

## Control of amplification without inversion in $H_2$ and LiH molecules: Dependence on relative magnitude of probe and coherent field Rabi frequencies in three-level $\Lambda$ system

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**Abstract.** Dependence of amplification without inversion (AWI) on the relative strength of probe and coherent field Rabi frequencies has been studied in  $H_2$  and LiH molecules for three-level  $\Lambda$  configuration. We have derived exact analytical expressions for coherences and populations keeping all the orders of probe field Rabi frequency ( $G$ ) and coherent field Rabi frequency ( $\Omega$ ) in the steady state limit. Previously, first-order approximation (i.e. keeping only the first-order term in  $G$ ) was used and hence AWI was studied for the condition  $\Omega \gg G$ . Here, by using the exact analytical expressions of coherences and populations, we have shown that AWI is maximum when  $\Omega$  is within the same order of probe field Rabi frequency  $G$  irrespective of the choice of different ro-vibrational transitions in both the molecules. However, the shape of the gain profile and the maximum value of gain on the probe field and the absorption on coherent field depend on the choice of different ro-vibrational levels as the upper lasing levels. Effect of bidirectional pumping, homogeneous and inhomogeneous broadening on AWI process has been studied. By solving the density matrix equations numerically it has been shown that both the transient and the steady state AWI can be obtained and the numerical values of coherences and populations at large time are in very good agreement with exact analytical values in the steady state limit. It has been shown that in molecules AWI can be obtained on probe field of smaller wavelength than that of the coherent field which has not been observed in atoms so far.

**Keywords.** Amplification without inversion;  $H_2$  molecule; LiH molecule;  $\Lambda$  system.

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### 1. Introduction

Amplification without inversion (AWI) and lasing without inversion (LWI) have been studied both theoretically [1–5] and experimentally [6,7] in three, four and multilevel systems by choosing different configurations, e.g. ladder,  $V$ ,  $\Lambda$  etc. Physical picture of AWI has also been analyzed in dressed state approach [8–11]. In most

of the theoretical studies, analytical expressions for coherences and populations in the steady state limit were derived by using first-order approximation, i.e. keeping only the first-order terms in probe field Rabi frequency ( $G$ ) and all orders in coherent field Rabi frequency ( $\Omega$ ). Hence the calculations were done for  $\Omega \gg G$ , within the validity of first-order approximation. In this work we have derived the exact expressions for populations and coherences in the steady state limit keeping all orders of both the probe field and coherent field Rabi frequencies and used these expressions to study the dependence of AWI on relative strength of  $\Omega$  and  $G$ . It is found that when  $\Omega$  and  $G$  are within the same order (e.g.  $\Omega \sim 2G, \Omega \sim 3G$ ), AWI is maximum and the values of gain differ significantly from those calculated under first-order approximation. We found that the values of populations and coherences obtained by solving the density matrix equations numerically at large time are in good agreement with the values obtained from the exact expressions in the steady state limit. However, results obtained with first-order approximation agree with the numerical results only when  $\Omega \gg G$ .

AWI were studied theoretically either by choosing different parameters for the process [12,13] or in real systems like atoms [14,15] and molecules [3–5,16–18]. The presence of rotational and vibrational states makes the study of LWI/AWI fascinating in molecules as these states influence the process of inversionless lasing. Choice of different ro-vibrational levels in the transition scheme results in variations in the gain profile of a molecule. Also, gain can be obtained in a wide range of frequency covering these vibrational and rotational states. Thus in a molecular system, one has the flexibility to choose from a wide frequency range as well as various shapes of the gain profiles. In atomic systems, the two upper levels and two lower levels respectively in the V and  $\Lambda$  schemes are generally taken to be hyperfine levels. In some studies, for example in the Rb experiment by Zibrov *et al* in 1995 [19], fine-structure components of an atomic level were considered. These levels being very closely spaced, very narrow bandwidth lasers are needed in order to single out the individual levels. In a molecular system, two widely spaced vibrational levels can be used and hence this restriction is not required, as long as coherence can be maintained. However, the lasers used should have bandwidth short enough to avoid resonant excitation from other vibrational levels. Earlier, our group has studied AWI in  $H_2$  molecule using resolvent operator technique [3–5], in which the dependence of gain on the choice of different vibrational levels was also shown considering the effect of two-photon near-resonant transitions to autoionizing states (the upper lasing level). In a recent calculation [20] on three-level ladder, V and  $\Lambda$  system in  $H_2$  and  $Li_2$  molecules we have shown that AWI can be obtained in VUV, and in violet region respectively from the first excited Rydberg state when two of the levels are coupled by a strong coherent field. We have also shown that AWI can be controlled by choosing different vibrational levels for coherent and probe transitions. Recently we have studied the feasibility of amplification without population inversion (AWI) in LiH molecule for three-level ladder, V and  $\Lambda$  schemes [21]. However, in these studies the effect of bidirectional pumping on the AWI process has not been considered. Moreover, in all these studies the Rabi frequency of probe  $G$  has been chosen much less than the Rabi frequency of coupling laser  $\Omega$  ( $G \ll \Omega$ ). Hence the laser intensity for coherent coupling was orders of magnitude greater than that for the probe field. Sometimes it becomes difficult to

get such a high-intensity laser in particular for the generation in the VUV range. In a preliminary calculation, we have shown that AWI is maximum when  $\Omega$  is within the same order of  $G$  (e.g.  $\Omega \sim 2G, \Omega \sim 3G$ ) for  $H_2$  molecule [22]. It has also been found that for the transitions considered in the previous calculation the absorption on the coherent field is maximum and orders of magnitude greater than AWI on the probe field. In the present work we have shown that by choosing different ro-vibrational transitions one can control the absorption on the coherent field to become orders of magnitude less than the AWI on the probe. Moreover, we have shown that similar conclusion can be drawn for AWI (in violet region) in LiH molecule in the presence of unidirectional and bidirectional pumping, and the magnitude of gain is different from that in  $H_2$  molecule. The essence of this work is that one can get the most efficient AWI in VUV and violet region by using coherent field laser intensity of the order of  $1 \text{ W/cm}^2$  or much less than the value under the Doppler free condition, keeping the absorption on the coherent field minimum. Here we have solved the density matrix equations of the  $\Lambda$  system numerically using *ab-initio* data for the potential energy curves and dipole transition moments of  $H_2$  [23,24] and LiH [25] and compared the values of matrix elements at large time with exact and approximate values [13,26–28] of coherences and populations in the steady state limit.

It has been shown here that under resonant condition the restriction on the relative values of the spontaneous decay width to get AWI (i.e. the spontaneous decay width on the probe field transition ( $\gamma_{13}$ ) should be less than the spontaneous decay width on the coherent field transition ( $\gamma_{12}$ )) in the presence of bidirectional pumping [13,26], is waived when the unidirectional pumping is considered [20,22]. However, under off-resonant condition AWI is feasible in both the cases (bidirectional and unidirectional pumping) if the detuning is less than a critical value (see eqs (16) and (17)).

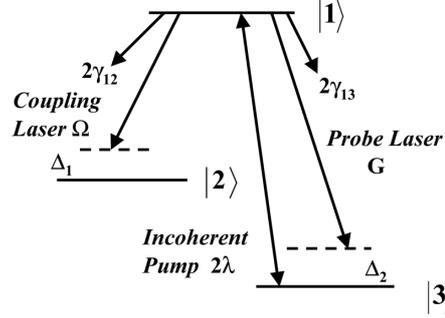
## 2. Theory

We consider a closed  $\Lambda$ -type three-level system with the ground state  $|3\rangle$  and two excited states  $|2\rangle$  and  $|1\rangle$ , as shown in figure 1. The transition  $|2\rangle \leftrightarrow |1\rangle$  of energy difference  $\hbar\omega_{12}$  is driven by a coherent coupling laser of frequency  $\omega_L$  with Rabi frequency  $\Omega$ . The transition  $|3\rangle \leftrightarrow |1\rangle$  of energy difference  $\hbar\omega_{13}$  is pumped with a rate  $2\lambda$  by an incoherent field.  $2\gamma_{13}$  ( $2\gamma_{12}$ ) is the spontaneous decay width from the state  $|1\rangle$  to the state  $|3\rangle$  ( $|2\rangle$ ). There is no dipole allowed coupling between the states  $|2\rangle$  and  $|3\rangle$ . A probe laser of frequency  $\omega_p$  with Rabi frequency  $G$  is applied to the transition  $|1\rangle \leftrightarrow |3\rangle$ .  $\Omega$  and  $G$  are chosen to be real. The Rabi frequencies for the coupling and probe fields respectively are given by

$$\Omega = d_c \cdot E_c / \hbar, \quad (1a)$$

$$G = d_p \cdot E_p / \hbar, \quad (1b)$$

where  $E_c$  and  $E_p$  are the electric field for the coupling and probe fields respectively while  $d_c$  and  $d_p$  are the dipole transition moments for the respective transitions. The detunings between the field and the system frequencies are given by



**Figure 1.** Schematic diagram for  $\Lambda$  transition scheme. The probe field of frequency  $\omega_p$  and the strong field of frequency  $\omega_L$  have been applied between levels  $|1\rangle \rightarrow |3\rangle$  and  $|1\rangle \rightarrow |2\rangle$  respectively. The level  $|1\rangle$  is pumped by the incoherent pumping field of rate  $2\lambda$  from level  $|3\rangle$ .  $2\gamma_{13}$  ( $2\gamma_{12}$ ) is the spontaneous decay width from the state  $|1\rangle$  to the state  $|3\rangle$  ( $|2\rangle$ ).  $\Delta_1$  and  $\Delta_2$  are the detunings for strong coupling laser and probe laser respectively.

$$\Delta_1 = \omega_{23} - \omega_L \tag{2a}$$

$$\Delta_2 = \omega_{13} - \omega_p. \tag{2b}$$

We obtain the density matrix equations from the master equation as

$$\dot{\rho}_{11} = i\Omega(\rho_{21} - \rho_{12}) + iG(\rho_{31} - \rho_{13}) - 2(\gamma_{12} + \gamma_{13} + \Lambda)\rho_{11} + 2\lambda\rho_{33} \tag{3a}$$

$$\dot{\rho}_{22} = i\Omega(\rho_{12} - \rho_{21}) + 2\gamma_{12}\rho_{11} \tag{3b}$$

$$\dot{\rho}_{33} = iG(\rho_{13} - \rho_{31}) + 2(\gamma_{13} + \lambda)\rho_{11} - 2\lambda\rho_{33} \tag{3c}$$

$$\dot{\rho}_{12} = i\Omega(\rho_{22} - \rho_{11}) + iG\rho_{32} - (\gamma_{13} + \gamma_{12} + \lambda + i\Delta_1)\rho_{12} \tag{3d}$$

$$\dot{\rho}_{13} = iG(\rho_{33} - \rho_{11}) + i\Omega\rho_{23} - (\gamma_{12} + \gamma_{13} + 2\lambda + i\Delta_2)\rho_{13} \tag{3e}$$

$$\dot{\rho}_{23} = i\Omega\rho_{13} - iG\rho_{21} + [i(\Delta_1 - \Delta_2) - \lambda]\rho_{23}. \tag{3f}$$

The closure relation for the system is  $\rho_{11} + \rho_{22} + \rho_{33} = 1$ . The gain-absorption coefficient for the probe laser coupled to the transition  $|1\rangle \leftrightarrow |3\rangle$  ( $|1\rangle \leftrightarrow |2\rangle$ ) is proportional to  $\text{Im}(\rho_{13})$  [ $\text{Im}(\rho_{12})$ ]. If  $\text{Im}(\rho_{13}) < 0$ , the probe laser will be amplified. Similarly, if  $\text{Im}(\rho_{12}) < 0$ , the coupling laser will be amplified.

We have analytically solved the above density matrix equations in the steady state limit without any approximation, i.e. keeping all the terms. When  $\Omega \sim G$ , one cannot neglect the terms containing  $G^2$ . Thus, to give the exact analytical solution of  $\text{Im}(\rho_{13})$ , we have considered all the terms, i.e. all the orders of  $G$  and  $\Omega$  and the analytical expression of  $\text{Im}(\rho_{13})$  is given as

$$v_{13} = \frac{[A_1 + \Omega^2 G^3 c \lambda (b + \lambda)] \lambda b}{R_1}. \quad (4)$$

Here

$$R_1 = (D_1 + G^2 \gamma_{12} b) C_1 + [G(\Delta_1 - \Delta_2) b + G \Delta_1 \lambda] P, \quad (5a)$$

$$A_1 = \Omega^4 G c b \lambda + \Omega^2 G \lambda^2 b c (b + \lambda) + \Omega^2 G b (\lambda \gamma_{12} + b \gamma_{13} + \lambda \gamma_{13}) \\ \times (\Delta_1 - \Delta_2)^2 + \Omega^2 G \lambda^2 \gamma_{12} \Delta_1^2 - 2 \Omega^2 G \lambda^2 \gamma_{12} \Delta_1 \Delta_2, \quad (5b)$$

where

$$C_1 = B_1 \lambda b + G^2 b (b + \lambda) (\Delta_1 - \Delta_2)^2 + \Omega^2 G^2 \lambda b + G^2 \lambda (b + \lambda) \\ + G^2 \lambda^2 b (b + \lambda) \quad (6a)$$

$$P = \Omega^2 G \Delta_2 c \lambda b - \Omega^2 G c b (b + \lambda) (\Delta_1 - \Delta_2) \\ - Q \gamma_{12} [G(\Delta_1 - \Delta_2) b + G \Delta_1 \lambda] \quad (6b)$$

$$D_1 = \lambda \gamma_{12} b^2 + (3\lambda + \gamma_{13}) \Omega^2 b + \Delta_1^2 \lambda \gamma_{12} \quad (6c)$$

$$B_1 = [\Omega^2 + (\Delta_1 - \Delta_2) \Delta_2]^2 + (b + \lambda)^2 (\Delta_1 - \Delta_2)^2 + 2 \Omega^2 \lambda (b + \lambda) \\ + \lambda^2 (b + \lambda)^2 + \lambda^2 \Delta_2^2 \quad (6d)$$

$$Q = \lambda (b + \lambda)^2 + G^2 (b + \lambda) + \Omega^2 (b + \lambda) + \Delta_2^2 \lambda \quad (6e)$$

$$c = \gamma_{13} - \gamma_{12} \quad (6f)$$

$$b = \gamma_{13} + \gamma_{12} + \lambda. \quad (6g)$$

The imaginary part of  $\rho_{12}$  is calculated as

$$v_{12} = \frac{\Omega b \lambda \gamma_{12} C_1}{R_1}. \quad (7)$$

The imaginary part of  $\rho_{23}$  is given by

$$v_{23} = \frac{\Omega P \lambda b}{R_1}. \quad (8)$$

The general expression of the steady state populations are given by

$$\rho_{11} = \frac{\Omega v_{12}}{\gamma_{12}} \quad (9)$$

$$\rho_{33} = [(\gamma_{13} + \lambda)\rho_{11} - G v_{13}]/\lambda \quad (10)$$

$$\rho_{22} = 1 - \rho_{11} - \rho_{33}. \quad (11)$$

With a strong coupling laser  $\Omega \gg \gamma_{ij}, \lambda, G$ , we can approximate the above steady state solutions considering all orders of  $\Omega$  and only first order of  $G$ . The solutions are given by

$$v_{13}^{(1)} = \frac{A_1}{B_1 D_1} \quad (12)$$

$$v_{12}^{(1)} = \frac{\Omega \lambda \gamma_{12} b}{D_1} \quad (13)$$

$$v_{23}^{(1)} = \frac{\Omega P}{B_1 D_1}. \quad (14)$$

When  $\Delta_1 = \Delta_2 = \Delta$ , eq. (12) reduces to

$$v_{13}^{(1)} = \frac{\Omega^4 G c b \lambda + \Omega^2 G \lambda^2 b c (b + \lambda) - \Omega^2 G \lambda^2 \gamma_{12} \Delta^2}{[(\Omega^2 + \lambda b + \lambda^2)^2 + \lambda^2 \Delta^2][\lambda \gamma_{12} b^2 + (3\lambda + \gamma_{13})\Omega^2 b + \Delta^2 \lambda \gamma_{12}]}. \quad (15)$$

When  $c$  is negative, i.e.  $\gamma_{13} < \gamma_{12}$ ,  $v_{13}^{(1)} < 0$  the probe will be amplified. But when  $c$  is positive, i.e.  $\gamma_{13} > \gamma_{12}$ , to achieve amplification we have to fulfill the condition

$$\lambda \gamma_{12} \Delta^2 > \Omega^2 c b + \lambda b c (b + \lambda).$$

As  $\Omega \gg \gamma_{ij}, \lambda, G$ ,

$$\Delta > \Omega \left[ \frac{(\gamma_{12} + \gamma_{13} + \lambda)(\gamma_{13} - \gamma_{12})}{\gamma_{12} \lambda} \right]^{1/2}. \quad (16)$$

Setting  $\rho_{22} - \rho_{33} = 0$ , we obtain a critical value of  $\Delta$  as

$$\Delta_c = \Omega \left[ \frac{(\gamma_{12} + \gamma_{13} + \lambda)\gamma_{13}}{\gamma_{12} \lambda} \right]^{1/2}. \quad (17)$$

When  $\Delta < \Delta_c$ , the population distribution satisfies  $\rho_{33} > \rho_{22} > \rho_{11}$ . But when  $\Delta > \Delta_c$ , the population distribution satisfies  $\rho_{22} > \rho_{33} > \rho_{11}$ , and the population inversion for the Raman transition  $|2\rangle \rightarrow |3\rangle$  occurs. The probe amplification occurs

due to the stimulated Raman scattering  $|2\rangle \rightarrow |1\rangle \rightarrow |3\rangle$  in which the molecules absorb a coupling-laser photon and then emit a probe-laser photon.

When  $\Delta_1 = \Delta_2 = 0$ , eq. (12) reduces to

$$v_{13}^{(1)} = \frac{\Omega^4 Gcb\lambda + \Omega^2 G\lambda^2 bc(b + \lambda)}{(\Omega^2 + \lambda b + \lambda^2)^2 [\lambda\gamma_{12}b^2 + (3\lambda + \gamma_{13})\Omega^2 b]}. \quad (18)$$

Thus when  $\Delta_1 = \Delta_2 = 0$  we can get amplification only if  $\gamma_{13} < \gamma_{12}$ . But when the replenishment of the lower lasing level is neglected,  $v_{13} \propto (\gamma_{13} - \lambda - \gamma_{12})$  at resonance. Thus for  $(\gamma_{13} - \lambda) < \gamma_{12}$ , we can get amplification.  $\lambda$  can be greater than or less than  $\gamma_{13}$ . But for  $\lambda > \gamma_{13}$ , the population will be inverted. Therefore,  $\lambda$  should be less than  $\gamma_{13}$  satisfying the condition  $(\gamma_{13} - \lambda) < \gamma_{12}$ . Thus, if we neglect the replenishment of the lower lasing level, we can get amplification for (a)  $\gamma_{12} > \gamma_{13}$  and (b)  $\gamma_{12} < \gamma_{13}$  at the resonant condition ( $\Delta_1 = \Delta_2 = 0$ ).

The real parts of  $\rho_{12}$ ,  $\rho_{13}$ ,  $\rho_{23}$  are given below:

$$\text{Re}(\rho_{12}) = u_{12} = \frac{Gv_{23} + \Delta_1 v_{12}}{(\gamma_{12} + \gamma_{13} + \lambda)}, \quad (19)$$

$$\text{Re}(\rho_{13}) = u_{13} = \frac{\Delta_2 v_{13} - \Omega v_{23}}{(\gamma_{12} + \gamma_{13} + 2\lambda)}, \quad (20)$$

$$\text{Re}(\rho_{23}) = u_{23} = -\frac{(\Delta_1 - \Delta_2)v_{23} + \Omega v_{13} + G v_{12}}{\lambda}. \quad (21)$$

With a resonant coupling laser and a resonant probe laser ( $\Delta_1 = \Delta_2 = 0$ ), we have found that  $\rho_{13}(t) = i \text{Im}[\rho_{13}(t)]$ ,  $\rho_{12}(t) = i \text{Im}[\rho_{12}(t)]$  and  $\rho_{23}(t) = \text{Re}[\rho_{23}(t)]$ . Thus the dispersive response for the probe and coupling laser vanishes, and the two-photon coherence  $\rho_{23}$  is real.

*Gain coefficient:* The gain coefficient for a three-level system is given as

$$\mathbf{G}' = \frac{-4\pi n\omega_p |d_p|^2}{\hbar c G} \text{Im}(\rho_p), \quad (22)$$

where  $n$  is the number density of molecules,  $\omega_p$  is the frequency of the probe field,  $d_p$  is the dipole transition moment for the probe transition and  $\rho_p$  is the corresponding coherence term.

*Absorption coefficient:* The absorption coefficient for the coherent field is given by

$$\mathbf{A} = \frac{-4\pi n\omega_L |d_c|^2}{\hbar c \Omega} \text{Im}(\rho_c), \quad (23)$$

where  $\omega_L$  is the frequency of the coherent field,  $d_c$  is the dipole transition moment for the coherent transition and  $\rho_c$  is the corresponding coherence term.

*Doppler broadening:* The Doppler width (FWHM) of a line of frequency  $\omega$  is given as

$$\Delta\omega = \frac{2\omega}{c} \sqrt{2 \ln 2 \frac{kT}{m}}, \quad (24)$$

where  $k$  is the Boltzmann constant,  $c$  is the velocity of light,  $T$  is the temperature and  $m$  is the mass of the molecule.

### 3. Schemes

#### • H<sub>2</sub> molecule

$X^1\Sigma_g^+(v=0, j=0) \rightarrow B^1\Sigma_u^+(v=2, 8, j=1)$  transition is coupled by the probe field and  $X^1\Sigma_g^+(v=1, j=0) \rightarrow B^1\Sigma_u^+(v=2, 8, j=1)$  transition is coupled by the coupling field.

For the first case, i.e when  $B^1\Sigma_u^+(v=2, j=1)$  is the upper lasing level,  $\gamma_{13} < \gamma_{12}$ . In the second case, when  $B^1\Sigma_u^+(v=8, j=1)$  is the upper lasing level,  $\gamma_{13} > \gamma_{12}$ .

In the first case, the coherent laser couples the  $B^1\Sigma_u^+(v=2, j=1)$  state and  $v=1, j=0$  level of ground state and the probe and pump field act between  $B^1\Sigma_u^+(v=2, j=1)$  state and the  $v=0, j=0$  level of ground state. The frequencies of the two transitions are  $88750.664 \text{ cm}^{-1}$  and  $92893.167 \text{ cm}^{-1}$  respectively. Now let us consider the effect of further transitions from other ro-vibrational levels which will be populated due to spontaneous decay from the excited levels, on the amplification process. It is well-known that under normal condition the population in  $v=0$  level of the ground state of a molecule is maximum and it decays exponentially with the increase in vibrational quantum number. The higher vibrational levels of the ground state will be populated due to spontaneous decay from these excited levels ( $v=1$  level of  $B^1\Sigma_u^+$  state). But in the large time limit these molecules will decay to ground state due to collisional and vibrational relaxations. Within the lifetime of an excited vibrational level, say  $v=1$  level of  $X^1\Sigma_g^+$  state (which is closest to  $v=0$  level), the fraction of the molecules lying in this level may be further excited by the probe and coherent fields to higher vibrational levels of  $B^1\Sigma_u^+$  state, e.g. coherent field will excite the molecule between  $v=1$  and 2 levels and probe field will excite the molecule between  $v=4$  and 5 levels and the amplification process will be repeated. But since this will be a second-order process and since the frequencies are far away from the exact resonance from these levels, effect of this second-order process will be of orders of magnitude less than the first-order process (i.e. the initial excitation from the  $v=0$  level of the ground state).

In H<sub>2</sub> molecules, the paramolecules (with nuclear spin  $I=0$ ) will occupy the states with even total angular momentum quantum number ( $j=0, 2, 4$  etc.) and the orthomolecules (with  $I=1$ ) will occupy the states with odd angular momentum quantum numbers. By using molecular jet one can get most of the populations in  $j=0$  and 1 levels. The energy difference between  $v=0, j=0$  of ground state of H<sub>2</sub> molecule and  $v=2, j=1$  of  $B^1\Sigma_u^+$  state is  $92893.167 \text{ cm}^{-1}$ . One can selectively excite  $v=0, j=0$  level of the ground state by choosing a probe laser of this transition frequency. A simple calculation can show that if  $v=0, j=1$  of the ground state is excited by a photon of this energy, it will reach very close to the  $v=1, j=3$  level of  $B^1\Sigma_u^+$  state (this is a forbidden transition) and it will be detuned from the  $j=2$  and  $j=0$  levels by approximately  $46.812 \text{ cm}^{-1}$  and  $153.674 \text{ cm}^{-1}$ . Again from the  $B^1\Sigma_u^+(v=1, j=1)$  level molecules may de-excite to  $v=1, j=0$  and  $v=1, j=2$  levels of the ground state. The energy difference between  $v=1, j=1$  level of  $B^1\Sigma_u^+$  state of H<sub>2</sub> molecule and  $v=1, j=0$  level of ground state is  $88750.664 \text{ cm}^{-1}$ . Thus by choosing the coherent coupling laser of this frequency, one can set the  $X^1\Sigma_g^+(v=1, j=0) \rightarrow B^1\Sigma_u^+(v=1, j=1)$  transition to be a resonant transition. The  $X^1\Sigma_g^+(v=1, j=2) \rightarrow B^1\Sigma_u^+(v=1, j=1)$  transition

will be detuned by approximately  $337.594 \text{ cm}^{-1}$ . The frequency of the coherent laser is much smaller than that for the probe laser. Therefore, if the coherent coupling laser excites the molecules from the  $v = 0, j = 0$  level of the ground state, it will be a detuned transition from  $B^1\Sigma_u^+(v = 0, j = 1)$  level by approximately  $1547.760 \text{ cm}^{-1}$ . Again if the coherent coupling laser excite the molecules from the  $v = 0, j = 1$  level of the ground state, it will be a detuned transition from  $B^1\Sigma_u^+(v = 0, j = 0)$  level and  $B^1\Sigma_u^+(v = 0, j = 2)$  level by approximately  $1380.488 \text{ cm}^{-1}$  and  $1497.346 \text{ cm}^{-1}$  respectively. Since the probability for the detuned transition is much weaker than the resonant transition, this excitation channel can be ignored.

• **LiH molecule**

$X^1\Sigma^+(v = 0, j = 0) \rightarrow A^1\Sigma^+(v = 0, 1, j = 1)$  is coupled by the probe field and  $X^1\Sigma^+(v = 1, j = 0) \rightarrow A^1\Sigma^+(v = 0, 1, j = 1)$  transition is coupled by the strong field.

In both the cases  $\gamma_{13} < \gamma_{12}$ . Here,  $v$  is the vibrational quantum number and  $j$  is the total angular momentum quantum number for the system.

To single out these particular vibrational levels for excitation, the frequencies of the lasers should be carefully chosen so as to avoid resonant excitation from other vibrational levels of the ground state to different vibrational levels of the excited state as discussed previously [21].

**4. Results and discussions**

For the three-level  $\Lambda$  scheme, the following aspects have been studied for H<sub>2</sub> and LiH molecules: (a) Dependence of AWI on probe field and absorption on coherent field on the relative strength of respective Rabi frequencies, (b) dependence of gain and absorption on the choice of different ro-vibrational transition in both the molecules, (c) influence of bidirectional and unidirectional pumping and (d) the effect of total decay width and Doppler broadening on AWI. Due to space restrictions we have not presented all the features as figures, we have instead discussed these features in this section.

*4.1 Dependence of gain and absorption on Rabi frequencies for the closed system*

In the course of deriving the exact analytical solutions of density matrix equations in the steady state limit we got the impression that when  $G$  and  $\Omega$  are of the same order, the amplification on the probe field and the absorption on the coherent field will be strongly affected. Hence, instead of choosing only  $\Omega \gg G$  (first-order approximation), we have studied the AWI process for different combinations of  $\Omega$  and  $G$ . First of all we have chosen  $\Omega = K\gamma_{13}$  where  $K = 1$  to  $4$  and we have varied the value of  $G$ . It is obvious that for  $\Omega = \gamma_{13}$ , AWI is absent. AWI starts from the value of  $\Omega$  slightly greater than  $2\gamma_{13}$  and the magnitude of gain decreases with the increase in the value of  $\Omega$  for a particular value of  $G$  at resonance for both the fields (table 1). But at a particular value of  $\Omega$  as the value of  $G$  is increased, the gain

Table 1. Dependence of gain on the choice of coherent field Rabi frequency ( $\Omega$ ) at resonance for two transition schemes of  $H_2$  molecule considering unidirectional pumping. Note:  $N(-a) = N \times 10^{-a}$ .

$v$	$\Omega/G$	$\Omega/\gamma_{13}$	Gain
2	3	2.21	0.105(-10)
		3.00	0.997(-11)
		4.00	0.719(-11)
8	2	2.21	0.155(-12)
		3.00	0.966(-13)
		4.00	0.198(-13)

increases and for the first transition scheme maximum gain is obtained for the value of  $G = \Omega/3$  (see table 2). If the value of  $G$  is further increased, the amplification occurs from Raman inversion. Similar feature is obtained for the second transition scheme, but the value of gain is maximum for  $G = \Omega/2$ . In table 2 we have given the values of exact and approximate (first-order approximation) gain on the probe field and the exact values of absorption on the coherent field as a function of  $G$  for  $\Omega = 2.21\gamma_{13}$  at resonance for two schemes in  $H_2$  molecule. By comparing the values of gain in columns 5 and 6, it is clear that the approximate values of gain deviate significantly from the exact values for  $G \sim \Omega/3$  and  $G \sim \Omega/2$  for the first and the second transition scheme respectively in  $H_2$  molecule. The reason for getting maximum gain at the resonance condition at this particular value of  $G$  and  $\Omega$  is that for this system the destructive interference between two absorption channels becomes maximum, leading to net emission from the molecule excited by the incoherent pumping. It is to be noticed here that for the first transition scheme ( $v = 2$  of  $B^1\Sigma_u^+$  state) of  $H_2$  molecule at  $\Omega \sim 3G$  the absorption on the coherent field is six times greater than the gain on the probe field. The reason for strong absorption is the high value of dipole transition moment for the coherent transition. But for the second transition scheme ( $v = 8$  of  $B^1\Sigma_u^+$  state), as the dipole moment for the coherent transition is much less than the dipole moment for the probe transition, the absorption is almost one order of magnitude smaller than the gain. Thus by choosing different ro-vibrational levels as the upper lasing level one can also control the relative strength of amplification and absorption.

The energy difference between ground vibrational level ( $v = 0, j = 0$ ) of  $X^1\Sigma_g^+$  state and the  $v = 2, 8 (j = 1)$  of  $B^1\Sigma_u^+$  state are  $92893.167 \text{ cm}^{-1}$  and  $99887.161 \text{ cm}^{-1}$  respectively which correspond to the wavelength  $105.86 \text{ nm}$  and  $100.53 \text{ nm}$  respectively. Therefore in  $H_2$  molecule, AWI can be obtained in VUV spectral region. It is found that for  $\Omega \sim 2.21\gamma_{13}$  and  $G = \Omega/3$  the intensities of lasers used as probe and coherent field are  $0.6$  and  $1.5 \text{ W/cm}^2$  respectively for the first transition scheme. One can obtain the intensity in the above-mentioned range by focusing a pulse of energy  $10 \text{ nJ}$  and duration  $\sim 1 \mu\text{s}$  into a focal area of  $1 \text{ mm}^2$ . Generation of these low-intensity VUV radiation is possible in laboratory [29]. In our previous work on AWI in  $H_2$  [20], the intensity of laser for coherent coupling

**Table 2.** Gain of the probe field and absorption of the coherent field (at resonance) in the steady state limit for closed three-level  $\Lambda$  configuration in  $H_2$  molecule for two transition schemes considering unidirectional pumping. All the quantities are expressed in atomic unit. To convert the gain and frequency in  $cm^{-1}$ , one has to multiply the tabulated values by  $1.889 \times 10^8$  and  $2.19475 \times 10^5$  respectively.

$v$	$G$	$\Omega(= 2.21\gamma_{13})$	$\lambda$	Gain		Absorption
				Exact	Approx.	(Exact)
2	0.002(-8)	0.18(-8)	0.06(-10)	0.084(-11)	0.084(-11)	-0.048(-10)
	0.006(-8)	0.18(-8)	0.18(-10)	0.254(-11)	0.255(-11)	-0.136(-10)
	0.019(-8)	0.18(-8)	0.66(-10)	0.704(-11)	0.728(-11)	-0.419(-10)
	0.024(-8)	0.18(-8)	0.93(-10)	0.863(-11)	0.909(-11)	-0.485(-10)
	0.059(-8)	0.18(-8)	2.20(-10)	1.092(-11)	1.323(-11)	-0.711(-10)
8	0.109(-8)	0.449(-8)	0.198(-8)	0.089(-12)	0.089(-12)	-0.116(-13)
	0.154(-8)	0.449(-8)	0.199(-8)	0.112(-12)	0.121(-12)	-0.137(-13)
	0.218(-8)	0.449(-8)	0.200(-8)	0.155(-12)	0.182(-12)	-0.148(-13)

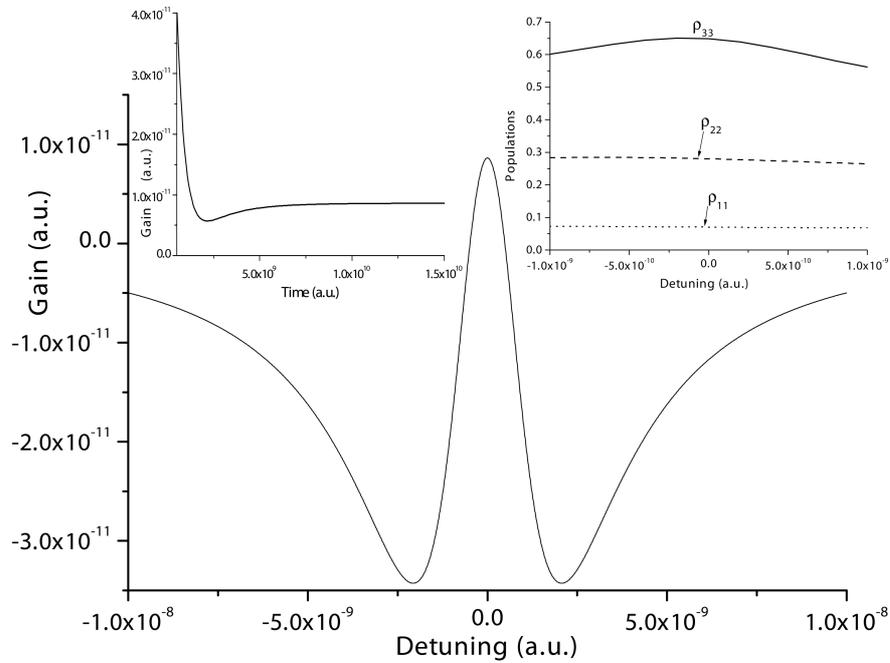
was two to three orders of magnitude greater and the probe field intensity was several orders of magnitude less than those used here. We found that when the strength of these two coupling lasers are almost comparable, the coherence of the system increases and gives rise to more efficient amplification.

It is to be noted that the values of gain and absorption which are given in tables 2 and 3, are obtained at the resonant condition of the coupling lasers. But for the first transition scheme at  $\Omega \sim 2.21\gamma_{13}$  and  $G = \Omega/3$  (see table 2), under detuned condition, amplification occurs from the stimulated Raman transition only. Therefore, we have plotted (in figure 2) the gain profile for first transition scheme in  $H_2$  molecule (keeping  $\Delta_1 = 0$ ) in the steady state limit, considering the unidirectional pumping of the ground state ( $X^1\Sigma_g^+$  ( $v = 0, j = 0$ )) for  $\Omega \approx 2.21\gamma_{13}$  and  $G = \Omega/10$  (see table 2). The right inset shows the steady state population distribution vs. probe laser detuning ( $\Delta_2$ ) at the above-mentioned values of  $\Omega$  and  $G$  when the unidirectional pumping of the ground state is considered. These curves show that gain can be obtained without population inversion since  $\rho_{33} > \rho_{22} > \rho_{11}$ . The left inset shows the time variation of numerically obtained value of gain at the peak position of the gain profile (i.e. at  $\Delta_1 = \Delta_2 = 0$ ), when the unidirectional pumping of the ground state is considered. This numerical value of gain at large time (shown in the left inset) agrees with the analytical value of gain at the resonance condition of the two coupling lasers in the steady state shown in the main figure.

In figure 3 we have plotted gain profile as a function of  $\Delta_2$  for the second transition scheme considering unidirectional pumping of the ground state when  $\Omega \approx 2.21\gamma_{13}$  and  $G = \Omega/2$  at the resonance condition of the two coupling lasers. The steady state population distribution vs. probe laser detuning ( $\Delta_2$ ) graph in the right inset at the above-mentioned values of  $\Omega$  and  $G$  indicate amplification without inversion in the steady state limit. The time variation of numerically obtained value of gain at the peak position of the gain profile (i.e. at  $\Delta_1 = \Delta_2 = 0$ ) has been shown in the left inset. From the figures and the tables it is to be noticed that the value of  $\Omega$

**Table 3.** Gain of the probe field and absorption of the coherent field in steady state for closed  $\Lambda$  system at the resonant condition of the coherent and probe lasers for the first scheme of  $H_2$  molecule (where  $B^1\Sigma_u^+(v=2, j=1)$  is the upper lasing level), considering the replenishment of the ground state, i.e. considering bidirectional pumping. All the quantities are expressed in atomic unit.

$v$	$G$	$\Omega(= 2.21\gamma_{13})$	$\lambda$	Gain		Absorption (Exact)
				Exact	Approx.	
2	0.002(-8)	0.18(-8)	0.06(-10)	0.083(-11)	0.083(-11)	-0.044(-10)
	0.006(-8)	0.18(-8)	0.18(-10)	0.246(-11)	0.247(-11)	-0.133(-10)
	0.019(-8)	0.18(-8)	0.73(-10)	0.681(-11)	0.703(-11)	-0.392(-10)
	0.024(-8)	0.18(-8)	0.98(-10)	0.787(-11)	0.826(-11)	-0.470(-10)
	0.059(-8)	0.18(-8)	2.70(-10)	0.901(-11)	1.083(-11)	-0.710(-10)



**Figure 2.** Gain profile for  $\Lambda$ -scheme in  $H_2$  molecule when  $B^1\Sigma_u^+(v=2, j=1)$  is the upper lasing level considering unidirectional pump (without replenishment of ground state). Here  $\gamma_{12} = 2.681 \times 10^{-9}$  a.u.,  $\gamma_{13} = 8.263 \times 10^{-10}$  a.u.,  $\lambda = 0.93 \times 10^{-10}$  a.u., the intensity for probe field =  $0.6 \text{ W/cm}^2$ , the intensity for the coherent field =  $1.5 \text{ W/cm}^2$ . In this calculation, the detuning of the strong field, i.e.  $\Delta_1 = 0$ ,  $\tau = 1 \times 10^{13}$  a.u. The right inset shows the steady state population distribution vs.  $\Delta_2$  curve. The left inset shows the time variation of numerically obtained value of gain at the peak position of the gain profile (i.e. at  $\Delta_1 = \Delta_2 = 0$ ).

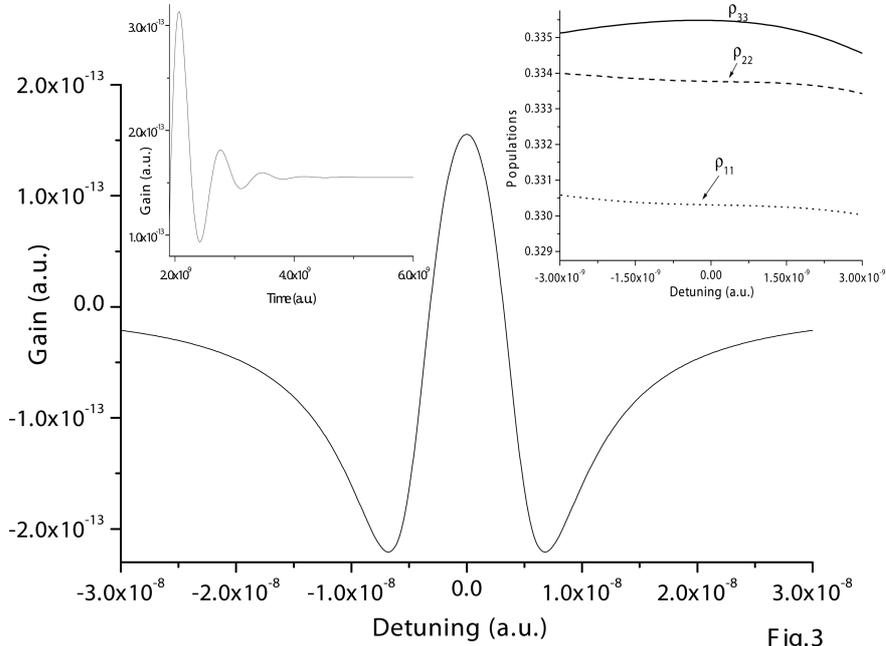


Fig.3

**Figure 3.** Gain profile for  $\Lambda$ -scheme in  $H_2$  molecule when  $B^1\Sigma_u^+(v=8, j=1)$  is the upper lasing level considering unidirectional pump (without replenishment of ground state). Here  $\gamma_{12} = 5.719 \times 10^{-11}$  a.u.,  $\gamma_{13} = 2.027 \times 10^{-9}$  a.u.,  $\lambda = 2 \times 10^{-9}$  a.u., the intensity for probe field =  $4 \text{ W/cm}^2$ , the intensity for the coherent field =  $530 \text{ W/cm}^2$ , the detuning of the strong field, i.e.  $\Delta_1 = 0$  and  $\tau = 1 \times 10^{13}$  a.u. The right inset shows the steady state population distribution vs.  $\Delta_2$  curve. The left inset shows the time variation of numerically obtained value of gain at the peak position of the gain profile (i.e. at  $\Delta_1 = \Delta_2 = 0$ ).

and hence the intensity of the coherent field should be chosen such that AC stark splitting is greater than the value of  $\gamma_{13}$ .

In table 3 we have given the results of gain and absorption in  $H_2$  molecule for the first transition scheme considering the bidirectional pumping for  $\Omega = 2.21\gamma_{13}$ . Comparing the results in tables 2 and 3, it is found that in the presence of bidirectional pumping both the absorption and amplification are damped for the same set of  $\Omega$  and  $G$ . For the results shown in table 3, since  $\lambda$  is much less than  $\gamma_{13}$ , the effect of damping is small. But it can be shown that when  $\lambda \geq \gamma_{13}$ , effect of bidirectional pumping will be prominent. Since for the second transition scheme ( $B^1\Sigma_u^+(v=8, j=1)$  is the upper lasing level)  $\gamma_{13} > \gamma_{12}$ , one can get gain only under off-resonant condition for both the fields in the presence of bidirectional pumping. To get AWI at  $\Delta_1 = \Delta_2$ , one has to satisfy the condition given in eq. (16) as well as the value of detuning should be less than a critical value (see eq. (17)). But for this particular ro-vibrational scheme as  $\gamma_{13} \gg \gamma_{12}$ , it is difficult to satisfy these two conditions (eqs (16) and (17)) simultaneously. If detuning of the two lasers are so chosen that  $\Delta > \Delta_c$ , the Raman inversion takes place and amplification occurs

**Table 4.** Gain of the probe field and absorption of the coherent field in steady state for the closed  $\Lambda$  system at the resonant condition of the coherent and probe lasers for LiH molecule, considering unidirectional pumping of the ground state. All the quantities are expressed in atomic unit.

$v$	$\gamma_{13}$	$G$	$\Omega$	$\lambda$	Gain	Absorption
0	0.095(-11)	0.094(-11)	0.284(-11)	0.022(-11)	0.123(-9)	-0.142(-8)
1	0.499(-11)	0.499(-11)	1.483(-11)	0.149(-11)	0.125(-9)	-0.104(-8)

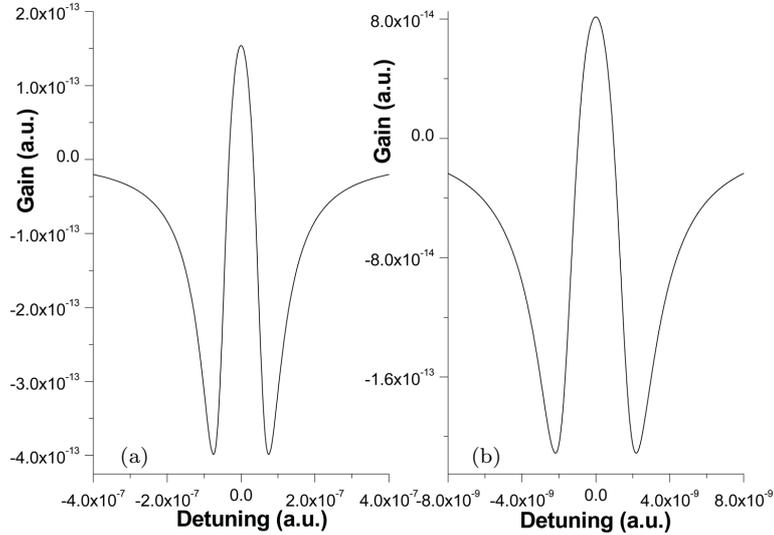
due to Raman transition. Therefore, for closed system AWI cannot be obtained in the presence of bidirectional pumping.

Recently our group has studied the feasibility of amplification without population inversion (AWI) in LiH molecule for three-level ladder, V and  $\Lambda$  schemes [21]. The LiH molecule serves as a good model for studying the dependence of gain on the choice of ro-vibrational levels, as the hyperfine splitting is much smaller than the spacing between two rotational ( $\sim 10^2$  GHz) or two vibrational levels ( $\sim 10^4$  GHz). We have done the calculations on  $\Lambda$  scheme in LiH molecule to show that AWI is more efficient when  $\Omega \approx G$  than when  $\Omega \gg G$ . To demonstrate this feature two vibrational levels  $v = 0, 1$  of  $A^1\Sigma^+(v = 0, 1, j = 1)$  state have been considered as the upper lasing level. In LiH molecule, amplification can be obtained for small values of the intensities of probe and coupling lasers since the decay widths are smaller than those in  $H_2$  molecule. In table 4 for  $v = 0, j = 1$  level of  $A^1\Sigma^+$  state as the upper lasing level the intensities of the probe field and the coupling lasers are  $2.7 \times 10^{-5}$  W/cm<sup>2</sup> and  $3.7 \times 10^{-5}$  W/cm<sup>2</sup> respectively. Similarly for  $v = 1, j = 1$  level of  $A^1\Sigma^+$  state, as the upper lasing level the intensities of the probe field and the coupling lasers are  $1.5 \times 10^{-4}$  W/cm<sup>2</sup> and  $2.5 \times 10^{-4}$  W/cm<sup>2</sup> respectively. Here, by choosing the values of  $\Omega$  and  $G$  close to each other, we have shown that the gain increases almost by four orders of magnitude than the previous values obtained by considering  $\Omega \gg G$  [21]. It can be shown that in the presence of bidirectional pumping, the magnitude of gain and absorption are reduced in LiH molecule as in the case of  $H_2$  molecule.

The main utility of AWI is to generate lasing in shorter wavelength region and we have shown here that AWI in VUV region and violet region is possible in  $H_2$  and LiH molecules respectively with coherent and probe field laser intensities which can be easily generated in the laboratory. Moreover, we have shown here that in these molecules amplification on probe field of shorter wavelength can be achieved using coupling laser of larger wavelength in  $\Lambda$ -transition scheme which has not been observed in atomic systems [30,31].

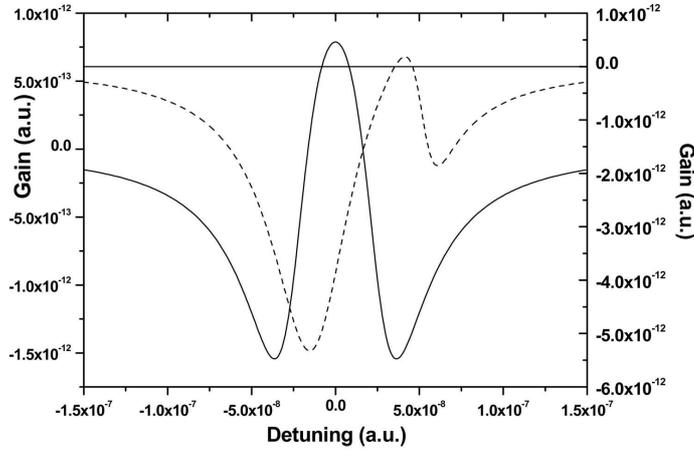
#### 4.2 Gain profile considering total spontaneous decay width

In the calculations described above, a closed system for the molecule is assumed, i.e. it has been assumed that the molecule in the excited electronic state decays spontaneously only to the ground vibrational level, from where it is excited. In



**Figure 4.** (a) Gain profile for  $\Lambda$ -scheme in  $H_2$  molecule when  $B^1\Sigma_u(v=2, j=1)$  is the upper lasing level considering the total spontaneous decay width in place of partial decay width (without replenishment).  $\gamma_{12} = \gamma_{13} = 0.197 \times 10^{-7}$  a.u.,  $\lambda = 0.1205 \times 10^{-7}$  a.u., the intensity for probe field =  $6.5 \times 10^2$  W/cm<sup>2</sup>, the intensity for the coherent field =  $1.6 \times 10^3$  W/cm<sup>2</sup>, the detuning of the strong field, i.e.  $\Delta_1 = 0$  and  $\tau = 1 \times 10^{13}$  a.u. (b) Gain profile for  $\Lambda$ -scheme in  $LiH$  molecule when  $A^1\Sigma^+(v=0, j=1)$  is the upper lasing level considering the total spontaneous decay width in place of partial decay width (without replenishment).  $\gamma_{12} = \gamma_{13} = 0.586 \times 10^{-9}$  a.u.,  $\lambda = 0.1205 \times 10^{-9}$  a.u., the intensity for probe field = 10 W/cm<sup>2</sup>, the intensity for the coherent field = 14 W/cm<sup>2</sup>, the detuning of the strong field, i.e.  $\Delta_1 = 0$  and  $\tau = 1 \times 10^{13}$  a.u.

practice, molecules in the first excited electronic state also decay to the higher vibrational levels of the ground electronic state and this should influence the strength of the gain profile. For example, it was shown by Renzoni *et al* [32] that losses towards levels not excited by the laser field results in reducing the contrast and width of the coherent population trapping. Therefore, we have considered here an open system in the molecule by taking into account spontaneous decay to all the allowed vibrational levels. Thus the  $\gamma$ 's in the density matrix eqs (3) have been replaced by  $\gamma' = \gamma + \gamma_h$ . But the total spontaneous decay width ( $\gamma_h$ ) of a state for an open system are larger than that for a closed system by orders of magnitude. In figure 4a we have presented the gain profile as a function of  $\Delta_2$  (keeping  $\Delta_1 = 0$ ) in the steady state limit, without considering the replenishment of the ground state for the first transition scheme of  $H_2$  molecule, i.e. when  $B^1\Sigma_u^+(v=2, j=1)$  is the upper lasing level. In figure 4b we have presented the gain profile as a function of  $\Delta_2$  (keeping  $\Delta_1 = 0$ ) in the steady state limit, without considering the replenishment of the ground state for  $LiH$  molecule when  $v=0, j=1$  of  $A^1\Sigma^+$  state is the upper lasing level. In our previous work [21], the intensities for the probe field and the coherent field were  $10^{-5}$  W/cm<sup>2</sup> and  $10^3$  W/cm<sup>2</sup> respectively. Here, for  $LiH$



**Figure 5.** Gain profile for  $\Lambda$ -scheme in  $H_2$  molecule when  $B^1\Sigma_u^+(v=8, j=1)$  is the upper lasing level considering the total spontaneous decay width in place of partial decay width. The solid line represents gain profile when unidirectional pumping is considered. Here  $\gamma_{12} = \gamma_{13} = 0.1 \times 10^{-7}$  a.u.,  $\lambda = 0.68 \times 10^{-8}$  a.u., the intensity for probe field  $= 0.85 \times 10^2$  W/cm<sup>2</sup>, the intensity for the coherent field  $= 2 \times 10^4$  W/cm<sup>2</sup>, the detuning of the strong field, i.e.  $\Delta_1 = 0$  and  $\tau = 1 \times 10^{13}$  a.u. The dashed line represents gain profile when bidirectional pumping is considered. The intensities for coherent and probe field and the value of  $\tau$  are kept same. Here  $\lambda = 0.35 \times 10^{-8}$  a.u. and  $\Delta_1 = 0.4 \times 10^{-7}$  a.u.

molecule the intensities for the probe field and the coherent field are 10 W/cm<sup>2</sup> and 14 W/cm<sup>2</sup> respectively, and the gain on the probe field increases by one order of magnitude. Therefore, under Doppler free condition AWI can be obtained in both the molecules when treated as open system and the laser intensities required can be generated in the laboratory with the present day facilities.

The calculation has been repeated considering the replenishment of the ground state, where we get amplification only at the off-resonant condition of the two coupling lasers as both the  $\gamma$ 's are equal. In figure 5 the solid line represents the gain profile as a function of  $\Delta_2$  considering unidirectional pumping (keeping  $\Delta_1 = 0$ ) in the steady state limit for the second transition scheme of  $H_2$  molecule, i.e. when  $B^1\Sigma_u^+(v=8, j=1)$  is the upper lasing level. At the peak position of the gain profile, absorption on the coherent field ( $5.3 \times 10^{-14}$  a.u.) is much less than the gain ( $7.8 \times 10^{-13}$  a.u.) on the probe field. The dashed line in this figure shows the gain profile as a function of  $\Delta_2$  (at  $\Delta_1 = 0.4 \times 10^{-7}$  a.u.) for this transition scheme considering bidirectional pumping. Since in this case (bidirectional pumping) both  $\gamma$ 's are equal, we get amplification only at the detuned condition of both the coupling lasers and get an asymmetric gain profile. When we consider the unidirectional pumping, the gain peak is present around the zero detuning ( $\Delta_2$ ) and the two minima are present around the AC Stark splitted levels. But when we consider the bidirectional pumping, we get AWI only at the off-resonant condition of the two coupling lasers

and the interference nature of gain profile is prominent. In the previous section (for closed system) we have shown that it is difficult to get AWI for the second transition scheme when bidirectional pumping is considered as  $\gamma_{13} \gg \gamma_{12}$ . But if we consider the open system, this difficulty is removed since in this case  $\gamma_{13} = \gamma_{12}$  and here we have chosen the values of  $\Delta$  which are less than the critical value to get AWI (see eq. (17)). In all the schemes, decay widths for an open system are larger than those for a closed system. Hence to establish coherence in the system, intensity of both the fields has been increased.

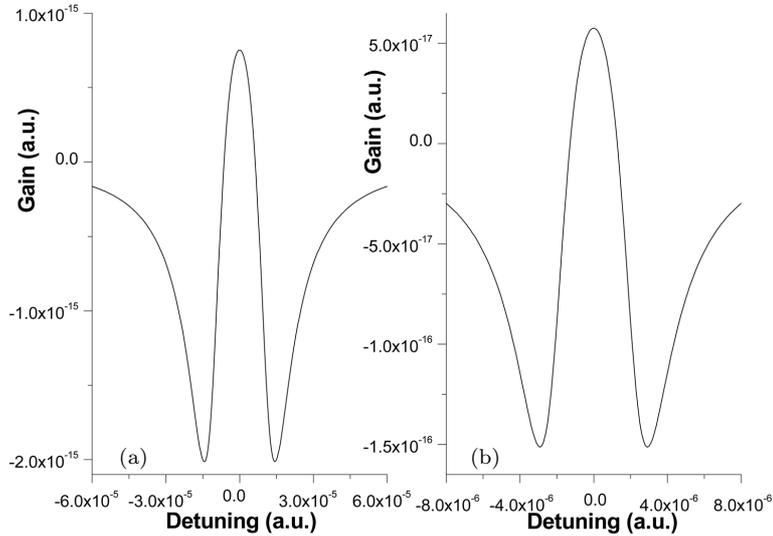
#### 4.3 Gain profile considering Doppler broadening

At room temperature, the Doppler broadening (see eq. (24)) is much greater than the natural broadening. Therefore the total linewidth increases by many orders of magnitude due to Doppler broadening. Thus the  $\gamma$ 's in the density matrix eqs (3) have been replaced by  $\gamma'' = \gamma + \gamma_h + \gamma_{inh}$ . But the Doppler broadening ( $\gamma_{inh}$ ) which arises due to the relative motion of the atoms with respect to the direction of the laser photons, are orders of magnitude larger than that for an open system. In  $H_2$  molecule when  $B^1\Sigma_u^+(v=2, j=1)$  is the upper lasing level, FWHM of Doppler broadening in the probe channel is  $\approx 3.7 \times 10^{-6}$  a.u.  $\approx 24.35$  GHz and that in the coherent coupling channel is  $\approx 3.53 \times 10^{-6}$  a.u.  $\approx 23.23$  GHz. The states involved in the lasing schemes are broadened and hence high intensity of lasers is required to induce coherence ( $\Omega > \gamma_{13}$ ). In this case, this is as high as  $\approx 10^7$  W/cm<sup>2</sup>. To obtain such intensity, a pulse of energy 10 nJ and duration  $\sim 10$  ns should be focused in a focal area of  $10 \mu\text{m}^2$ , i.e. a short pulse with tight focusing is required.

Figure 6a shows the gain profile of the  $H_2$  molecule, for the Doppler broadened  $\Lambda$ -system (when  $B^1\Sigma_u^+(v=2, j=1)$  is the upper lasing level) without considering the replenishment of the ground state. The nature of the profile is similar to the profile in figure 4a where we have considered the total spontaneous decay width, but the magnitude of gain is reduced by two orders of magnitude than that for the open system.

Figure 6b shows the gain for AWI for  $LiH$  molecule considering the Doppler broadening where  $v=0, j=1$  of  $A^1\Sigma^+$  state is the upper lasing level, neglecting the replenishment of the ground state (keeping  $\Delta_1 = 0$ ). Here FWHM of Doppler broadening in the probe channel is  $\approx 0.729 \times 10^{-6}$  a.u.  $\approx 4.802$  GHz and that in the coherent coupling channel is  $\approx 0.769 \times 10^{-6}$  a.u.  $\approx 5.03$  GHz. Choosing  $\Omega$  and  $G$  within the same order, it has been noticed that AWI becomes more efficient and the gain on the probe channel is higher than that obtained in the previous work [21].

The calculation has been repeated considering the replenishment of the ground state, where we get amplification only at the off-resonant condition of the two coupling lasers as  $\gamma_{13} > \gamma_{12}$ . Magnitude of gain is damped in the presence of bidirectional pumping than that for unidirectional pumping as mentioned in the previous section. Due to space restriction all the figures are not shown here. Thus we can conclude that one can get AWI in the steady state region even in the presence of strong Doppler damping.



**Figure 6.** (a) Gain profile for  $\Lambda$ -scheme in  $H_2$  molecule when  $B^1\Sigma_u(v=2, j=1)$  is the upper lasing level considering the Doppler width (without replenishment).  $\gamma_{12} = 0.354 \times 10^{-5}$  a.u.,  $\gamma_{13} = 0.37 \times 10^{-5}$  a.u.,  $\lambda = 0.231 \times 10^{-5}$  a.u., the intensity for probe field =  $2.4 \times 10^7$  W/cm<sup>2</sup>, the intensity for the coherent field =  $6 \times 10^7$  W/cm<sup>2</sup>, the detuning of the strong field, i.e.  $\Delta_1 = 0$  and  $\tau = 1 \times 10^{13}$  a.u. (b) Gain profile for  $\Lambda$ -scheme in LiH molecule when  $A^1\Sigma^+(v=0, j=1)$  is the upper lasing level considering the Doppler width (without replenishment).  $\gamma_{12} = 0.729 \times 10^{-6}$  a.u.,  $\gamma_{13} = 0.769 \times 10^{-6}$  a.u.,  $\lambda = 0.479 \times 10^{-6}$  a.u., the intensity for probe field =  $1.8 \times 10^7$  W/cm<sup>2</sup>, the intensity for the coherent field =  $2.5 \times 10^7$  W/cm<sup>2</sup>, the detuning of the strong field, i.e.  $\Delta_1 = 0$  and  $\tau = 1 \times 10^{13}$  a.u.

We have shown that under Doppler free condition (closed/open) as well as in the presence of Doppler broadening, AWI is feasible in  $H_2$  and LiH molecules and it can be tested in the laboratory. In this calculation, we have used molecular density of  $10^9/\text{cm}^3$  and get the amplification which is comparable to that of the He–Ne laser in the Doppler free situation of  $H_2$  molecule. For LiH molecule this is one order of magnitude higher than that of the He–Ne laser under Doppler free condition. But when we consider the Doppler broadened system, the molecular density should be increased to  $10^{14}/\text{cm}^3$  (pressure is less than 1 Torr), one can get gain ( $\sim 10^{-3} \text{ cm}^{-1}$ ) of the order of that in He–Ne lasers. At that high molecular density, the collisional linewidth is  $\approx 3$  kHz (at room temperature and pressure 3.1 mTorr) which is much less than the Doppler width ( $\approx 24$  GHz). The molecular density for LiH molecule (Doppler broadened) should be increased to  $10^{15}/\text{cm}^3$ . At this situation, the gain will be comparable to that of the He–Ne laser. At that high molecular density, the collisional linewidth is  $\sim 10$  MHz (at room temperature and a pressure of 1 Torr [33]) which is also much less than the Doppler width ( $\sim 5$  GHz). Since the Doppler width is much larger for both the molecules, inclusion of collisional broadening has no effect on the results.

## 5. Conclusion

We have derived the exact expressions of coherences and populations from the density matrix equations for three-level  $\Lambda$ -system in the steady state limit keeping all the orders of probe field Rabi frequency and the nature of the gain profiles in  $H_2$  and LiH molecules can be explained by analyzing those analytical expressions. It has been shown that AWI is more efficient when  $\Omega \sim 2G$  or  $3G$  than when  $\Omega \gg G$ . We have shown both analytically and numerically that in small diatomic molecules like  $H_2$  and LiH, AWI is feasible in VUV and violet region respectively in the case of both inhomogeneous and homogeneous broadening of levels, with and without considering the replenishment of the ground state. The advantage of choosing molecular system is that the profile of AWI and the magnitude of the maximum gain and absorption can be controlled by choosing different ro-vibrational levels as the upper lasing level of the  $\Lambda$ -system. Moreover, we have shown here that in  $\Lambda$ -system one can generate high frequency lasing on probe channel using low frequency coherent coupling laser in this molecule.

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