{e}^3(2n-2)^{2n+1}}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
\approx& \left(\frac{4n}{\pi}\right)^{1/4}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}
\end{aligned}
$$

4.1.2 Using 3D harmonic wavefunction

$$\begin{aligned}
\psi(r=0) =& \frac{1}{\sqrt{4\pi}}\sqrt{\sqrt{\frac{m^3\omega^3}{4\pi\hbar^3}}\frac{2^{n+3}n!}{(2n+1)!!}}L_n^{1/2}(0)\\
=& \frac{1}{\sqrt{4\pi}}\sqrt{\sqrt{\frac{m^3\omega^3}{4\pi\hbar^3}}\frac{2^{n+3}2^n}{(2n+1)!}}n!L_n^{1/2}(0)\\
\approx& \frac{1}{\sqrt{4\pi}}\sqrt{\sqrt{\frac{m^3\omega^3}{\pi\hbar^3}}\frac{2^{2n+2}}{(2n+1)!}}\frac{n!2\sqrt{n}}{\sqrt{\pi}}\\
\approx& \frac{1}{\sqrt{4\pi}}\sqrt{\frac{\pi 2^{2n+2}\mathrm{e}^{2n+1}}{\sqrt{2\pi(2n+1)}(2n+1)^{2n+1}}}\sqrt{2\pi n}\Big(\frac{n}{\mathrm{e}}\Big)^n\frac{2\sqrt{n}}{\sqrt{\pi}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
=& \sqrt{\frac{2\pi(2n)^{2n+2}\mathrm{e}}{\pi\sqrt{2\pi(2n+1)}(2n+1)^{2n+1}}}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\\
\approx& \left(\frac{4n}{\pi}\right)^{1/4}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}
\end{aligned}$$

which agrees with the other method.
The Raman Rabi frequency to the motional state is therefore,

$$\begin{aligned}
\Omega_R' =& \Omega_R\psi'(r=0)\left(\frac{\pi^3\hbar^3}{m^3\omega_1\omega_2\omega_3}\right)^{1/4}\\
=& \Omega_R\left(\frac{4n}{\pi}\right)^{1/4}\Big(\frac{m\omega}{\pi\hbar}\Big)^{3/4}\left(\frac{\pi^3\hbar^3}{m^3\omega_1\omega_2\omega_3}\right)^{1/4}\\
=& \Omega_R\left(\frac{4n\omega^3}{\pi\omega_1\omega_2\omega_3}\right)^{1/4}
\end{aligned}$$

The decay rate is

$$\begin{aligned}
\gamma =& \frac{2\pi\Omega_R'^2}{2\omega}\\
=& \frac{\pi\Omega_R^2}{\omega}\sqrt{\frac{4n\omega^3}{\pi\omega_1\omega_2\omega_3}}\\
=& \Omega_R^2\sqrt{\frac{2\pi E}{\omega_1\omega_2\omega_3}}
\end{aligned}$$

where E is the energy (in frequency unit) of the continuum energy we are coupling to.