Asymmetry in population distribution with respect to detuning caused by EIT/coherent scattering

May 9, 2023

1 Introduction

While simulating a simple three-level system with a Raman transition coupling two ground states and an excited state with a finite lifetime, I noticed that there's a difference in the dynamic and the final/steady-state population distribution when the two-photon detuning changes sign.

This effect cannot be reproduced in a two-level system if we assume the two ground states scatters independently, even if we include the difference in the scattering rate of the two states caused by the slight difference in the single photon detuning when a non-zero two photon detuning. It could be reproduced, OTOH, if the initial state of the scattering is assumed to be a superposition of the two ground states. This difference is of course very important since it's also where EIT comes from.

While the result of the simulation is pretty clear and with a 2-by-2 density matrix it shouldn't be that difficult to write out the full master equation and solve it directly, I do want to understand the origin of this asymmetry better and here are some of the approaches I can think of to understand this phenomenon.

2 System description

Hamiltonian,

$$H = \frac{1}{2}(\delta\sigma_z + \Omega\sigma_x) \tag{1}$$

For scattering, we'll assume that the state $|\psi_s\rangle \equiv \dfrac{|0\rangle + |1\rangle}{\sqrt{2}}$ scatters at a rate of γ with a 50/50 branching ratio back to the $|0\rangle$ and $|1\rangle$ states.

3 Steady state solution in the eigen basis of the Hamiltonian

The eigen states of the Hamiltonian is,

$$|\psi_1\rangle = \begin{pmatrix} -\sqrt{\frac{1}{2} - \frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \\ \sqrt{\frac{1}{2} + \frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \end{pmatrix} \tag{2}$$

$$|\psi_2\rangle = \begin{pmatrix} \sqrt{\frac{1}{2} + \frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \\ \sqrt{\frac{1}{2} - \frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \end{pmatrix} \tag{3}$$

Since there is no coupling between these two states and there's no coherence in the scattering final state (because of the 50/50 branching ratio) we can complete ignore any coherence between these two states and simply treat this using a scattering rate equation. The overlap between these two states and the scattering state $|\psi_s\rangle$ is,

$$\langle\psi_1|\psi_s\rangle = \frac{1}{2}\left(\sqrt{1+\frac{\delta}{\sqrt{\delta^2+\Omega^2}}} - \sqrt{1-\frac{\delta}{\sqrt{\delta^2+\Omega^2}}}\right) \tag{4}$$

$$\langle\psi_2|\psi_s\rangle = \frac{1}{2}\left(\sqrt{1+\frac{\delta}{\sqrt{\delta^2+\Omega^2}}} + \sqrt{1-\frac{\delta}{\sqrt{\delta^2+\Omega^2}}}\right) \tag{5}$$

Scattering rates,

$$\begin{aligned}
\gamma_1 =& \gamma|\langle\psi_1|\psi_s\rangle|^2 \\
=& \frac{\gamma}{4}\left(\sqrt{1+\frac{\delta}{\sqrt{\delta^2+\Omega^2}}} - \sqrt{1-\frac{\delta}{\sqrt{\delta^2+\Omega^2}}}\right)^2 \\
=& \frac{\gamma}{4}\left(1+\frac{\delta}{\sqrt{\delta^2+\Omega^2}} + 1 - \frac{\delta}{\sqrt{\delta^2+\Omega^2}} - 2\sqrt{1+\frac{\delta}{\sqrt{\delta^2+\Omega^2}}}\sqrt{1-\frac{\delta}{\sqrt{\delta^2+\Omega^2}}}\right) \\
=& \frac{\gamma}{2}\left(1-\frac{\Omega}{\sqrt{\delta^2+\Omega^2}}\right)
\end{aligned} \tag{6}$$

$$\begin{aligned}
\gamma_2 =& \gamma|\langle\psi_2|\psi_s\rangle|^2 \\
=& \frac{\gamma}{2}\left(1+\frac{\Omega}{\sqrt{\delta^2+\Omega^2}}\right)
\end{aligned} \tag{7}$$

Since the branching ratio for the scattering is still 50/50 in this basis, the population ratio at steady state is the inverse of the scattering rate ratio of the two states.

$$\begin{aligned}
\frac{p_{\psi_1}}{p_{\psi_2}} =& \frac{\gamma_2}{\gamma_1} \\
=& \frac{\sqrt{\delta^2+\Omega^2}+\Omega}{\sqrt{\delta^2+\Omega^2}-\Omega}
\end{aligned} \tag{8}$$

$$p_{\psi_1} = \frac{\sqrt{\delta^2+\Omega^2}+\Omega}{2\sqrt{\delta^2+\Omega^2}} \tag{9}$$

$$p_{\psi_2} = \frac{\sqrt{\delta^2+\Omega^2}-\Omega}{2\sqrt{\delta^2+\Omega^2}} \tag{10}$$

$$\tag{11}$$

The density matrix

$$
\begin{aligned}
\rho =&|\psi_1\rangle p_{\psi_1}\langle\psi_1| + |\psi_2\rangle p_{\psi_2}\langle\psi_2| \\
=&\begin{pmatrix} -\sqrt{\frac{1}{2}-\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} & \sqrt{\frac{1}{2}+\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \end{pmatrix} \frac{\sqrt{\delta^2+\Omega^2}+\Omega}{2\sqrt{\delta^2+\Omega^2}} \begin{pmatrix} -\sqrt{\frac{1}{2}-\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \\ \sqrt{\frac{1}{2}+\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \end{pmatrix} + \\
&\begin{pmatrix} \sqrt{\frac{1}{2}+\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} & \sqrt{\frac{1}{2}-\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \end{pmatrix} \frac{\sqrt{\delta^2+\Omega^2}-\Omega}{2\sqrt{\delta^2+\Omega^2}} \begin{pmatrix} \sqrt{\frac{1}{2}+\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \\ \sqrt{\frac{1}{2}-\frac{\delta}{2\sqrt{\delta^2+\Omega^2}}} \end{pmatrix} \\
=&\begin{pmatrix} \sqrt{\delta^2+\Omega^2}-\delta & -\Omega \\ -\Omega & \sqrt{\delta^2+\Omega^2}+\delta \end{pmatrix} \frac{\sqrt{\delta^2+\Omega^2}+\Omega}{4(\delta^2+\Omega^2)} + \\
&\begin{pmatrix} \sqrt{\delta^2+\Omega^2}+\delta & \Omega \\ \Omega & \sqrt{\delta^2+\Omega^2}-\delta \end{pmatrix} \frac{\sqrt{\delta^2+\Omega^2}-\Omega}{4(\delta^2+\Omega^2)} \\
=&\begin{pmatrix} \big(\sqrt{\delta^2+\Omega^2}-\delta\big)\big(\sqrt{\delta^2+\Omega^2}+\Omega\big) & -\Omega\big(\sqrt{\delta^2+\Omega^2}+\Omega\big) \\ -\Omega\big(\sqrt{\delta^2+\Omega^2}+\Omega\big) & \big(\sqrt{\delta^2+\Omega^2}+\delta\big)\big(\sqrt{\delta^2+\Omega^2}+\Omega\big) \end{pmatrix} \frac{1}{4(\delta^2+\Omega^2)} + \\
&\begin{pmatrix} \big(\sqrt{\delta^2+\Omega^2}+\delta\big)\big(\sqrt{\delta^2+\Omega^2}-\Omega\big) & \Omega\big(\sqrt{\delta^2+\Omega^2}-\Omega\big) \\ \Omega\big(\sqrt{\delta^2+\Omega^2}-\Omega\big) & \big(\sqrt{\delta^2+\Omega^2}-\delta\big)\big(\sqrt{\delta^2+\Omega^2}-\Omega\big) \end{pmatrix} \frac{1}{4(\delta^2+\Omega^2)} \\
=&\begin{pmatrix} \delta^2+\Omega^2-\delta\Omega & -\Omega^2 \\ -\Omega^2 & \delta^2+\Omega^2+\delta\Omega \end{pmatrix} \frac{1}{2(\delta^2+\Omega^2)}
\end{aligned} \tag{12}
$$

In another word, the population of $|0\rangle$ and $|1\rangle$

$$p_0 = \frac{1}{2}\left(1-\frac{\delta\Omega}{\delta^2+\Omega^2}\right) \tag{13}$$

$$p_1 = \frac{1}{2}\left(1+\frac{\delta\Omega}{\delta^2+\Omega^2}\right) \tag{14}$$

showing the asymmetry between the two states when the detuning changes sign.

We can compare this with the numerical simulation result (Fig. 1) showing very good agreement.

4 Qualitative discussion based on EIT

From the previous calculation, we can see that the density matrix in the z basis has non-diagonal terms meaning that there is actually coherence between the $|0\rangle$ and $|1\rangle$ states. This agrees with the observation that the selection of scattering state being important in causing the asymmetry.

We also see, as we expected, that when $\delta = 0$ the steady state density matrix represents a pure state $|\psi_d\rangle = \frac{|0\rangle - |1\rangle}{\sqrt{2}}$ which is the EIT dark state. For $\delta \neq 0$, the steady state is not the dark state but it remains fairly closed to the dark state for small detuning where the asymmetry is the most pronounced. This suggests that the dark state (and the eigen basis for scattering) is likely a better starting point / basis to understand the scattering process and potentially the asymmetry.

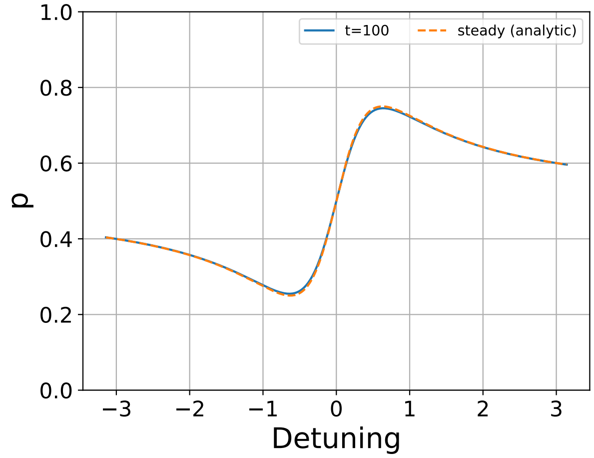

Figure 1: Comparison between the analytic steady state population of $|1\rangle$ and numerical result from master equation simulation.

We'll start by looking at the unitary evolution of the dark state $|\psi_d\rangle$ and see if there's any asymmetry when δ changes sign.

If the initial state of the system is the dark state $|\psi_d\rangle$, the time derivative of the wavefunction is,

$$\begin{aligned}\frac{\mathrm{d}|\psi\rangle}{\mathrm{d}t} =& -\mathrm{i}H|\psi_d\rangle\\ =& -\frac{\mathrm{i}}{2}(\delta\sigma_z + \Omega\sigma_x)\frac{|0\rangle - |1\rangle}{\sqrt{2}}\\ =& -\frac{\mathrm{i}}{2\sqrt{2}}((\delta - \Omega)|0\rangle + (\delta + \Omega)|1\rangle)\end{aligned} \tag{15}$$

While I'm not sure how it affects the dynamic yet, this indeed shows some asymmetry between $|0\rangle$ and $|1\rangle$ depending on if δ and Ω have the same sign or not. The amplitude of the derivative on $|1\rangle$ is larger than that on $|0\rangle$ if the two have the same sign, and smaller if the signs are different.

The change in the amplitude of both the $|0\rangle$ and the $|1\rangle$ states are out-of-phase with their original amplitude so there isn't any first order change in the state probabilities at $t = 0$. We can calculate the second order change to the wavefunction,

$$\begin{aligned}\frac{\mathrm{d}^2|\psi\rangle}{\mathrm{d}t^2} =& -H^2|\psi_d\rangle\\ =& -\frac{1}{4}(\delta\sigma_z + \Omega\sigma_x)^2\frac{|0\rangle - |1\rangle}{\sqrt{2}}\\ =& -\frac{1}{4\sqrt{2}}\left(\delta^2 + \Omega^2\right)(|0\rangle - |1\rangle)\end{aligned} \tag{16}$$

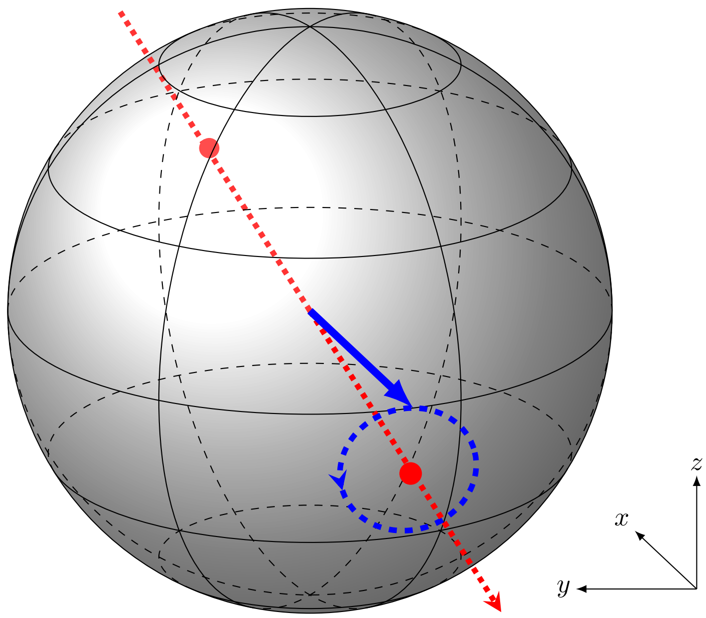

Figure 2: Unitary evolution of the dark state on the Bloch sphere. The blue arrow represent the dark state $|\psi_d\rangle$ and the red dotted arrow is the rotation axis determined by the Hamiltonian. The blue dashed circle is the trajectory the state make on the Bloch sphere.

And from this the second order change to the $|0\rangle$ probability,

$$\begin{aligned}\frac{\mathrm{d}^2|\langle 0|\psi\rangle|^2}{\mathrm{d}t^2} =& \frac{\mathrm{d}}{\mathrm{d}t}\left(\langle 0|\psi\rangle\frac{\mathrm{d}\langle\psi|0\rangle}{\mathrm{d}t} + \frac{\mathrm{d}\langle 0|\psi\rangle}{\mathrm{d}t}\langle\psi|0\rangle\right)\\ =&\langle 0|\psi\rangle\frac{\mathrm{d}^2\langle\psi|0\rangle}{\mathrm{d}t^2} + \frac{\mathrm{d}^2\langle 0|\psi\rangle}{\mathrm{d}t^2}\langle\psi|0\rangle + 2\frac{\mathrm{d}\langle 0|\psi\rangle}{\mathrm{d}t}\frac{\mathrm{d}\langle\psi|0\rangle}{\mathrm{d}t}\\ =&\frac{1}{4}(\delta-\Omega)^2 - \frac{1}{4}\left(\delta^2+\Omega^2\right)\\ =&\frac{\delta\Omega}{2}\end{aligned} \tag{17}$$

i.e. depending on the sign of $\delta\Omega$ the time evolution of the dark state will put more or less population in the $|0\rangle$ state. Indeed, we can see this clearly if we simply look at the time evolution on the Bloch sphere (Fig. 2). The initial vector is at $-\hat{x}$ (blue arrow). The hamiltonian corresponds to a rotation around the vector $\delta\hat{x} + \Omega\hat{z}$ (red dotted arrow). Depending on the sign of δ (actually $\delta\Omega$), the rotation axis (and therefore the time evolution of the state shown by the blue dashed circle) may be either above or below the equator.

Based on these observation, here is a semi-quatitative explaination of this phenomena.

1. The scattering process preferentially pumps the system into the dark state $\dfrac{|0\rangle - |1\rangle}{\sqrt{2}}$.
2. The Hamiltonian rotates the system around the axis $\delta\hat{x} + \Omega\hat{z}$ on the Bloch sphere, which is out of the equator plane when $\delta \neq 0$.
3. The interaction between these two effects means that the system is being pumped towards one side of the rotation axis and since the rotation axis is tilted the resulting state will have an uneven distribution between the $|0\rangle$ and $|1\rangle$ states.

In short, the symmetry in x is broken by the scattering/optical pumping term and this asymmetry is projected onto z by the Hamiltonian.

This can also explain the rough shape of the asymmetry. As δ increases, the coupling between x and z increases and there's more asymmetry in the steady state population. The coupling maximizes when $\delta = \Omega$ (i.e. $45°$ tilted axis) after which point the steady state population asymmetry decreases again.

We could also extend this explaination to other system configurations as well. If somehow the initial state of the scattering is random but the final state is always $\dfrac{|0\rangle + |1\rangle}{\sqrt{2}}$, this would result in an optical pumping effects towards $+x$ direction which would produce an asymmetry in the population in the other direction (confirmed by simulation). Similarly, if the OP process (either because of the initial or final state) is along the y axis, there will be no asymmetry based on the detuning since it is orthogonal to the rotation axis. There will, however, still be an asymmetry in the population since the rotation caused by the Hamiltonian will move the dark state closed to one of the poles.