Notes on two photon scattering of the molecular state

November 23, 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. 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.

3. 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.

4. $|F^{Cs} = 3, m_F^{Cs} = 3; F^{Na} = 2, m_F^{Na} = 2\rangle$ 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.

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

6. $v = 12$ 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.

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

8. 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 frequencies is detuned by few GHz to a THz and the other one is scanned around the resonance by up to a few

[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.

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.

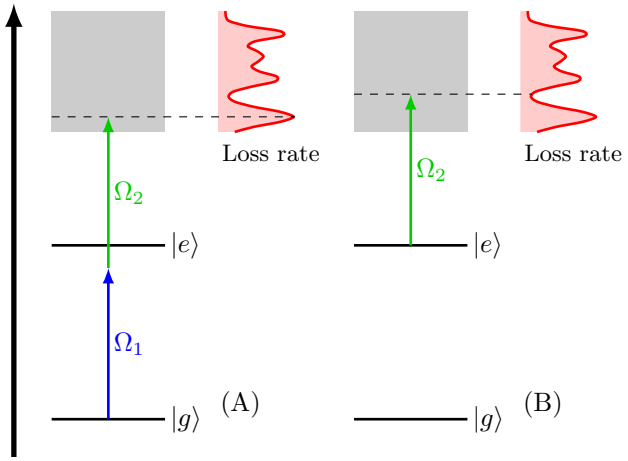

Figure 1: Two photon coupling to a (lossy) continuum. (A) Off resonance measurement, (B) On resonance measurement.

[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.

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

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 $-\Delta \equiv \omega - \omega_0$.

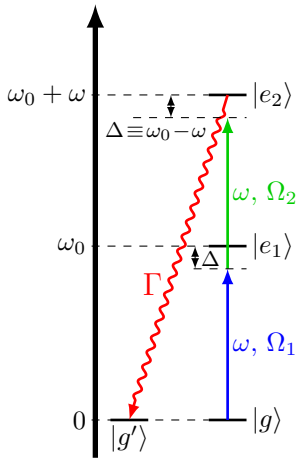

Figure 2: Four level model.

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]

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 linewidth measurement, we can first couple the two excited states together with Ω_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 $\left(\frac{2\omega}{\omega + \omega_0}\right)^3$.

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 $\Omega_\pm = \frac{\Omega_1}{\sqrt{2}}$.
The scattering from the two excited states are

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

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.

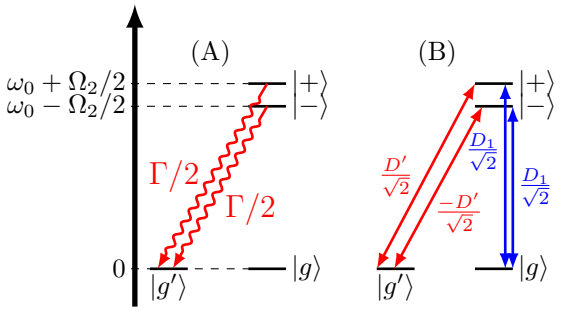

Figure 3: Coupled excited states. (B) Dipole moments between new states.

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.

3.2 Two photon coupling down