



Feasibility study of optically pumped molecular lasers with small quantum defect

A. Ratanavis *, N. Campbell, W. Rudolph

Department Physics and Astronomy, University of New Mexico, Albuquerque, NM 87131, USA

ARTICLE INFO

Article history:

Received 28 September 2009

Received in revised form 29 October 2009

Accepted 30 October 2009

Keywords:

Gas lasers

Lasers

Optical pumping

ABSTRACT

An analytical analysis of continuous wave (CW) optically pumped molecular lasers with small quantum defect is presented. The results show the conditions under which lasing with small Stokes shifts are possible. The general results are applied to HCN as an example gas. The predictions from the analytical approach are in good quantitative agreement with numerical calculations. The effective vibrational lifetimes and rotational relaxation rates of the molecule determine the principal behavior of such lasers. High overall laser efficiencies are possible for long interaction length of pump laser and gas media, which for example can be achieved in waveguide structures. This makes such lasers potentially interesting for coherently combining the output of mutually incoherent fiber and or diode lasers.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Since their first demonstration [1,2] optically pumped atomic vapor lasers have gained a great deal of interest as a potential tool to coherently combine the output of mutually incoherent fiber and or diode lasers. Such applications have become attractive after high power diode and fiber sources were successfully spectrally narrowed to a few GHz and below, which is necessary in order to match to the width of the absorption transition of the alkali metal vapors. The quantum efficiency of these three-level lasers is extremely high, for example 98% for Rb and 95% for Cs, enabling large laser slope efficiencies and overall efficiencies. Diode pumped alkali lasers (DPALs) lasers with slope efficiencies exceeding 80% (Cs) [3] and 70% (Rb) [4] have been reported. As with most optically pumped lasers, the beam profile and spectral brightness can also be improved considerably when comparing the pump laser (s) to the output of the atomic vapor laser, and several mutually incoherent pump sources can be employed. This was demonstrated with an Rb laser pumped by two laser diodes [5].

Optically pumped molecular lasers (OPMLs) have typically been excited by vibrational overtone transitions producing laser output radiation with large Stokes shifts [6–9]. In diatomic molecules the laser frequency is roughly an integer fraction of the pump frequency. When compared to atomic vapors, molecules have additional degrees of freedom, which create relaxation pathways that can channel excitation energy away from the desired laser transition. In diatomic molecules collision induced energy transfer among rotational states and vibrational relaxation can represent such competing energy dissipation pathways. In tri-atomic gas

molecules lasing with large Stokes shifts has been observed on transitions between the pumped level and a vibrational level other than the ground state [10,11].

To use OPMLs in a similar fashion as alkali vapor lasers, that is with a very small quantum defect, lasing has to occur between the pumped vibrational level and the vibrational ground state. Such molecular lasers have a number of attractive features. They can be operated at room temperature, and a certain amount of frequency tunability exists owing to the multitude of possible vibration–rotation transitions. A review of theoretical and the experimental work related to this concept for diatomic molecules and polyatomic molecules can be found in reference [12]. However, only cases were considered where the relaxation between excited state and ground state does not involve intermediate levels, restricting the applicability of the conclusions drawn.

In this contribution we analyze theoretically the potential of continuous wave (CW) molecular lasers operating on vibration–rotation transitions with small quantum defect for a broader class of molecules and transitions by allowing for the existence of an intermediate state. The analysis applies to any optically pumped molecular laser with small quantum defect. This includes polyatomic molecules where lasing can occur between a combination state or overtone state and the ground state. The model also describes diatomic molecules where relaxation back to the ground state involves intermediate levels because of vibrational ladder climbing.

We evaluate an OPML subject to some simplifying assumptions analytically and then use a computer model to test these results under more realistic conditions using the molecular parameters of HCN (as far as they are known) as an example. HCN, C₂H₂ and HI are attractive OPML candidates because they have absorption bands that coincide with the emission of C-band fiber lasers in

* Corresponding author. Tel.: +1 505 277 1868.

E-mail address: amarin@unm.edu (A. Ratanavis).

the 1.5 μm [13–15] region suggesting laser output in the eye-safe spectral region. The necessary long interaction lengths between the active gas and high pump intensities can in principle be realized using hollow fibers which have been used for example for saturated absorption spectroscopy with C_2H_2 [16] and for stimulated Raman scattering in H_2 [17].

2. Laser model

2.1. Analytical model

A schematic energy level diagram of the molecules to be considered is shown in Fig. 1. The pump laser excites a transition between a rotational level (rotational quantum number J) of the vibrational ground state $|o\rangle$ and a rotational level with quantum number $J' = J + \Delta J$ of an excited vibrational level $|e\rangle$ ($\Delta J = \pm 1$ for an R and P transition, respectively). The molecule can relax back to the vibrational ground state via an intermediate state $|i\rangle$. The time τ_e represents a characteristic time for collision induced vibrational relaxation back to the ground state if an intermediate state $|i\rangle$ is not occupied, that is $\tau_o = 0$. In general, other vibrational states can be involved in the relaxation paths shown in Fig. 1 with the two effective time constants being a simple representation of this situation. Such relaxation paths can include intermediate vibrational states that are located energetically between $|e\rangle$ and $|o\rangle$ and states with energies higher than $|e\rangle$ (vibrational ladder climbing).

For an analytical approach let us first assume that rotational relaxation within one vibrational state is fast so that at all times a thermalized rotational population in $|o\rangle$ and $|e\rangle$ exists. The occupation numbers in a certain rotational state J at temperature T are thus given by

$$n_{o,e}(J) = (2J+1)e^{-\beta_{o,e}(J)} \frac{N_{o,e}}{S_{o,e}}, \quad (1)$$

where $S_{o,e} = \sum_J (2J+1)e^{-\beta_{o,e}(J)}$ with $\beta_{o,e}(J) = E_{o,e}(J)/(k_B T)$. For a simple diatomic or linear molecule the rotational energy relative to the rotational ground state of the respective vibrational level $E_{o,e} = h^2 J(J+1)/(2I)$ where I is the moment of inertia, $N_{o,e}$ is the total population density in the vibrational states, and k_B is the Boltzmann constant.

The gain coefficient for a transition from $|e, j\rangle$ to $|o, j + \Delta j\rangle$ can be written as

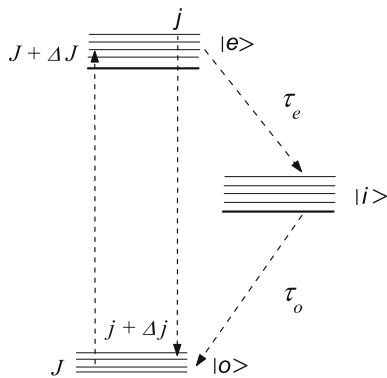


Fig. 1. Simplified molecular energy level diagram. $|o\rangle$ and $|e\rangle$ denote vibrational states with rotational manifolds that in thermal equilibrium are populated according to a Boltzmann's distribution. $|i\rangle$ can represent an ensemble of states that participate in the relaxation of the excited vibrational state $|e\rangle$ to the ground state $|o\rangle$. τ_e and τ_o are effective relaxation times indicating how fast $|e\rangle$ is depopulated and $|o\rangle$ populated.

$$g = \sigma_j \left[n_e(j) - \frac{2j+1}{2(j+\Delta j)+1} n_o(j+\Delta j) \right], \quad (2)$$

where σ_j is the gain cross section. The occupation numbers needed to calculate g can be obtained from Eq. (1). The rate equations for the total occupation numbers in steady state are

$$\frac{dN_o}{dt} = -R_p \frac{2J+1}{S_o} e^{-\beta_o(J)} \left[N_o - \frac{S_o}{S_e} e^{\Delta\beta_{ej}} N_e \right] + \frac{N_i}{\tau_o} = 0, \quad (3a)$$

$$\frac{dN_i}{dt} = \frac{N_e}{\tau_e} - \frac{N_o}{\tau_o} = 0, \quad (3b)$$

$$N_t = N_e + N_o + N_i, \quad (3c)$$

where $\Delta\beta_{ej} = \beta_o(J) - \beta_e(J + \Delta J)$, and the pump rate (photons/s) $R_p = \phi_p \sigma_j$ is the product of the pump photon flux and the absorption cross section.

Assuming $S_o = S_e = S$, the gain coefficient becomes:

$$g(J, J + \Delta J, j, j + \Delta j) = \sigma_j \frac{2j+1}{S} e^{-\beta_e(j)} N_t \left[\frac{\tau_e R_p e^{-\beta_o(J)} (1 - e^{\Delta\beta_{ej} + \Delta\beta_{ej}}) - e^{\Delta\beta_{ej}}}{\frac{S}{2j+1} + R_p e^{-\beta_o(J)} (\tau_e + \tau_o + \tau_e e^{\Delta\beta_{ej}})} \right], \quad (4)$$

where $\Delta\beta_{ej} = \beta_e(j) - \beta_o(j + \Delta j)$.

From Eq. (4) it is obvious that positive gain can occur only if $\Omega = \Delta\beta_{ej} + \Delta\beta_{ej} < 0$. This means that the laser photon energy must be smaller than the pump photon energy. The condition:

$$\Omega = -\frac{h^2}{2I} (\Delta J^2 + 2J\Delta J + \Delta J + \Delta j^2 + 2j\Delta j + \Delta j) / k_B T < 0, \quad (5)$$

determines which combinations of pump and lasing transitions are not possible for lasing to occur independent of the pump rate and relaxation parameters. For example, excitation of a P-line transition and lasing on a R-line transition would result in $\Omega > 0$, which according to Eq. (4) does not permit positive gain.

To include the lasing process from state $|e, j\rangle$ to state $|o, j + \Delta j\rangle$ we can add to Eq. (3a) a term:

$$R_L \frac{2j+1}{S} e^{-\beta_o(j+\Delta j)} [e^{-\Delta\beta_{ej}} N_e - N_o], \quad (6)$$

where $R_L = \sigma_j \phi_L$ is the product of the laser emission cross section σ_j and the laser photon flux ϕ_L . The rate equation for ϕ_L after steady state has been reached can be written as,

$$\frac{d\phi_L}{dt} = c\sigma_j \phi_L \frac{L_g}{L} \left\{ \frac{2j+1}{S} e^{-\beta_o(j+\Delta j)} [e^{-\Delta\beta_{ej}} N_e - N_o] \right\} - \frac{\phi_L}{\tau_c} = 0, \quad (7)$$

where τ_c is the photon cavity lifetime, L is the cavity length and L_g is the length of the gain medium. The cavity lifetime $\tau_c = 2L/c / [-\ln(1 - L_f) - \ln(R_m)]$ is determined by the reflectance R_m of the outcoupler and the combined other linear losses per roundtrip, L_f .

Note that to force this one-mode laser operation on the transition from j to $j + \Delta j$, if necessary, a suitable spectral filter can be placed in the cavity preventing all other lasing transitions. In steady state the gain coefficient equals the cavity loss coefficient:

$$g_L = \sigma_j \frac{L_g}{L} \left\{ \frac{2j+1}{S} e^{-\beta_o(j+\Delta j)} [e^{-\Delta\beta_{ej}} N_e - N_o] \right\} = \frac{1}{c\tau_c}. \quad (8)$$

Eqs. (8) and (3a) with the addition of the laser term Eq. (6) yield the steady-state laser output:

$$P_L = h\nu_j A L_g Q \left\{ R_p \frac{2J+1}{S} e^{-\beta_o(J)} \left[N_t (1 - e^{\Omega}) - \frac{S'}{\gamma\tau_c} (1 + \gamma e^{\Delta\beta_{ej}}) \right] - \frac{1}{\tau_e} \left[\frac{S'}{\tau_c} + N_t e^{\Delta\beta_{ej}} \right] \right\}, \quad (9)$$

where h is the Plank constant, ν_j is the laser frequency and A is the cross section of the laser beam which is assumed to be the same as the pump beam cross section:

Download English Version:

<https://daneshyari.com/en/article/1539513>

Download Persian Version:

<https://daneshyari.com/article/1539513>

[Daneshyari.com](https://daneshyari.com)