Pump induced Autler-Townes effect and A-T mixing in four level atoms

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Abstract

It is shown by theoretical simulation that tuning of the pump power can induce mixing and crossing of Autler-Townes (A-T) components of closely spaced transitions in atoms. Pump radiation also leads to small shifts of the central hole of A-T doublet. Off-resonance pumping gives an asymmetry in the A-T components and by controlling pump frequency detuning it is also possible to mix the A-T components.
I Introduction:

The interaction of a strong electromagnetic field with two, three and four-level atomic systems leads to many interesting nonlinear effects like electromagnetically induced transparency (EIT) [1,2], lasing without inversion (LWI) [3,4] and enhancement or suppression of atomic refractive index [5,6,7]. The strong pump field leads to the dynamic (ac) Stark splitting or Autler-Townes (A-T) splitting [8], in this case the system is said to be dressed by the strong pump field. This energy level splitting is well resolved when the Rabi-frequency of the pump field is larger than the atomic decay rates. Detailed studies on the splitting have been carried out by many researchers both experimentally and theoretically [9-13]. In the weak probe limit this well known nonlinear effect has drawn much attention in recent years after successful achievement of EIT and LWI in various atomic vapor systems [2,10,14-15].

Interaction of atomic systems having closely spaced energy levels with electromagnetic radiations leads to interesting effect on the line shape of the observed spectra [16-19]. Schlössberg and Javan [16] first studied the interaction of a three level system (TLS) having two closely spaced upper levels with two incident radiation fields with two closely spaced monochromatic frequencies separated by an amount larger than the natural line widths of the atomic resonances. Double resonance line shape arising from the interaction of a four-level atomic system with two closely spaced intermediate levels under the influence of a fixed pump frequency has been studied [17]. Almost all of the theoretical and experimental work on A-T effect deal with the three level systems (TLS) and in a TLS two peaks of the A-T doublet occur at probe detunings $\delta_1 = \frac{\delta_2}{2} \pm \frac{1}{2} \sqrt{\delta_2^2 + 4|\chi|^2}$ [10] where $\delta_2$ and $\chi$ are the pump detuning and pump Rabi-frequency respectively. If pump frequency is on-resonance ($\delta_2 = 0$) the two symmetric peaks appear at $\delta_1 = \pm \chi$ with identical linewidths. For non-zero detuning, the peaks are asymmetric and one peak has larger linewidth, while the other has smaller linewidth. This change of linewidth occurs in such a way that the sum of the linewidths is equal to the unperturbed linewidth of the A-T components [10].

Hyperfine transitions in alkaline atoms like Rubidium are widely used for laser cooling [20]. Rb-D$_2$ hyperfine transitions of both the isotopes involve a single lower level and three closely spaced upper levels. Recently we reported [21] a theoretical simulation based on rate equations of a four level atom with a common ground level and three closely spaced upper levels interacting with a standing wave. They were solved numerically to show the difficulty of isolating the components in Lamb dip spectroscopy. Effect of pump Rabi frequency and detuning on A-T components of atomic system with three closely spaced upper levels, commonly found in the hyperfine splitting of alkali atoms, has not been studied theoretically earlier. Here we present a perturbative analytical treatment of such a system with the pump frequency held near resonance with one of the hyperfine transitions ($F_g = 2 \rightarrow F_e = 1$ for $^{85}$Rb-D$_2$). It leads to Autler-Townes splitting of the two closely spaced probe transitions ($F_g = 2 \rightarrow F_e = 2$ and 3). It is found that tuning of the pump power
At higher pump power the mixing of the A-T components leads to a significant outward shift in the holes created by the overlap of the A-T doublets. Effect of off-resonance pumping on the line shape of the A-T components is also discussed.

II Theory:

The four-level system (Fig.1) with energy levels $E_1$, $E_2$, $E_3$ and $E_4$ interacts with two coherent monochromatic radiation fields of frequencies $\Omega_1$ and $\Omega_2$. The strong pump frequency $\Omega_1$ is nearly equal to transition frequency $\omega_1 = \frac{(E_2 - E_1)}{\hbar}$, and the field $\Omega_2$ probes the transitions $1 \rightarrow 3$ and $1 \rightarrow 4$ with the transition frequencies $\omega_2 = \frac{(E_3 - E_1)}{\hbar}$ and $\omega_3 = \frac{(E_4 - E_1)}{\hbar}$ respectively.

The Hamiltonian of the system can be written as

\[ H = H_0 + H_I \]  

where $H_0$ is the unperturbed atomic Hamiltonian with the eigenvalues $E_1$, $E_2$, $E_3$ and $E_4$.

$H_I$ is the interacting Hamiltonian of the four-level system and can be written as

\[ H_I = -2\varepsilon_1 \mu \cos \Omega_1 t - 2\varepsilon_2 \mu \cos \Omega_2 t \]  

where the $\varepsilon_1$ and $\varepsilon_2$ are the electric-field amplitudes of the pump and probe radiations respectively and $\mu$ is the transition dipole moment. In a semiclassical treatment for the atom-field interaction we define the Rabi-frequencies as $\chi_1 = \frac{2\mu \varepsilon_1}{\hbar}$, $\chi_2 = \frac{2\mu \varepsilon_2}{\hbar}$ and $\chi_3 = \frac{2\mu \varepsilon_3}{\hbar}$.

The time evolution of the density matrix is given by [22,23],

\[ i\hbar \frac{\partial \rho}{\partial t} = [H, \rho] + \text{(relaxation terms)} \]  

The detunings are defined as $\Delta \omega_1 = (\Omega_1 - \omega_1)$, $\Delta \omega_2 = (\Omega_2 - \omega_2)$ and $\Delta \omega_3 = (\Omega_2 - \omega_3)$ and the energy level differences are $\Delta_1 = \frac{(E_3 - E_2)}{\hbar}$, $\Delta_2 = \frac{(E_1 - E_2)}{\hbar}$ and $\Delta_3 = \frac{(E_4 - E_3)}{\hbar}$. We introduce $T_1$, $T_2$ as the longitudinal and transverse relaxation constants. Under the effect of the applied field a macroscopic polarization will be developed in the system, which can be written as

\[ P = N[\mu_{21} \rho_{12} \exp(i\Omega_1 t) + \mu_{31} \rho_{13} \exp(i\Omega_2 t) + \mu_{41} \rho_{14} \exp(i\Omega_2 t) + c.c]. \]  

where $N$ is the number of atoms per unit volume. This allows us to introduce the real and imaginary parts of the pump and probe polarizations associated with the three allowed transitions, given by $P_{pr} + iP_{pi} = N\mu_{21} \rho_{12}$, $P_{sr} + iP_{si} = N\mu_{31} \rho_{13}$ and $P_{sr} + iP_{si} = N\mu_{41} \rho_{14}$. The population differences are $\Delta N_1 = N(\rho_{11} - \rho_{22})$, $\Delta N_2 = N(\rho_{11} - \rho_{33})$, and $\Delta N_3 = N(\rho_{11} - \rho_{44})$. They relax to $\Delta N_1^\circ$, $\Delta N_2^\circ$ and $\Delta N_3^\circ$ with relaxation time $T_1$. The transitions $2 \leftrightarrow 3$, $2 \leftrightarrow 4$ and $3 \leftrightarrow 4$ are not induced by any radiation but $\rho_{23}$, $\rho_{24}$ and $\rho_{34}$ are not equal to zero. This will involve the non-linear terms given by $P_{nr} + iP_{ni} = N\rho_{23} \mu_{31}$, $P_{mr} + iP_{mi} = N\mu_{41} \mu_{12} \rho_{24}$ and $P_{qr} + iP_{qi} = N\mu_{41} \mu_{13} \rho_{34}$.
Fifteen steady-state optical Bloch equations can be written in terms of polarizations, population differences and the non-linear parameters.

**III Line Shape of the Probe transitions:**

The analytical expression of the total probe polarization, is the sum of $P_{s1}$ and $P_{s2}$. The analytical closed forms of the $P_{s1}$ and $P_{s2}$ are very complicated, and to get a simplified form we consider the relaxation parameters $T_1 = T_2 = T$ and probe Rabi-frequencies $\chi_2 = \chi_3 = \chi_p$. Since pump electric field amplitude is much greater than the probe electric field amplitude; i.e., $\varepsilon_1 >> \varepsilon_2$ we get $\chi_1 >> \chi_p$.

The probe polarization in zeroth order is

$$P_{\text{probe}} = -\left[ \frac{C_1}{1 + \Delta \omega_2^2 T^2 + \frac{\chi_1^2 T^2}{4}} \left( 1 - \frac{(2\Delta \omega_2 - \Delta \omega_1)^2 T}{\frac{4}{T} + (\Delta \omega_1 - \Delta \omega_2)^2 T + \frac{2\chi_1^2}{T}} \right) + \frac{C_2}{1 + \Delta \omega_3^2 T^2 + \frac{\chi_1^2 T^2}{4}} \left( 1 - \frac{(2\Delta \omega_3 - \Delta \omega_1)^2 T}{\frac{4}{T} + (\Delta \omega_1 - \Delta \omega_3)^2 T + \frac{2\chi_1^2}{T}} \right) \right]$$  \hspace{1cm} (5)

The parameters $C_1$ and $C_2$ are proportional to $\Delta N_2^0 \mu_{13}$ and $\Delta N_3^0 \mu_{14}$ respectively. The first and second terms of eq.(5) correspond to the Lorentzian line shapes of $1 \rightarrow 3$ and $1 \rightarrow 4$ transitions respectively.

When pump frequency is on-resonance with the transition $1 \rightarrow 2$ having transition frequency $\omega_1$, the corresponding detuning is equal to zero (i.e. $\Delta \omega_1 = 0$). In that condition the first term of the eq.(5) becomes

$$P_{\text{probe}}(\Delta \omega_2) = -\left[ \frac{C_1}{1 + \Delta \omega_2^2 T^2 + \frac{\chi_1^2 T^2}{4}} \left( 1 - \frac{4\Delta \omega_2^2 T}{\frac{4}{T} + \frac{4\chi_1^2}{T} + \frac{2\chi_1^2}{T}} \right) \right]$$  \hspace{1cm} (6)

Eq.(6) gives a Lorentzian line shape centered at $\Delta \omega_2 = 0$, when we increase the power of pump-beam it is split into two symmetrical components which are commonly known as A-T doublet [8]. Similar result will be obtained from the second term of eq.(5). The separation of the A-T components increases with increase in the value of $\chi_1 T$. They are not observed at lower value of $\chi_1 T$.

To see the effect of probe power on absorptive line we also present here the expression of $P_{\text{probe}}$ after retaining up to second order term of the probe Rabi-frequencies $\chi_p$

$$P_{\text{probe}} = \left[ \frac{C_1}{1 + \Delta \omega_2^2 T^2 + \frac{\chi_1^2 T^2}{4}} \left( 1 - \frac{4\Delta \omega_2^2 T}{\frac{4}{T} + \frac{4\chi_1^2}{T} + \frac{2\chi_1^2}{T}} \right) \right]$$  \hspace{1cm} (7)
Where the above parameters $a_1$, $b_1$, $c_1$, $d_1$ and $e_1$ are defined as

$$a_1 = 1 + T^2 \left[ \chi_p^2 + \frac{1}{4} (\chi_1^2 + \chi_p^2) + \Delta \omega_2^2 \right]$$

$$b_1 = \frac{(2\Delta \omega_2 - \Delta \omega_3)(2\Delta \omega_2 - \Delta \omega_1)\chi_1^2\chi_p^2 T^4}{8\left[ \frac{1}{T} + (\Delta \omega_1 - \Delta \omega_2)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2) \right]\left[ \frac{1}{T} + (\Delta \omega_2 - \Delta \omega_3)^2 T + \frac{T}{2}\chi_p^2 \right]}$$

$$c_1 = \frac{(2\Delta \omega_2 - \Delta \omega_1)^2 \chi_1^2 T^3}{4\left[ \frac{1}{T} + (\Delta \omega_1 - \Delta \omega_2)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2) \right]}$$

$$d_1 = \frac{(2\Delta \omega_2 - \Delta \omega_3)^2 \chi_p^2 T^3}{4\left[ \frac{1}{T} + (\Delta \omega_2 - \Delta \omega_3)^2 T + \frac{T}{2}\chi_p^2 \right]}$$

$$e_1 = \frac{\chi_1^2 \chi_p^2 T^4 \left[ \frac{3}{16} + \frac{(2\Delta \omega_2 - \Delta \omega_1)(2\Delta \omega_2 - \Delta \omega_3)T}{\frac{1}{T} + (\Delta \omega_1 - \Delta \omega_2)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2)} \right]^2}{16 \left[ \frac{1}{T} + (\chi_1^2 + \chi_p^2)^2 T^4 - \frac{T^2}{4} \left( \frac{\chi_p^2(2\Delta \omega_1 - \Delta \omega_3)^2}{\frac{1}{T} + (\Delta \omega_1 - \Delta \omega_3)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2)} + \frac{\chi_p^2(2\Delta \omega_1 - \Delta \omega_2)^2}{\frac{1}{T} + (\Delta \omega_1 - \Delta \omega_2)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2)} \right) \right]}$$

Similarly $a_2$, $b_2$, $c_2$, $d_2$ and $e_2$ are

$$a_2 = 1 + T^2 \left[ \chi_p^2 + \frac{1}{4} (\chi_1^2 + \chi_p^2) + \Delta \omega_3^2 \right]$$

$$b_2 = \frac{(2\Delta \omega_3 - \Delta \omega_1)(2\Delta \omega_3 - \Delta \omega_2)\chi_1^2\chi_p^2 T^4}{8\left[ \frac{1}{T} + (\Delta \omega_1 - \Delta \omega_3)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2) \right]\left[ \frac{1}{T} + (\Delta \omega_2 - \Delta \omega_3)^2 T + \frac{T}{2}\chi_p^2 \right]}$$

$$c_2 = \frac{(2\Delta \omega_3 - \Delta \omega_1)^2 \chi_1^2 T^3}{4\left[ \frac{1}{T} + (\Delta \omega_1 - \Delta \omega_3)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2) \right]}$$

$$d_2 = \frac{(2\Delta \omega_3 - \Delta \omega_2)^2 \chi_p^2 T^3}{4\left[ \frac{1}{T} + (\Delta \omega_2 - \Delta \omega_3)^2 T + \frac{T}{2}\chi_p^2 \right]}$$

$$e_2 = \frac{\chi_1^2 \chi_p^2 T^4 \left[ \frac{3}{16} + \frac{(2\Delta \omega_1 - \Delta \omega_1)(2\Delta \omega_3 - \Delta \omega_2)T}{\frac{1}{T} + (\Delta \omega_1 - \Delta \omega_3)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2)} \right]^2}{16 \left[ \frac{1}{T} + (\chi_1^2 + \chi_p^2)^2 T^4 - \frac{T^2}{4} \left( \frac{\chi_p^2(2\Delta \omega_1 - \Delta \omega_3)^2}{\frac{1}{T} + (\Delta \omega_1 - \Delta \omega_3)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2)} + \frac{\chi_p^2(2\Delta \omega_1 - \Delta \omega_2)^2}{\frac{1}{T} + (\Delta \omega_1 - \Delta \omega_2)^2 T + \frac{T}{4}(\chi_1^2 + \chi_p^2)} \right) \right]}$$
IV Computation of line shape in Rubidium atom:

For the purpose of numerical calculations we have chosen a four-level system that corresponding to $^{85}\text{Rb}-D_2$ hyperfine transitions (Fig. 1). The lower energy level 1 corresponds to the $F_g = 2$ hyperfine level in $^5S_{1/2}$ ground state of $^{85}\text{Rb}-D_2$, while the upper levels 2, 3 and 4 are the $F_e = 1, 2, 3$ components of the $^5P_{3/2}$ excited state. The energy level differences in the upper states of $^{85}\text{Rb}$ are $\Delta_1 = 0.029\text{GHz}$, $\Delta_2 = 0.092\text{GHz}$ and $\Delta_3 = 0.063\text{GHz}$ respectively. The value of the pump frequency used in this work is $384615.38\text{GHz} (\lambda \approx 780.0\text{nm})$. The values of pump Rabi-frequencies used for computation are very close to the experimental situation of $\text{Rb}$ vapor system [11,24]. We choose the value of relaxation constants $\frac{1}{\tau} = \Gamma = 0.01\text{GHz}$ [11].

Fig. 2A shows the probe polarization vs. detuning $\left(\Omega_2 - \frac{(\omega_2 + \omega_3)}{2}\right)$ curve, for two different pump powers when the pump frequency is held on-resonance with the transition $1 \rightarrow 2$. The pump field Rabi-frequencies used in Fig. 2A are 0.01 and 0.04 $\text{GHz}$. At low pump Rabi-frequency two absorption lines appear corresponding to $1 \rightarrow 3$ and $1 \rightarrow 4$ transitions (Fig. 2A(a)) when the probe is tuned across the lines. As we increase the pump Rabi-frequency keeping $\Gamma$ constant the two absorption lines become power broadened and each line is split into two A-T components (Fig. 2A(b)) denoted as (A$_1$, A$_2$) and (B$_1$, B$_2$).

It is observed that at a pump Rabi-frequency of 0.06$\text{GHz}$ the nearby split components of two A-T doublets corresponding to the transitions $1 \rightarrow 3$ and $1 \rightarrow 4$ are mixed with each other (A$_2$ and B$_1$ of Fig. 2B). This mixing occurs at a frequency $\frac{(\omega_2 + \omega_3)}{2}$ and forms a single stronger absorption line. Thus one can obtain three A-T components instead of four (Fig. 2B(a)). In order to find the effect of the finite probe power the probe polarization(eq.7) is plotted in Fig. 2B(b), the values of $\chi_1$ and $\Gamma$ remain unchanged; value of $\chi_p$ is 0.006$\text{GHz}$. The effect of the finite probe power is revealed as a small change of line shape.

In Fig. 2C we see the effect of further higher pump powers on the absorption line. At pump Rabi-frequency 0.1$\text{GHz}$ the central line of Fig. 2B is again split and they cross the zero detuning line (Fig. 2C(a)). With further increase in pump power the separation between two A-T components increases but they never mix again. As a consequence positions of the holes produced by the A-T splitting also shifts with pump power, this is observed after they cross the zero probe detuning.

In Fig. 3 we present two spectra (A,B) where we show the effect of off-resonance pumping on the probe absorption. In both the spectra (A,B) dotted curve(a) shows the probe absorption when $\Delta\omega_1 = 0$ and $\chi_1 = 0.03\text{GHz}$ and it leads to the two A-T doublets corresponding to two allowed transitions. The solid curve(b) of Fig. 3A plotted with $\chi_1 = 0.03$ and $\Delta\omega_1 = -0.012\text{GHz}$ gives four A-T peaks with asymmetric linewidths because of the non-zero detuning of the pump field. A-T components A$_1$ and B$_1$ are weaker and have lower linewidth than A$_2$ and B$_2$. We can control the pump detuning in such a way that at a value of -0.026$\text{GHz}$ the A-T component(B$_1$)
of $1 \rightarrow 4$ transition nearly mixes with the A-T component $(A_2)$ of the $1 \rightarrow 3$ transition (Fig. 3B) solid curve (b). So by controlling the pump detuning (i.e. coherent control) we are able to mix the nearby A-T component keeping pump power fixed. In both the cases mentioned above the sum of the linewidths of two A-T components is equal to the unperturbed linewidth [10] for each transition.

V Conclusions:

The power-broadened A-T doublets in a four-level system interacting with a near resonant strong pump and a weak probe field have been investigated by analytical solution of the optical Bloch equations. Numerical computation of the line shape of $^{85}$Rb-D$_2$ transitions is reported. In a four level system two A-T doublets are generally expected in the presence of a pump. Our calculation of the A-T splitting in a four-level system with three closely spaced upper levels reveals that the pump power can be tuned so that the nearby A-T components of two nearby transitions mix with each other. Such mixing of the A-T components may also be achieved by varying detuning of the pump frequency. Further increase of pump power will cause the mixed components to split again and they cross to the other sides of the mean frequency resulting in four components. It is noteworthy that the positions of the holes of the A-T doublets shifts with increase of pump Rabi-frequency. This results from increased A-T splitting. So one can manipulate the atomic response through pump field either by pump power or by pump detuning. We also performed the computation with another $^{85}$Rb-D$_2$ transitions (i.e. $F_g = 3 \rightarrow F_e = 2, 3, 4$) where the upper state hyperfine energy level spacings are higher than those of the $^{85}$Rb-D$_2$ transitions corresponding to $F_g = 2 \rightarrow F_e = 1, 2, 3$. In this case much higher pump power is required for the level mixing.

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Figure Captions

Figure 1: Energy level diagram of a four-level atom interacting with two radiation fields. Inset: the Energy-level of $^{85}\text{Rb} - D_2$ transitions.

Figure 2A: Probe polarization $P_{probe}$ vs. $\left( \Omega_2 - \frac{\omega_2 + \omega_3}{2} \right)$ curve for (a) $\chi_1 = 0.01$ and (b) $\chi_1 = 0.04 \text{ GHz}$. The pump field is on-resonance with $1 \rightarrow 2$ level. The parameters used in calculations are $\Gamma = 0.01 \text{ GHz}$ and $\omega_1 = 384615.38 \text{ GHz}$

Figure 2B: Probe polarization $P_{probe}$ vs. $\left( \Omega_2 - \frac{\omega_2 + \omega_3}{2} \right)$ curve. (a) $\chi_1 = 0.06 \text{ GHz}$. The curve(b) based on eq.(7) uses $\chi_1 = 0.06$ and $\chi_p = 0.006 \text{ GHz}$. Other parameters remain the same as in Fig. 2A.

Figure 2C: Probe polarization $P_{probe}$ vs. $\left( \Omega_2 - \frac{\omega_2 + \omega_3}{2} \right)$ curve for (a) $\chi_1 = 0.1$ and (b) $\chi_1 = 0.12 \text{ GHz}$. Other parameters remain the same as in Fig. 2A.

Figure 3: Probe polarization $P_{probe}$ vs. $\left( \Omega_2 - \frac{\omega_2 + \omega_3}{2} \right)$ curve for figure (A) and (B) the dotted curve (a) $\chi_1 = 0.03 \text{ GHz}$ and $\Delta\omega_1 = 0$. In figure (A) for (b) $\chi_1 = 0.03$ and $\Delta\omega_1 = -0.012 \text{ GHz}$. In figure (B) for (b) $\chi_1 = 0.03$ and $\Delta\omega_1 = -0.026 \text{ GHz}$. Other parameters remain the same as in Fig. 2A.
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