The Role of Doppler Broadening in Electromagnetically Induced Transparency and Autler-Townes Splitting in Open Molecular systems

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(Dated: April 1, 2022)

Abstract

We describe in this Letter how inhomogeneous line broadening affects the Autler-Townes (AT) splitting in a three level open molecular cascade system. For moderate Rabi frequencies in the range of 300 to 500 MHz the fluorescence line shape from the uppermost level $|3\rangle$ in this system depends strongly on the frequency ratio of the two laser fields. However, the fluorescence spectrum of the intermediate level $|2\rangle$ appears as expected. We provide a description of the conditions for optimally resolved AT splitting in terms of the probe laser/coupling field frequency ratio and laser propagation geometry based on our theoretical analysis of the Doppler integral. This is important for applications such as molecular angular momentum alignment as well as for the measurement of the transition dipole moment matrix element.

PACS numbers: 33.40.+f, 42.50.Hz
Studies of coherence and quantum interference effects such as Autler-Townes (AT) splitting and electromagnetically induced transparency (EIT) in atomic and molecular systems have attracted a great deal of attention, because they yield a number of important potential applications including production of slow light, lasing without inversion, pulse matching effects and control of the index of refraction. EIT was demonstrated for the first time by Boller et al. in an atomic Λ-type Strontium system. It renders an otherwise optically thick medium transparent to a weak laser field. Most of the previous theoretical and experimental work on EIT is based on various three-level closed atomic systems with different energy level configurations using both pulsed and continuous wave (cw) lasers. To overcome inhomogeneous Doppler broadening it is generally expected that high coupling field intensities are required for observation of AT splitting and EIT. Nevertheless, it has been shown that with proper selection of experimental parameters and beam geometry it is possible to overcome this difficulty and to demonstrate these effects in both atomic and molecular systems with coupling field Rabi frequencies produced by commercially available cw lasers.

In our previous papers we emphasized that the AT effect can be used to directly measure the absolute value of the molecular transition dipole moment as well as to facilitate control of molecular angular momentum alignment. In this Letter we investigate the role of the inhomogeneous line broadening and how it affects the AT splitting in the fluorescence spectrum of levels |2⟩ and |3⟩ in a 3-level open molecular cascade system, when the coupling field is on resonance with the |2⟩ − |3⟩ rovibronic transition. We have observed that the fluorescence line shape originating from the uppermost level of the cascade excitation scheme critically depends on the probe laser/coupling field wavenumber ratio, while at the same time the fluorescence line shape observed from the intermediate level is hardly affected at all. Based on our theoretical analysis to explain these observations, we provide here a description of the optimal conditions for AT splitting in terms of the coupling field wavenumber compared to that of the probe laser as well as their relative propagation direction.

For the experimental set up adopted in our study we refer the reader to. Schematic illustrations of the Na₂ energy levels for the cascade configuration discussed here are displayed in the two panels of Fig. Case a) illustrates a situation in which the absolute value of the ratio $k_1/k_2$ of the probe laser wave number to that of the coupling field is smaller than unity. We define this ratio as positive for co-propagating beams and negative for the
FIG. 1: The Na$_2$ cascade excitation scheme for two different probe laser/coupling field wavenumber ratio $x = k_1/k_2$, where $k_1$ is the weak probe laser (thin arrow) and $k_2$ is the strong coupling field (thick arrow). In case (a) this ratio is smaller than 1 and in case (b) it is larger than 1. The energy levels chosen for case a) are: $|1\rangle = X^1\Sigma^+_g(0,19)$, $|2\rangle = A^1\Sigma^+_u(0,20)$, $|3\rangle = 2^1\Pi_g(0,19)$, $|4\rangle = A^1\Sigma^+_u(0,18)$ or $A^1\Sigma^+_u(0,20)$, and $|5\rangle = X^1\Sigma^+_g(3,21)$. Those for case b) are: $|1\rangle = X^1\Sigma^+_g(1,19)$, $|2\rangle = A^1\Sigma^+_u(3,18)$, $|3\rangle = 4^1\Sigma^+_g(0,17)$, $|4\rangle = A^1\Sigma^+_u(1,16)$ and $|5\rangle = X^1\Sigma^+_g(2,19)$. The laser frequencies for transitions $|1\rangle$ to $|2\rangle$ and $|2\rangle$ to $|3\rangle$ are $L_1$ (14647.547 cm$^{-1}$) and $L_2$ (15888.065 cm$^{-1}$) for case a) and $L_1$ (14828.639 cm$^{-1}$) and $L_2$ (13284.554 cm$^{-1}$) for case b). Single channel side fluorescence wavenumbers were 15771.91$\pm$0.3 cm$^{-1}$ and 14167.36$\pm$0.3 cm$^{-1}$ for case a) transitions $|3\rangle$ to $|4\rangle$ and $|2\rangle$ to $|5\rangle$, respectively. These values were 13522.93$\pm$0.3 cm$^{-1}$ and 14672.59$\pm$0.3 cm$^{-1}$ for case b). The lifetimes are 12.2 ns for level $|2\rangle$ in cases a) and b) [15], and 21.0 ns and 12.7 ns for level $|3\rangle$ in cases a) and b) [16, 17, 18, 19].

counter propagating configuration. In case b) the absolute value of the $k_1/k_2$ ratio is greater than one instead. In addition to obeying this constraint on the transition frequencies, the selected energy levels were chosen to allow the generation of the largest Rabi frequency of the coupling field, given the available laser sources [narrowband frequency stabilized dye and Titanium Sapphire lasers (Coherent Autoscan 699-29 and 899-29)]. Intermediate level $|2\rangle$ belongs to the Na$_2$ $A^1\Sigma^+_u$ state and the uppermost level belongs to the $2^1\Pi_g$ state in case a) and to the $4^1\Sigma^+_g$ state in case b), respectively, as indicated in the caption of Fig. 1 where, in addition, we provide the fluorescence detection wavelengths in wavenumber units. With the coupling laser tuned to resonance, we scanned the probe laser and detected fluorescence from level $|2\rangle$ and from level $|3\rangle$ by isolating the desired fluorescence from the background radiation with a Spex 1404 double monochromator used as a narrow band filter. In both
FIG. 2: Observation of EIT by fluorescence detection from the intermediate level $|2\rangle$ as a function of the probe laser detuning for counter-propagating beams with a) $|k_1/k_2| = 0.922$ and $\Delta_2 = 0$ MHz and b) $|k_1/k_2| = 1.116$ and $\Delta_2 = 60$ MHz. The Doppler width is 1.1GHz corresponding to 625K sample temperature. The dotted line represents the simulations.

In cases a) and b) the fluorescence from the intermediate level $|2\rangle$ displayed the EIT pattern expected in an open molecular system [12] as illustrated in Figs. 2 a) and b). However, the AT splitting, which is generally present in the fluorescence spectrum from level $|3\rangle$ for a sufficiently large coupling field Rabi frequency, is quite sensitive to the wavenumber ratio $x = k_1/k_2$. In case a) when the absolute value of this ratio is smaller than unity, we observed a clear AT splitting, as shown in Fig. 3a, whereas in case b) with $|k_1/k_2| > 1$, we did not observe evidence of this effect at all (see Fig. 3b). The Rabi frequencies of the coupling field were 400MHz and 530MHz in case a) and b), respectively. Thus case b) should be more favorable for the observation of AT splitting with regard to this parameter.

We simulated the experimental data by using stationary solutions of the density matrix equations of motion as shown in Figs. 2 and 3. Our Rabi frequencies were 6 MHz and 400
MHz for the probe and coupling fields, respectively, for case a) and 36 MHz and 530 MHz for case b). These values were determined from the calculated transition dipole moment matrix elements, Franck Condon factors for known molecular potentials and the measured E field amplitude in the interaction region. Our results show that the presence (or absence) of a splitting in the fluorescence spectrum of the upper level \( |3\rangle \) depends in a sensitive way on the wavenumber ratio of the probe and coupling field lasers. As we explain below, the origin of this result is the Doppler broadening.

In order to gain physical insight into the behavior of the system we used the perturbative model discussed by Qi et al. in Ref. for an open molecular cascade system. We have investigated the conditions under which the fluorescence spectrum from level \( |3\rangle \) displays the experimentally observed shown behavior in Figs. 3a. and 3b. The calculated Doppler averaged fluorescence spectrum from the upper level \( |3\rangle \) (summed over the magnetic sublevels of the \( |2\rangle \rightarrow |3\rangle \) transition), as a function of the detunings \( \Delta_1 = \omega_{21} - \omega_1 + k_1 v_z \) and \( \Delta_2 = \omega_{32} - \omega_2 + k_2 v_z \) of the probe and coupling fields, respectively, is given by

\[
I_3(\Delta_1, \Delta_2) = \sum_M \int_{-\infty}^{+\infty} \rho_{33}^M(v_z) N(v_z) dv_z ,
\]

(1)

where \( z \) is the axis of propagation of the laser beams, \( v_z \) is the z-component of the thermal velocity of the molecules, \( N(v_z) \) is the Gaussian velocity distribution, and \( \rho_{33} \) is the analytical expression for the population of level \( |3\rangle \), given in Ref. 12. We were able to carry out the integration of the Doppler integral analytically. However, the details of this calculation with the full result will be reported elsewhere, because of the complexity of the mathematical expressions and the limited space of the current manuscript. Based on this calculation the intensity of the fluorescence spectrum \( I_3(\Delta_1, \Delta_2) \) from level \( |3\rangle \) is proportional to the following factors:

\[
I_3(\Delta_1, \Delta_2) \propto \frac{w(z_i)}{z_1 - z_2} ,
\]

(2)

where \( w(z_i) \) is the Faddeeva function and \( z_1 \) and \( z_2 \) are the roots of the denominator of \( \rho_{33}(\Delta_1, \Delta_2) \) from Ref. 12. Our analysis indicates that the behavior of \( I_3 \) stems from the two factors, \( w(z_i) \) and \( (z_1 - z_2)^{-1} \).

Based on the analytically calculated \( I_3(\Delta_1, \Delta_2) \) we have derived expressions for the threshold Rabi frequency \( \Omega_{\text{Th}} \) as a function of the ratio \( k_1/k_2 \) above which AT splitting can be experimentally observed. For this we have used the condition for change in the curvature of
FIG. 3: Level $|3\rangle$ fluorescence under the presence of a strong coupling field for: a) $k_1/k_2 = -0.922$ and b) $k_1/k_2 = -1.116$. The dotted line represents the simulations.

the $I_3(\Delta_1, \Delta_2)$ at zero detuning of the probe laser:

$$\left\{ \frac{\partial^2(\Delta_1, \ldots, \Omega_2^{M-1}, \Omega_2^M, \Omega_2^{M+1}, \ldots, x)}{\partial \Delta_1^2} \right\}_{\Delta_1=0} = 0 \quad (3)$$

The results are summarized in Fig. 4 which shows that region II is most favorable for the observation of AT splitting with the smallest required coupling field strength. This corresponds to a configuration in which the probe and coupling laser beams counter-propagate and $|k_1/k_2|$ is smaller than unity. The dots in the figure identify the values of the $k_1/k_2$ ratio that were used in our experiments. We have also investigated the dependence of this threshold $\Omega_T^2$ on the inhomogeneous Doppler linewidth $\Delta \nu_D$. The 3 dimensional plot of Fig. 5 shows a simulation with the same parameters as Fig. 4 where the 3rd dimension is represented by $\Delta \nu_D$. The threshold Rabi frequency $\Omega_T^2$ grows very rapidly with increasing Doppler linewidth in the region $k_1/k_2 < -1$ and $k_1/k_2 > 0$ and has no $\Delta \nu_D$ dependence at all in region $-1 < k_1/k_2 < 0$. This result is unexpected, and quite interesting. It shows that
FIG. 4: Threshold Rabi frequency $\Omega^T_2$ of the coupling field, as a function of the ratio $k_1/k_2$. AT splitting can be observed only in region II. The $\Omega^T_2$ value is much smaller for $-1 < k_1/k_2 < 0$ than for $-1 > k_1/k_2$ or $k_1/k_2 > 0$ (co-propagating geometry). The two dots indicate the $k_1/k_2$ ratio of our experiments.

FIG. 5: Calculated threshold Rabi frequency $\Omega^T_2$ as a function of the wavenumber ratio $x = k_1/k_2$ and the Doppler linewidth $\Delta \nu_D$. $\Omega^T_2$ does not depend on $\Delta \nu_D$ in the interval $-1 < k_1/k_2 < 0$, in contrast there is very strong dependence in $-1 > k_1/k_2$ or $k_1/k_2 > 0$.

only when the two beams counter propagate, and the coupling laser has a larger wavenumber $k_2$ than the probe laser $k_1$, AT splitting of the same magnitude can be observed independently of the molecular/atomic weight or the temperature of the system. In this region only a moderate coupling field Rabi frequency is required to split the experimental Optical Optical Double Resonance (OODR) spectrum and to observe the AT effect independent of the
Doppler linewidth $\Delta \nu_D$. In order to explain this behavior we have to consider the expression for $I_3(\Delta_1, \Delta_2)$ in Eq. 2. First for $-1 < x < 0$ our analysis shows that the splitting is mainly determined by:

$$\frac{1}{z_1 - z_2} = \frac{2x(1 + x)[(\Delta_1 - i\Gamma)^2 + x(1 + x)\Omega_2^2]^\frac{1}{2}}{[(\Delta_1^2 - \Gamma^2 + x(1 + x)\Omega_2^2)^2 + 4\Gamma^2\Delta_1^2]^\frac{1}{2}}$$  \hspace{1cm} (4)

where $\Gamma$ is defined as $\Gamma = \gamma_{12}(1 + x) - \gamma_{13}x$.

This expression is independent of $\Delta \nu_D$. Therefore the threshold $\Omega_2^T$ becomes independent of the Doppler linewidth. This has an important physical implication in that the AT splitting will be equally well resolved independent of the magnitude of the Doppler broadening $\Delta \nu_D$. On the contrary, in the region where the ratio is $x < -1$ or $x > 0$ the AT splitting and the threshold value of $\Omega_2^T$ are determined by

$$w(z_i) \propto e^{-z_i^2} \propto e^{-\frac{f(\Omega_2, x, \gamma_{ij})}{(\Delta \nu_D)^2}}$$  \hspace{1cm} (5)

for sufficiently large $|z_i|$, indicating a strong dependence on $\Delta \nu_D$. Thus the increase in the value of $\Delta \nu_D$ leads to a rapid increase in the threshold Rabi frequency $\Omega_2^T$. This explains why the experimental spectrum in Fig. 4 does not show AT splitting, since our available coupling Rabi frequency was 300~350MHz, while the required value is of the order of a GHz, for this particular experiment.

In conclusion, our experimental results show profoundly different behavior of the AT splitting in the spectra from the upper level in a Doppler broadened open molecular cascade system, while the fluorescence spectrum emanating from the intermediate level is almost unaffected. We have shown that this unexpected behavior is based on the presence of Doppler broadening and that it is strongly dependent on the ratio of the probe and coupling laser wavenumbers. By analyzing the dependence of the threshold Rabi frequency $\Omega_2^T$ on the probe laser-coupling field wavenumber ratio and the Doppler linewidth $\Delta \nu_D$ we have provided an overview of the optimal conditions for observing AT splitting in this excitation scheme.

**Acknowledgments**

This work was supported by National Science Foundation Awards PHY 0245311 and PHY 0216187. We gratefully acknowledge valuable discussions with Prof. Lorenzo M. Narducci.
of Drexel University.

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