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Lasing without population inversion in molecules

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Abstract. We have described here the physical basis for lasing without population inversion (LWOPI). This type of amplification is obtained basically by two mechanisms: (i) one is based on atomic interference and (ii) the other is based on Fano-type interference. We have shown here, in H_2 molecules, amplification without population inversion is feasible by considering both the mechanisms.

Keywords. Autoionization; lasing.

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1. Introduction

By LWOPI or AWOPI we mean lasing or amplification of weak probe radiation by atomic/molecular system in the absence of population inversion between the upper and lower lasing levels of the system. Though a decade has passed since this subject of lasing or amplification without population inversion (LWOPI/AWOPI) was brought to light, yet experimental observation in the laboratory has started only during the last few years.

For this new type of laser-LWOPI, the idea of inversion is no longer the criterion of lasing. It is the positive balance of photons emitted to photons absorbed, which has become the primary criterion for amplification. Depending upon the inversion or non-inversion of the population, the decision is made whether it is a conventional laser or a new type laser (LWOPI) respectively. This non-inversion lasing (LWOPI), is basically possible due to interference between energy absorbing transitions in the atoms/molecules of the lasing medium. In other words, in atoms or molecules, the probability of absorbing the photons drop to near zero, due to this interference, maintaining the probability of emission. As a result a few atoms in the excited state are enough to produce lasing. To operate this type of LWOPI, two well controlled lasers are required, one to create the interference and the other to trigger light emission, and a source of ordinary incoherent light to pump the upper lasing level bypassing the blocked level.

Theoretical investigation of AWOPI had started since 1962 [1]. Through these years many groups have shown AWOPI/LWOPI in various schemes [2]. But, basically, there are

two mechanisms for amplification without population inversion. One mechanism is based on atomic interference and the other is based on Fano-type interference. The idea behind the first mechanism is associated with the interference of different transitions, when there is a coherent superposition of atomic levels. In this mechanism, there are two types of (Λ and V) schemes. In the former case, the interference of transitions are between the two closely spaced lower levels and an excited upper level, while in the latter case the interference of transitions are between the two closely spaced excited levels and a lower level. In the Fano type interference, the upper lasing level is an autoionizing level, which lies above the lowest ionization threshold. The principle behind this theory is the destructive interference between the direct ionization channel and the autoionization channel creating an asymmetric absorption profile, whereas the interference is absent for stimulated emission.

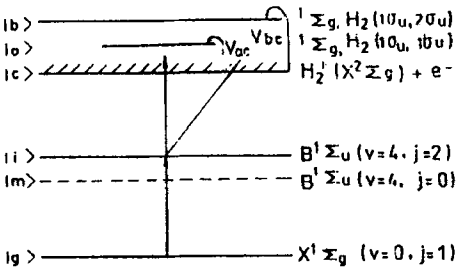
Most of the calculation on LWOPI and AWOPI done so far are parametric [2]. Recently, attempts are being made to study the feasibility of LWOPI in real atomic/molecular systems [3] both theoretically and experimentally. However, incidentally, our group for the first time, explored the feasibility of LWOPI in a molecular system like H_2 [4]. We have studied the feasibility of lasing in H_2 molecules by considering both the mechanisms. For the second mechanism, we have considered a $(1 + 1')$-photon transition scheme involving bound Rydberg levels and autoionizing level. In H_2 molecules, the effect of close lying rovibrational levels in the intermediate step and the presence of higher AI states embedded into the same continuum cannot be ignored. Inclusion of these effects give rise to new features: (i) LWOPI can be obtained from the higher AI state, although it is not being externally pumped and (ii) lasing at different close by frequencies can be obtained when the probe field frequency is tuned through resonances with intermediate rovibrational levels. This has been found to be suitable for obtaining gain in the UV and VUV frequency range. Furthermore, we have shown that persistence of LWOPI can be controlled by changing the intensity of the coherent pump field [4].

Recently, we have studied the first mechanism by considering the three level and four level schemes involving ground state and the bound Rydberg levels of H_2 molecules, where the coherence is created by coupling two of the levels by strong driving field coherently. We have shown that the inversionless amplification is feasible in UV and VUV range in H_2 molecules. Further study in alkali molecules is also in progress. Recently, Scully and coworkers have demonstrated LWOPI in atoms using a similar scheme [5].

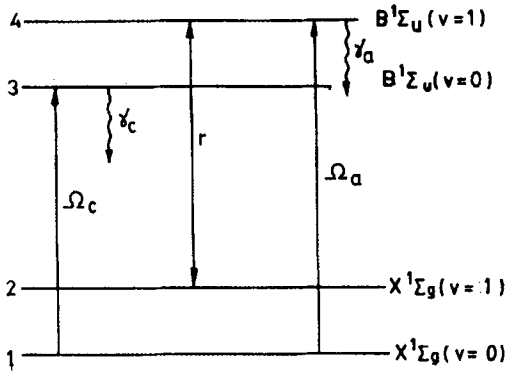
After these elaborate theoretical studies, clear experimental evidence of AWOPI and LWOPI was reported only recently. The first experimental report on AWOPI was in 1992 [6] using sodium atoms. However the first clear-cut evidence of AWOPI was reported in 1993 [7] using picosecond-pulses, in atomic samarium vapour. Many groups [8] have done experimental study on AWOPI in various atoms. However, this new type of lasing is still confined to the laboratory.

2. Transition schematics

The two types of transition schemes considered here are shown in figures 1a and 1b. In the first scheme involving autoionizing states (figure 1a), we have considered here a $(1 + 1')$-photon resonant transition scheme from the ground $X^1\Sigma_g(v = 0; j = 1)$ state to the lowest and the first excited AI states of $^1\Sigma_g$ symmetry via the intermediate resonant



(1a)



(1b)

Figures 1(a) and (b). Schematic diagrams for the transition schemes considered in the H_2 molecule.

$B^1\Sigma_u(v=4; j=2)$ state of the H_2 molecule. Two AI states are coupled to each other via electronic continuum by configuration interaction coupling. The transition scheme has been shown schematically in figure 1a. To obtain the effect of autoionization via near-resonant rovibrational levels on the resonant channel, we have considered transitions via $(v=4, j=0)$, $(v=5, j=0)$, $(v=5, j=2)$ and $(v=3, j=2)$ levels separately. In this case the probe field intensities for the lower and the upper transitions are much less than 10^8 W/cm^2 and hence the coupling between these near-resonant rovibrational levels has been ignored [9]. But the coupling between resonant and the near-resonant levels via continuum has been considered.

For this scheme, the lower probe field has been kept fixed around the resonance between the $X^1\Sigma_g(v=0, j=1)$ level and $B^1\Sigma_u(v=4, j=2)$ level, i.e., around the wavelength 105 nm. The mechanism for generation in VUV range is the frequency mixing of a visible wavelength ($\sim$ 5250Å) from a Nd-Yag pumped dye laser with two UV wavelengths ($\sim$ 2625Å), which can be obtained by frequency doubling the visible wavelength [10]. In our calculation we have shown that the peak power required for this probe field is $\sim$ 1 W/cm^2. This peak power can easily be obtained, if one can generate 10 ps pulses of typical energy 10×10^{-15} J and by focussing it to a spot area of 0.1 mm^2. If the pulse energy obtained is less than the above value, one will have to focus the probe beam more tightly, in the interaction region, so that the peak power can be raised to 1W/cm^2.

The wavelength of the upper probe field is varied around the resonances with the lower and the upper AI states from the intermediate level, i.e., around 315 nm and 234 nm respectively. These wavelengths can be obtained from Nd-Yag pumped dye lasers (by frequency doubling the fundamental) [11]. The peak power required in our calculation is $\sim 10^6$ W/cm^2. This can be easily obtained from these lasers by focussing 10 ps probe pulses of typical energy 10 nJ into the area of 0.1mm^2. Nevertheless, if the pulse energy is lower than this value, tight focussing is required, to obtain the above mentioned peak power. Therefore, for both the probe pulses, the focussing criterion can be relaxed, if the pulse energy is higher than the above mentioned values.

The second scheme (figure 1b)is a four-level scheme [12], where the two rovibrational levels of the ground $X^1\Sigma_g$ state are coupled to a pair of rovibrational levels of the excited $B^1\Sigma_u$ state by three fields. A strong driving field with Rabi frequency Ω_c and a weak probe field with Rabi frequency Ω_a are assumed to be quasimonochromatic. These fields have linewidths much less than atomic radiative decay rates (γ_a and γ_c). The third (pump) field is taken to be incoherent with a very broad linewidth and the pumping rate is represented by 'r' (figure 1b).

3. Theory

We have used the resolvent operator technique and the density matrix formalism to study the LWOPI process in the first and the second scheme respectively. We will describe here the resolvent operator technique only. The density matrix formalism can be obtained in the literature [13]. We will show here that by using resolvent operator technique, one can study the evolution of a system and hence the gain. The product states considered here are $|g\rangle\ |n\rangle$, $|i\rangle\ |n-1\rangle$, $|m\rangle\ |n-1\rangle$, $|a\rangle\ |n-2\rangle$, $|b\rangle\ |n-2\rangle$ and $|c\rangle|n-2\rangle$ for the ground, intermediate (resonant and near-resonant), lowest and first excited AI states and continuum respectively. Starting from the resolvent operator equation

$$(Z-H)\,G(Z)=1 \tag{1}$$

one obtains a set of equations for the matrix elements of resolvent operator as follows:

$$(Z-E_g)\,G_g - D_{gi}G_i - D_{gm}G_m = A \tag{2a}$$

$$(Z-E_i)\,G_i - D_{ig}G_g - D_{ia}G_a - D_{ib}G_b - \int D_{ic}G_c\,\mathrm{d}E_c = 0 \tag{2b}$$

$$(Z-E_m)\,G_m - D_{mg}G_g - D_{ma}G_a - D_{mb}G_b - \int D_{mc}G_c\,\mathrm{d}E_c = 0 \tag{2c}$$

$$(Z-E_a)\,G_a - D_{ai}G_i - D_{am}G_m - \int V_{ac}G_c\,\mathrm{d}E_c = B \tag{2d}$$

$$(Z-E_b)\,G_b - D_{bi}G_i - D_{bm}G_m - \int V_{bc}G_c\,\mathrm{d}E_c = 0 \tag{2e}$$

$$(Z-E_c)\,G_c - V_{ca}G_a - V_{cb}G_b - D_{ci}G_i - D_{cm}G_m = 0 \tag{2f}$$

where D_{pq} is the dipole transition moment between product states $|p\rangle$ and $|q\rangle$ and V_{cp} is the configuration interaction coupling of state $|p\rangle$ with the continuum $|c\rangle$. E_g, E_i, E_m, E_a, E_b and E_c are the energies of the product states as specified by the subscripts on E. The matrix element of resolvent operator G_{rs} can be written as $G_{rs} = \langle r|(1/(Z-H))|s\rangle$, where H is the total hamiltonian for the system. The choice of the second subscript on G_{rs} depends on the boundary condition and has been omitted in the above equations for the matrix

elements of resolvent operators. It is to be mentioned here that by formally solving for G_c from (2f) and substituting it in other equations, one can write down a set of equations in terms of ionization widths $\gamma_i(=2\pi|D_{ic}|^2)$ and $\gamma_m(=2\pi|D_{mc}|^2)$, autoionization widths $\Gamma_a(=2\pi|V_{ac}|^2)$ and $\Gamma_b(=2\pi|V_{bc}|^2)$, the coupling strength between two AI states via common continuum, two photon coupling strength between two intermediate levels via continuum and the detunings from these AI-levels (δ_a and δ_b) and from the intermediate levels (δ_i and δ_m). The coupling between the two AI levels depends on the system itself. But the two photon coupling between the two intermediate levels depends on the intensity of the radiation provoking the transition. We have solved the above equations twice with two different boundary conditions: $A = 1$, $B = 0$ for absorption and $A = 0$, $B = 1$ for emission. By solving the above equations one can obtain formally the elements of the resolvent operator as $G_p = (f_p(Z)/F(Z))$, where $F(Z)$ is the polynomial in Z and the roots of the equation $F(Z) = 0$ give the energies of the dressed states. The corresponding matrix element for the evolution operator $U_p(t)$ can be obtained by inverse Laplace transforms of $G_p(Z)$. The population of the product state $|p\rangle$ can be obtained as $|U_p(t)|^2$. We denote $U_p(t)$ as ${U_p}^{(a)}(t)$for absorption and ${U_p}^{(e)}(t)$ for emission.

The probability of absorption can be given as

$$P_a(t) = 1 - |{U_g}^{(a)}(t)|^2 - |{U_i}^{(a)}(t)|^2 - |{U_a}^{(a)}(t)|^2 - |{U_m}^{(a)}(t)|^2 - |{U_b}^{(a)}(t)|^2,$$

and the probability of emission can be given as

$$P_e(t) = |U_g^{(e)}(t)|^2 + |U_i^{(e)}(t)|^2 + |U_m^{(e)}(t)|^2.$$

Hence the gain as a function of time can be given as

$$E(t) = R_a P_e(t) - R_g P_a(t),$$

where R_a and R_g are the pumping rates to the lowest AI state $|a\rangle$ and the ground state $|g\rangle$ respectively. We have chosen $R_a = R_g = 1$ for this calculation. The positive value of $E(t)$ as a function of time corresponds to lasing or amplification. To ensure that the gain is obtained under the condition of population non-inversion we have studied the evolution of different product states $|{U_p}^{(a)}(t)|^2$.

4. Calculation

The dipole transition moments for the transitions from the ground state to the intermediate levels, from the intermediate levels to the lowest AI state and the continuum and also the autoionization width of the lowest AI state for the H_2 molecules have been obtained from our previous calculations [9]. Considering the ratio of the autoionization width of the first excited AI state and the lowest AI state to be 0.096 as obtained from the literature [10], we have calculated the autoionization width (Γ_b) of the first excited AI state to be 7.07×10^{-5} a.u. where the calculated value [9] of the autoionization width (Γ_a) of the lowest AI state is 7.369×10^{-4} a.u. The dipole transition moments for the transition from the intermediate levels to the excited AI state has been chosen to be equal to those for the lowest AI states. We have found that the effect of relative variation of these two dipole transition moments is insignificant to the amplification process. For the second scheme, the density matrix equations have been solved numerically for the steady state. The computed imaginary part

of coherence between two lasing levels, which is proportional to gain is shown in figure 6. The dipole transition moments between the ground state and the two excited states are taken to be the same. The decay time of the two excited states are of the order of 100 ns.

5. Results and discussion

Results for the first scheme are shown in figures (2)–(5). In figure 2, we have shown gain profiles around both the autoionizing (AI) states at the dimensionless time $\Gamma_a \tau = 60$ a.u. i.e. $\tau = 1.97$ ps. This shows prominent gain peaks around the resonance frequencies with the lower and upper AI states from the intermediate rovibrational levels of $B^1\Sigma_u$ state. The values of ϵ_a corresponding to resonance frequencies for the following transitions are: (i) $\epsilon_a = 0$ (from the lowest AI state to the $B^1\Sigma_u(v = 4, j = 2)$ level), (ii) $\epsilon_a = 1.2$(from the lowest AI state to the $B^1\Sigma_u(v = 4, j = 0)$ level) , (iii) $\epsilon_a = 136$ (from the excited AI state to the $B^1\Sigma_u(v = 4, j = 2)$ level) and (iv) $\epsilon_a = 137.2$ (from the excited AI state to the $B^1\Sigma_u(v = 4, j = 0)$ level) and hence the wavelengths for the resonant transitions are 315 nm, 314 nm, 234 nm and 233.5 nm respectively. The probe field intensities for this calculation are 0.96 W/cm^2 and 9.6×10^5 W/cm^2 for the lower and the upper probe respectively. In figure 3, we have shown absorption, emission and gain profiles around the first excited autoionizing state for higher upper probe field intensity ($9.6\times$ 10^6 W/cm^2). It is found that for this type of transition, the absorption is neglibly small and hence the net emission gives rise to the net gain.

The persistence of gain around $\epsilon_a = 136$ (i.e., at $\epsilon_a = 136.2$ and 135.8) has been shown in figure 4 and this sets the time limit for obtaining positive gain. Positive gain at $\epsilon_a = 136.2$ persists for the duration of $\tau = 3.14$ ps. The inversionless gain can be obtained

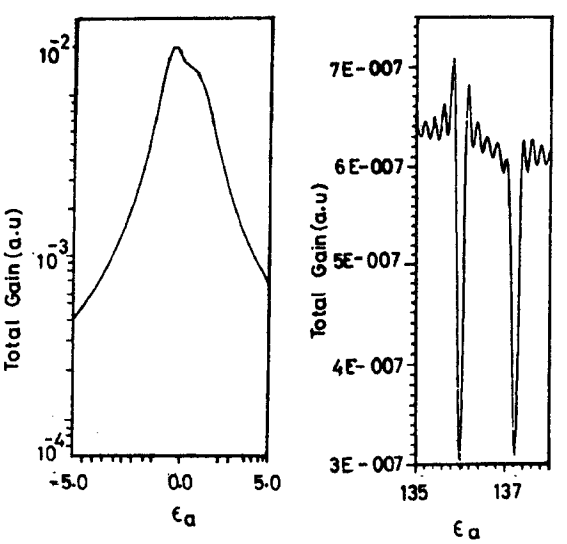


Figure 2. Total gain from the two lowest AI-states as a function of $\epsilon_a = \delta_a/(\Gamma_a/2)$. $(\Gamma_a \tau) = 60$. $\Gamma_a = 7.369 \times 10^{-4}$ a.u. and $\Gamma_b = 7.07 \times 10^{-5}$ a.u.. Intensities of the lower and the upper probe fields are 0.96W/cm^2 and 9.6×10^5 W/cm^2 respectively.

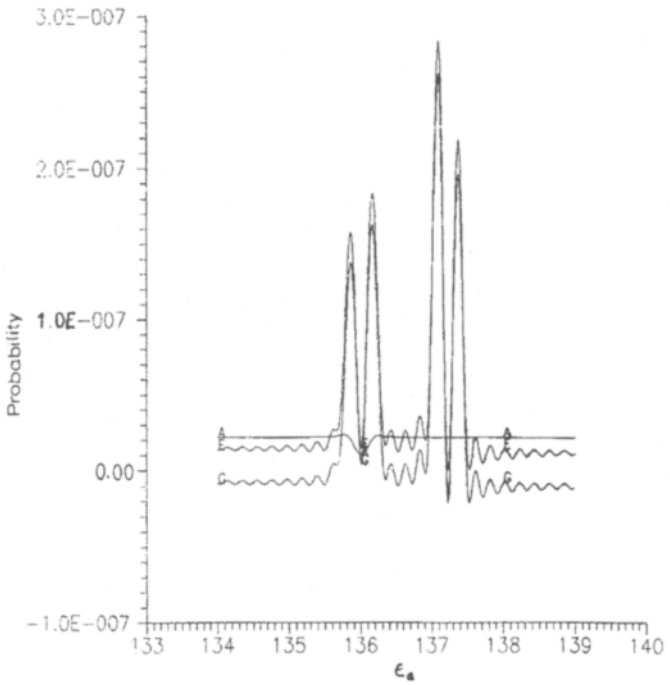


Figure 3. Absorption-(A), emission-(E) and gain-(G) profiles from the first excited AI-state as a function of dimensionless detuning ϵ_a, with the upper probe field intensity $9.6 \times 10^6 \text{W/cm}^2$. Other parameters are as in figure 2.

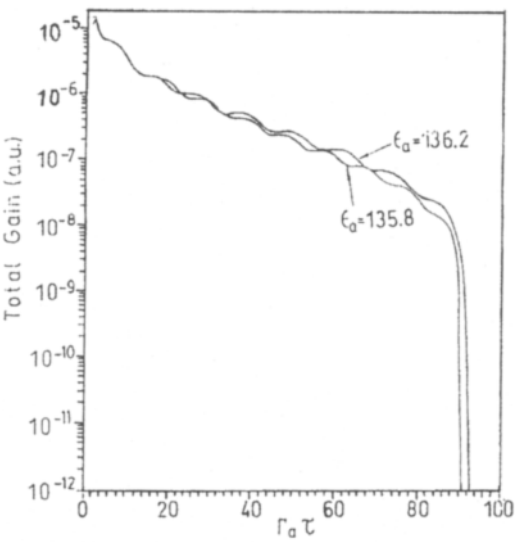


Figure 4. Persistence of total gain from the first excited AI-state at $\epsilon_a = 135.8$ and 136.2. Other parameters are same as in figure 2.

for a finite time and its duration is less for the gain obtained from the excited AI state than that from the lowest AI state [4]. It has been shown that the persistence of gain can be controlled by changing the probe field intensities [4].

So far we have considered the effect of near-resonant channel via $v = 4, j = 0$ of $B^1\Sigma_u$ state which is the closest near-resonant level to the resonant ($v = 4, j = 2$) level of the same electronic state. To show that the presence of nearby rovibrational levels can affect the gain on the probe fields in and around resonances with the two AI states from the $B^1\Sigma_u$ ($v = 4, j = 2$) level, we have calculated gain profiles for three other nearby rovibrational levels like $v = 5, j = 0$; $v = 5, j = 2$ and $v = 3, j = 2$ of $B^1\Sigma_u$ state. The coupling between these near-resonant rovibrational levels has been neglected because of the fact that the intensities of the probe fields are much lower than that required for these effects to be significant [9]. The gain profiles obtained for the emission from the excited AI state have been plotted in figure 5. We find that when the upper probe field frequency is tuned to the resonances between the AI level and the nearby rovibrational levels, gain peaks are obtained at several values of ϵ_a. The gain peaks for the transitions between the first excited AI state and the near-resonant levels (i.e. $v = 5, j = 2; v = 5, j = 0; v = 3, j = 2$) are at $\epsilon_a = 121.4$, 122.6, and 151.1 respectively (figure 5). Similarly, the gain peaks for the transitions between lowest AI state and the above-mentioned near-resonant levels are at $\epsilon_a = -14.6, -13.4$, and 15.0 respectively [4]. It is found that the presence of near-resonant channels via $v = 4, j = 0$ and $v = 5, j = 0$ (being the closest on either side of $v = 4, j = 2$) affect the gain at $\epsilon_a = 0$ and around $\epsilon_a = 136$. Hence, the effect of these

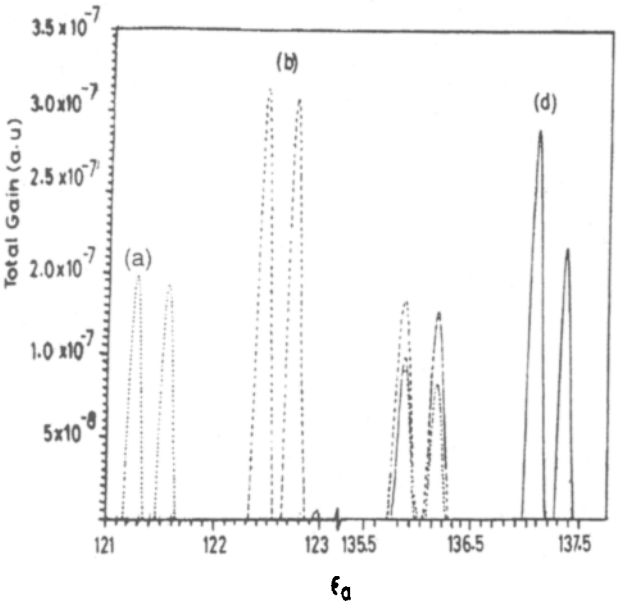


Figure 5. Total gain profiles for the first excited AI-state at $\Gamma_a\tau = 60$, considering the effect of different near-resonant rovibrational levels of $B^1\Sigma_u$; curves: (a) for the (v = 5, j = 2); (b) for the (v = 5, j = 0); and (d) for the (v = 4, j = 0). For curves (a), (b) and (d), peaks at $\epsilon_a = 135.85$ and 136.16 are due to the presence of $v = 4, j = 2$ level at $\epsilon_a = 136$.

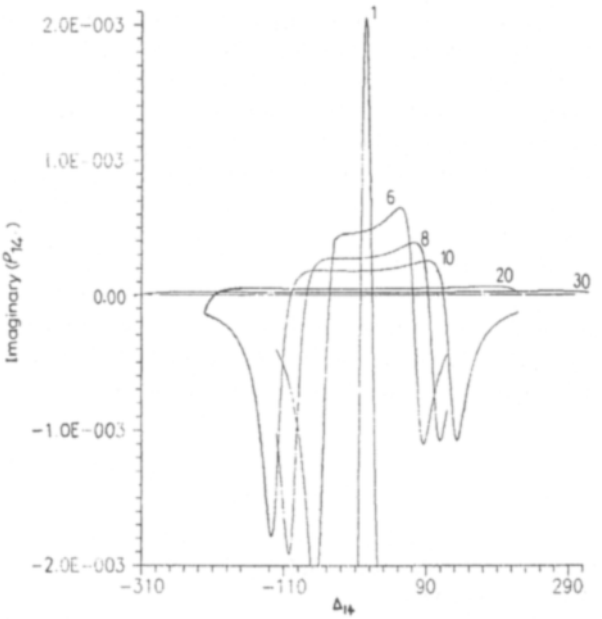


Figure 6. The imaginary part of the off-diagonal density matrix element between lasing levels (1 and 4) (in figure 1b) has been plotted as a function of detuning (Δ_{14}), from the upper lasing level $B^1\Sigma_u$ (v=1), for different values of driving field Rabi-frequency Ω_c (with values mentioned alongside the curves). The Rabi frequency of the weak probe field Ω_a = 0.8 MHz and the incoherent pumping rate r = 0.1 MHz.

near-resonant channels depends on the proximities of these nearby levels to the $v = 4$, $j = 2$ level of $B^1\Sigma_u$ state and the relative strength of dipole transition moments from these levels to the others (bound-bound and bound-continuum both). A detailed analysis has revealed (not shown here) that to obtain inversionless gain at resonances between the AI levels and the intermediate nearby rovibrational levels ($v = 4$, $j = 0$; $v = 5$, $j = 0$; $v = 5$, $j = 2$ and $v = 3$, $j = 2$), one will have to use probe pulses of duration ~ 0.35 ps which is much shorter than that obtained at $\epsilon_a = 0$, 135.8 and 136.2. This is because of the fact that the gain at these resonances oscillate between non-inversion and inversion, unlike the gains at $\epsilon_a = 0$, 135.8 and 136.2.

In the second scheme, we have shown the profile of the imaginary part of the density matrix element (ρ_{14}), which is proportional to gain (figure 6), as a function of the detuning (Δ_{14}) from the bound Rydberg state $B^1\Sigma_u$ ($v = 1$), for different strengths of the strong driving field Rabi frequency Ω_c (figure 1b). Here the weak probe Rabi frequency $\Omega_a = 0.8$ MHz and the incoherent pumping rate $r = 0.1$ MHz. It has been shown that this profile becomes broader with the increase in driving field intensity, at the cost of its peak value.

In conclusion, in the first scheme, we have shown that the AWOPI from an excited AI state in H_2 molecules can be obtained although this level is not being directly pumped. The lowest AI state is pumped incoherently and the population accumulated in the excited state due to the coherent coupling with the lowest AI state via continuum gives rise to AWOPI from the excited AI state. The main advantage for this type of scheme is that,

AWOPI can be obtained from an excited AI state by pumping the lower AI state embedded into the same continuum. Moreover, in molecular systems like H_2 molecules, one can obtain gain at several laser frequencies, by tuning the upper probe field frequency around the intermediate rovibrational levels and by controlling the pulse duration of the probe fields. In the second scheme, it has been shown that amplification in the VUV range can be obtained in H_2 molecules in absence of population inversion.

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