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Weakly Aligned Molecules: From Molecular Detectors to Room-Temperature Tunable Masers

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Abstract.

We have developed and demonstrated a new mechanism of manipulation of population in molecular rotational levels in a weakly aligned molecules. The mechanism is based on an adiabatically changing electric field interacting with the molecules with dipole moments. Treating molecules as simple rotators, we have described their behavior using the density matrix taking into account the relaxation processes. We have considered the interaction of the weakly aligned molecules with a microwave field in a high finesse cavity. We have found that, on one hand, the population inversion can be reached in the ensemble of the weakly aligned molecules to be used for the maser operation at room temperature. On the another hand, we have found that the enhancement of the absorption can reach the theoretical limit and be used for gas sensing with high sensitivity and selectivity. Such sensors can efficiently analyze the multi-gas mixtures and be used for a huge range of applications – stretching from technology, sciences, control of environment, biology and medicine.

1. Introduction

The maser (microwave amplification by stimulated emission of radiation), the coherent source of the microwave radiation similar to the laser, was demonstrated in 1954 [1]. However, unlike lasers [2], the masers are much less widely used because in order to function, they must be cooled to temperatures close to absolute zero. In 2012, it was demonstrated that a maser could operate at room temperature using the organic molecule pentacene. However, it only produced short pulses of maser radiation. For continuous operation of the maser, the crystal would likely have melted [3]. Only recently, it was reported that a new maser is created that operates continuously [4].

Meanwhile, we have to underline here that the maser technology being used where sensitive detection of microwave radiation is essential. The masers could be used in a range of applications

such as medical imaging and airport security scanning. They have more traditionally been used in deep space communication and radio astronomy. As well as medical imaging and airport security scanning, masers could play a pivotal role in improving sensors to remotely detect bombs, new technology for quantum computers, and might even improve space communication methods to potentially find life on other planets. Thus, the development tunable maser working at room temperature is very important.

On the other hand, to detect molecular gas dispersion or absorption or to create population inversion in molecular levels at room temperature is difficult for rotational frequencies because the population difference is very low at room temperature and probability to absorb photon is very low. Indeed, the probability for photon being absorbed is given by

$$P_{abs} = 1 - \exp\left[-\frac{\omega_{ab}\wp_{ab}^2 N_0 L}{c\hbar\Gamma_{ab}} n_{ab}\right] \tag{1}$$

where ω_{ab} is the frequency of transition, $\wp_{ab}$ is the dipole moment, Γ_{ab} is the width of transition, N_0 is the gas density, and n_{ab} is the population difference between corresponding levels. It is clear that low frequency ω_{ab} and small n_{ab} results in low absorption probability. But it would be desirable to enhance the probability of absorption for individual molecules to 100 %, so each molecule can absorb one photon. Then, this can be extremely useful to reach maximal absorption for gas detectors as well as creation of population inversion in a molecular gas for the maser operation.

In this paper, we show that developing techniques to control and to manipulate the populations in the rotational molecular levels can enhance the sensitivity of gas detection, and also it can open possibilities of creation population inversion that can be used for developing masers working at room temperature.

Applying a dc electric field aligns molecules along the field, and modifies the rotational energy levels of molecules [5, 6] (see Figure 1(a)). Aligned molecules can be viewed as simple pendulums that have the energy structure of a simple harmonic oscillator (see Figure 1(a)). Even a small electric field produces the weak alignment that can be take advantage for development various techniques of quantum control of rotational molecular states. The control is based on an adiabatically changing electric field that interacts with the rotational structure of the molecules with dipole moments. In particular, these new techniques are important for developing gas masers at room temperature as well as the developing of new gas sensors with higher selectivity and sensitivity [7, 8].

There are several physical mechanisms to change absorption that are related to the so-called quantum coherent effects. Quantum coherence effects, such as coherent population trapping [9] and electromagnetically induced transparency (EIT) [10, 11, 12], have been the focus of a broad range of research activity for the past two decades since they drastically change the optical properties of the media. In EIT, for example, absorption practically vanishes in both the CW and the pulsed regime [11]. A medium with an excited quantum coherence, phaseonium [10], can be used to make an ultra-dispersive prism [13] which will have several orders of magnitude greater angular spectral dispersion compared to a conventional one [14, 15, 16, 17, 18]. The importance and success of proper quantum engineering was demonstrated, in particular, for the pulse shaping in coherent Raman spectroscopy [19] that allows researchers to improve sensitivity. Here, we propose a new approach that results in the enhancement of the population difference between corresponding molecular levels and reaches the theoretical maximum of absorption.

The new approach to the problem is based on introducing mechanism based on an adiabatically changing electric field interacting with the rotational structure of the molecules with dipole moments. Normally the gases do not absorb radiation if optical density is very small. Contrary, using this approach, every gas molecule absorbs a photon. This can be reached by using an adiabatically changing electric field that is sweeping the resonant frequency through

the frequency of the ac probe field resulting in complete population transfer in the molecular system. Together with the change of photon numbers, the phase change of the probe field occurs. Both changes can be used as signals for gas sensing. Using proper tailoring of the adiabatically changing electric field allows researchers to resonantly enhance the sensor signal reaching the theoretical limit.

2. Interaction with chirping adiabatic pulses

Let us consider the interaction of the adiabatically changing fields with a two-level system. For the two-level atom, the Hamiltonian is given by

$$\hat{H} = \hbar\Delta|a\rangle\langle a| + \hbar\Omega(|a\rangle\langle b| + |b\rangle\langle a|), \tag{2}$$

where a and b are the excited and ground levels of the atom, $\Delta = \omega_{ab} - \omega$ is the detuning between frequency of the field ω and the resonance frequency of the transition ω_{ab}, Ω is the coupling due to interaction with adiabatically changing field. The eignevalues are

$$\lambda_\pm = \omega_\pm = \frac{\Delta}{2} \pm \sqrt{\left(\frac{\Delta}{2}\right)^2 + \Omega^2} \tag{3}$$

and eigenvectors are given by

$$|\pm\rangle = \frac{\omega_\pm|a\rangle + \Omega|b\rangle}{\sqrt{\omega_\pm^2 + \Omega^2}} \tag{4}$$

Let us note here that for $\Omega = 0$ and $\Delta > 0$: $\omega_+ = \Delta$, $|+\rangle = |a\rangle$ and $\omega_- = 0$, $|-\rangle = |b\rangle$, and for $\Omega = 0$ and $\Delta < 0$: $\omega_+ = 0$, $|+\rangle = |b\rangle$ and $\omega_- = \Delta$, $|-\rangle = |a\rangle$. For $\Omega \neq 0$, adiabatically changing Δ from $-\infty$ to $+\infty$ leads to adiabatical changes $\omega_\pm \rightarrow \omega_\mp$ and $|\pm\rangle \rightarrow |\mp\rangle$, as

$$\omega_+(\Delta) = \left\{ \begin{array}{ll} 0, & \Delta \rightarrow -\infty \\ \Delta, & \Delta \rightarrow +\infty \end{array} \right. \text{ and } |+\rangle(\Delta) = \left\{ \begin{array}{ll} |b\rangle, & \Delta \rightarrow -\infty \\ |a\rangle, & \Delta \rightarrow +\infty \end{array} \right. \tag{5}$$

$$\omega_-(\Delta) = \left\{ \begin{array}{ll} \Delta, & \Delta \rightarrow -\infty \\ 0, & \Delta \rightarrow +\infty \end{array} \right. \text{ and } |-\rangle(\Delta) = \left\{ \begin{array}{ll} |a\rangle, & \Delta \rightarrow -\infty \\ |b\rangle, & \Delta \rightarrow +\infty \end{array} \right. \tag{6}$$

and the states $|a\rangle$ and $|b\rangle$ can be expressed in terms of $|\pm\rangle$ states as

$$|a\rangle = \frac{\sqrt{\omega_+^2 + \Omega^2}|+\rangle - \sqrt{\omega_-^2 + \Omega^2}|-\rangle}{2\sqrt{\left(\frac{\Delta}{2}\right)^2 + \Omega^2}} \text{ and } |b\rangle = \frac{-\omega_-\sqrt{\omega_+^2 + \Omega^2}|+\rangle + \omega_+\sqrt{\omega_-^2 + \Omega^2}|-\rangle}{2\Omega\sqrt{\left(\frac{\Delta}{2}\right)^2 + \Omega^2}}. \tag{7}$$

Then, an arbitrary state $|\psi_i\rangle = A|a\rangle + B|b\rangle$ changes into $|\psi_f\rangle = Be^{i\phi_a}|a\rangle + Ae^{i\phi_b}|b\rangle$, performing swap the populations in the states. It is a remarkable because it can lead to the maximal absorption on one hand, and to creation of population inversion on the other. Below there are applications of this swap of populations to the gas sensing and to the maser operation.

3. Theoretical Model

For simplicity and to understand essential physics, we consider molecules with electric dipole moments $\vec{\wp}$, and the Hamiltonian in the dc electric field $\vec{\mathcal{E}}_0$ is given by

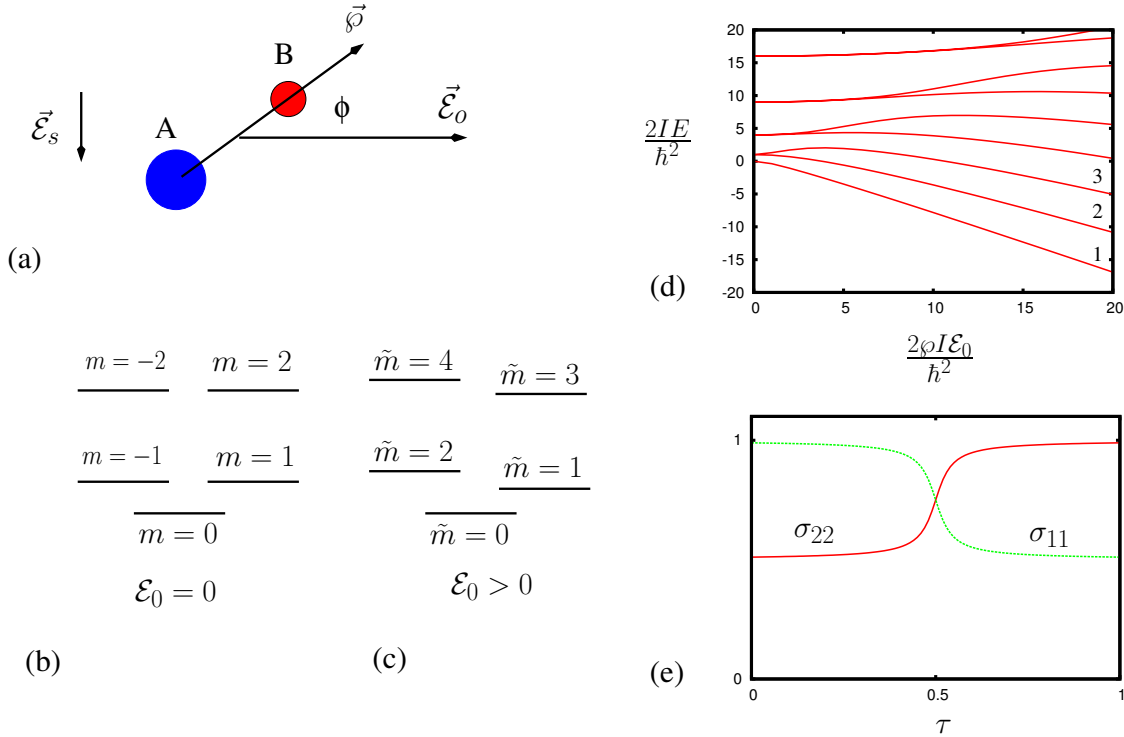$$\hat{H} = B\hat{J}_z^2 - \vec{\wp} \cdot \vec{\mathcal{E}}_0 \tag{8}$$

Figure 1. (a) Molecular rotator with electric dipole $\vec{\wp}$, where ϕ is the angle of rotation with respect to the electric field $\vec{\mathcal{E}}_0$, and the ac electric field $\vec{\mathcal{E}}_s$ orthogonal to the dc field that is tuned to the resonance with the molecules transitions; (b) the energy structure of the molecular rotator with no electric field ($\mathcal{E}_0 = 0$); (c) the energy structure of the molecular rotator with electric field ($\mathcal{E}_0 > 0$); (d) Molecular energy $2IE_{\tilde{m}}/\hbar^2$ dependence on $2I\wp\mathcal{E}_0/\hbar^2$. The numbers 1, 2, and 3 correspond to the states with $\tilde{m} = 0$, $\tilde{m} = 1$, and $\tilde{m} = 2$ correspondingly. (e) Dependence of the population in levels $\tilde{m} = 1$ and $\tilde{m} = 2$ on time τ.

Here, we can view symmetric molecules as simple rotators (see Figure 1(a)) to simplify our consideration [20] ($B = \hbar^2/2I$, I is the moment of inertia).

The first excited rotational states are split in the dc electric field due to Stark effect (see Figure 1(b,c)), and this energy splitting depends on $\mathcal{E}_0$

$$\frac{2I(E_2 - E_1)}{\hbar^2} = F\left(\frac{2\wp I\mathcal{E}_0}{\hbar^2}\right)$$

The splitting can be tuned to any frequency $\omega_0 = \dfrac{(E_2 - E_1)}{\hbar}$, and the transition can be used for maser operation and for spectroscopy as well. The strong (dc or slowly varying) electric field control the population and the splitting between levels, and the ac electric field orthogonal to the dc field is tuned to the resonance with the molecules transitions. To pump maser the ac pump field is used. Then, for maser operation and spectroscopy, the maser field and the probe field are ac fields with frequencies from RF to microwave range. The ac pump and maser and probe fields $\mathcal{E}_s$ can be created by the high finesse resonance circuits or high finesse microwave cavities. The equation for the Rabi frequency $\Omega_s = \wp\mathcal{E}_s/\hbar$ is given by

$$\dot{\Omega}_s = -i(\omega_0 - \omega_s)\Omega_s + i\Omega_a^2\rho_{ab} \tag{9}$$

$$\hat{H} = -B\frac{\partial^2}{\partial\phi^2} \qquad\qquad \hat{H} = -B\frac{\partial^2}{\partial\phi^2} - \wp E_0 \cos\phi \cdot$$

$$|m| = 3$$
$$|m| = 2$$
$$|m| = 1$$

$$E_n = \frac{\hbar^2 m^2}{2I} = Bm^2$$

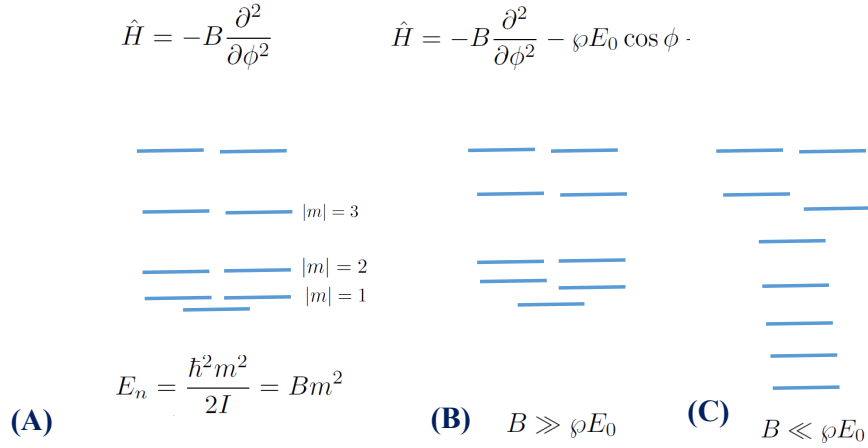**(A)**   **(B)** $B \gg \wp E_0$ **(C)** $B \ll \wp E_0$

Figure 2. Molecular level structure in the different dc electric fields. (A) $\mathcal{E}_0 = 0$, no dc field, level structure of rotator; (B) $\mathcal{E}_0$ is weak ($B \gg \wp\mathcal{E}_0$), one can see small splitting of the low levels of rotator; (C) $\mathcal{E}_0$ is strong ($B \ll \wp\mathcal{E}_0$), one can see that the low levels of rotator look similar to the levels of the simple harmonic oscillator (see Eq. (15) and Eq. (16)).

where ω_s is the frequency of the resonance circuit, $\Omega_a^2 = \dfrac{2\pi\omega_0\wp^2 N}{\hbar}$ is the cooperative frequency, and ρ_{ab} is the molecular coherence between the molecular states involved, ρ_{ab} is the element of the density matrix ρ.

Taking into account relaxation in all processes described above is very important. For this purposes, we use the Liouville-von Neumann equation for the density matrix to describe the time evolution of the molecules. The decoherence of the individual three-level atomic system is taken into account using Lindblad superoperator formalism [21].

$$\frac{\partial\rho}{\partial t} = \frac{i}{\hbar}[\rho, H] - \hat{\Gamma}[\rho] \tag{10}$$

where $\hat{\Gamma}[\rho]$ is the relaxation matrix for all components of the density matrix ρ. The set of density matrix equations are given by

$$\dot{\rho}_{ab} = -(\Gamma_{ab} + i\Delta)\rho_{ab} + in_{ab}\Omega_s \tag{11}$$

$$\dot{\rho}_{bb} = i(\rho_{ba}\Omega_s - \Omega_s^*\rho_{ab}) \tag{12}$$

$$\dot{\rho}_{aa} = i(\rho_{ab}\Omega_s^* - \Omega_s\rho_{ba}) \tag{13}$$

where $n_{ab} = \rho_{aa} - \rho_{bb}$, and $\Delta = \omega_{ab}(\Omega_0) - \omega_0$.

The modification of the energy structure leads also to redistribution of population in the rotational levels. Namely, the population difference between levels $\tilde{m} = 1$ and $\tilde{m} = 2$ (see in Figure 1(d) and even more clear see Figure 2(A), (B), (C)) is given by

$$N_2 - N_1 = \frac{\hbar^2 N_0}{2Ik_B TZ}F\left(\frac{2\wp I\mathcal{E}_0}{\hbar^2}\right), \text{ where } Z = \sum_{\tilde{m}=0}^{\infty} e^{-\frac{E_{\tilde{m}}}{k_B T}} \tag{14}$$

where k_B is the Boltzmann constant, T is the room temperature, and N_0 is the molecular gas density. The population distribution for case of the electric field strong enough as shown
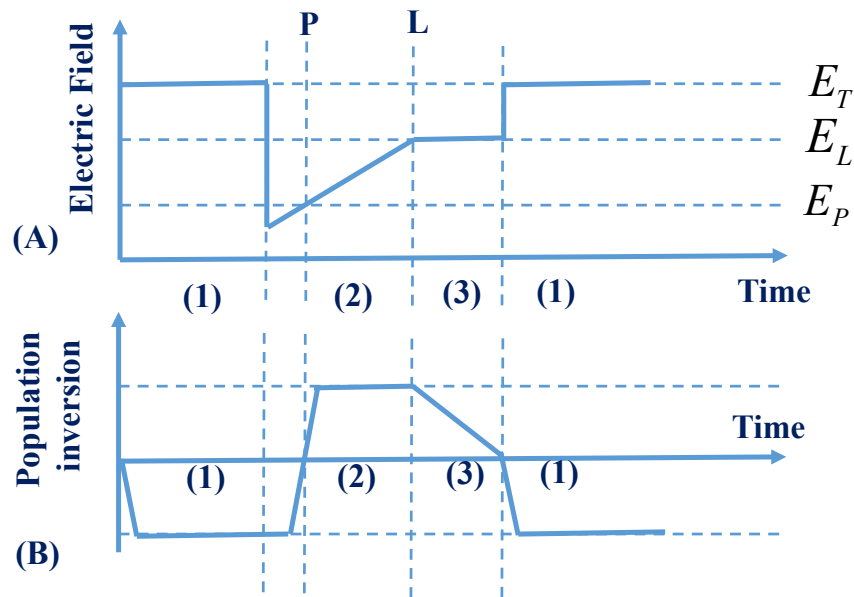
Figure 3. Time dependence of the adiabatically changing strong electric field (arbitrary units) $\mathcal{E}_0$ shown in (A). The corresponding population inversion between corresponding levels (arbitrary units) is shown in (B). Different time periods corresponds different phases of molecules in the cavity. (1) - the thermal pumping; (2) - adiabatic change of electric field to produce population inversion between corresponding levels due to some strong field (as shown in Figure 1(e)); (3) - reaching the resonance with maser transition, microwave amplification, depleting all population inversion and then returning to the pumping stage (1).

in Figure 2(C) will transfer to the same population difference in Figure 2(B) with adiabatic change of the electric field $\mathcal{E}_0$. We can see that this is similar to the magnetic cooling with the adiabatically changing magnetic field [22, 23, 24].

In our approach, we use a molecular gas with weakly aligned molecules in a dc electric field. For complete alignment, a much stronger dc electric (or strong ac electric fields) has to be applied. Indeed, for the total molecular alignment, we need the energy difference between molecular levels be larger than the thermal energy, i.e.,

$$\hbar\omega_E \gg k_B T$$

where k_B is the Boltzman constant, and

$$\wp\mathcal{E}_0 \gg \left(\frac{k_B T}{\hbar B}\right)^2 \hbar B$$

that gives a simple estimation of the dc electric field to be of the order of $\mathcal{E}_0 \simeq 100$ MV/cm. We consider the very weak alignment of molecules in the dc field which leads to the resonance with the ac electric field. For the weak alignment, we need to have the dc electric field be sufficiently strong that the coupling is comparable with the rotational splitting, i.e.
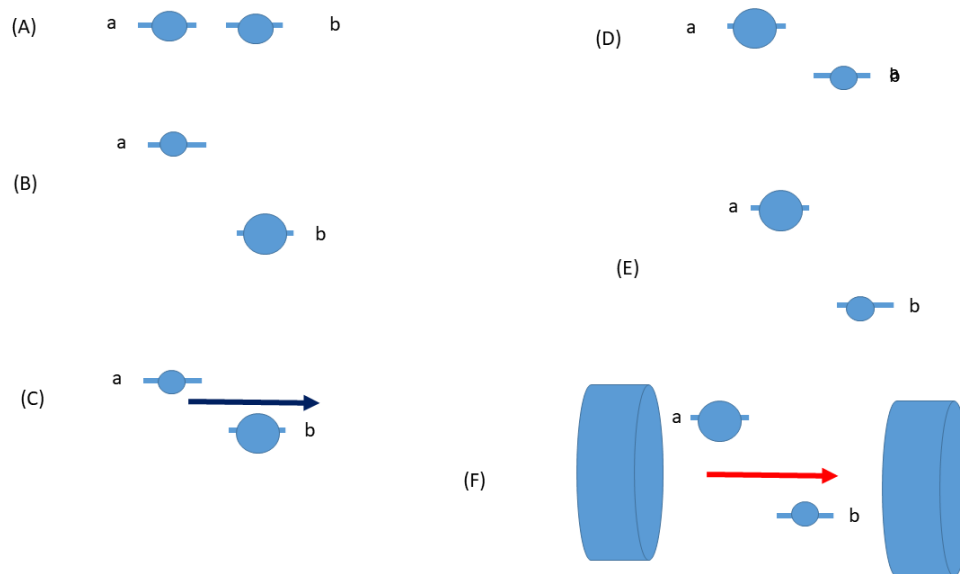
$$\wp\mathcal{E}_0 \simeq \hbar B,$$

Figure 4. Plot shows all stages of population preparation for maser action. (A) Two selected levels from the levels of the molecular rotator that are chosen for serve as a maser transition; (B) Applying a dc field that splits levels, and it creates population difference by thermal equilibrium at room temperature; (C) The splitting tuned to be in resonance with low frequency field to swap population between levels; (D) The swap of population occurs between levels a and b; (E) The transition is tuned by adiabatic field to the maser cavity frequency; (F) The molecular population inverted levels tuned to cavity frequency and in the cavity produce maser radiation.
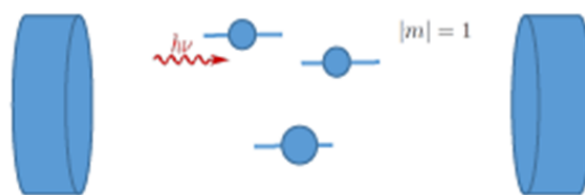

Figure 5. It is schematically shown molecular levels of interests (more molecular levels can be seen in Figure 1) with some populations and resonant microwave radiation in the microwave cavity.

that gives the field to be of the order of $\mathcal{E}_0 \simeq 10$ kV/cm. Our estimations have been made for two frequencies: $\omega_0 = (2\pi)5$ MHz, corresponding to be of the order of $\mathcal{E}_0 \simeq 1$ kV/cm and $\omega_0 = (2\pi)10$ GHz, to be of the order of $\mathcal{E}_0 \simeq 30$ kV/cm.

Applying a dc electric field modifies the rotational energy levels of molecules (see

Figure 1(b,c)). The states $|\tilde{m}\rangle$ in the dc electric field with the rotational states $|m\rangle$ (without electric field) are related as

$$|\tilde{0}\rangle \rightarrow |m = 0\rangle, \quad \text{for } m > 0, \ |\tilde{m} = 2|m| - 1, 2|m|\rangle \rightarrow |\pm|m|\rangle$$

The energy modification can be easily understood by naively considering aligned molecules in the dc electric field [5, 6] (see Figure 2). Aligned molecules can be viewed as simple pendulums. For levels with low energy excitation, they have the energy structure of a simple harmonic oscillator (see Figure 1(a)). For higher excitation levels, the molecules retain rotational structure.

The dependence of the molecular energy $\frac{2IE}{\hbar^2}$ on the $\frac{2I\wp\mathcal{E}_0}{\hbar^2}$ is shown in Figure 1(d). It shows that for the larger fields $\wp\mathcal{E}_0 \gg \frac{\hbar^2}{I}$, the electric dipole under the presence of the electric field looks like a simple harmonic oscillator, with the Hamiltonian given by

$$\hat{H} = -\frac{\hbar^2}{2I}\frac{\partial^2}{\partial\phi^2} + \frac{I\omega_E^2}{2}\phi^2 - \wp\mathcal{E}_0, \tag{15}$$

where $\omega_E^2 = \frac{\wp\mathcal{E}_0}{I}$. As can be seen in Figure 1(b), the ground and lower states have energies

$$E_{\tilde{m}} \simeq -\wp\mathcal{E}_0 + \hbar\omega_E(\tilde{m} + 1/2). \tag{16}$$

Meanwhile, for larger $\tilde{m}$ such that $\frac{\hbar^2}{2I}\tilde{m}^2 \gg \wp\mathcal{E}_0$, the fast molecular rotation completely averages out the effect of the dc electric field, and the molecular energy is $E_{\tilde{m}} \simeq \frac{\hbar^2}{2I}\tilde{m}^2$. One can see that changes in Figure 2 where the molecular level structure is shown in the different dc electric fields. (A) $\mathcal{E}_0 = 0$, no dc field, level structure of rotator; (B) $\mathcal{E}_0$ is weak ($B \gg \wp\mathcal{E}_0$), one can see small splitting of the low levels of rotator; (C) $\mathcal{E}_0$ is strong ($B \ll \wp\mathcal{E}_0$), one can see that the low levels of rotator look similar to the levels of the simple harmonic oscillator. (see Eq. (15) and Eq. (16)).

Then, the proposed maser scheme works in the following stages (see Figures 3 and 4). First, we can chose the levels that can be used for our maser transition (see Figure 4(A)), these can be levels with $|m| = 1$ (see Figure 1). Maser starts at the stage (1), at some relatively strong dc electric field $\mathcal{E}_0$ that causes the weak alignment of molecules. The maser levels a and b are split (see Figure 4(B)). The molecules have the corresponding thermal distribution between molecular levels. Then, the electric field $\mathcal{E}_0$ changes to the vicinity of the electric field $\mathcal{E}_P$ that corresponds coherent pump field (see Figure 4(C)). Time dependence of the adiabatically changing strong electric field (arbitrary units) $\mathcal{E}_0$ shown in Figure 3(A). The corresponding population inversion between corresponding levels (arbitrary units) is shown in Figure 3(B). Stage (2): the dc field adiabatically changes the electric field to tune the molecular transition through the resonance with relatively strong ac pumping field to produce adiabatic population transfer (see Figure 4(D)) that creates the population inversion between corresponding levels (as shown in Figure 1(e)). Stage (3): the dc field adiabatically changes the electric field to tune the molecular transition till it reaches the resonance with the maser cavity mode (see Figure 4(E,F)). One can see the molecular rotational level structure with, for example, $|m| = 1$ (see in Figure 5) that can be used for the maser operation. During this stage the microwave amplification occurs depleting all population inversion and then returning to the pumping stage (1).

Using the set of equations for density matrix and the equation for the microwave cavity field, We estimate the gain for some simple molecules such as [25], using $\wp \simeq 1D$ as a typical value, and for frequency $\omega_0 = (2\pi)10$ GHz, and $N \simeq 2 \ 10^{16}$ cm^{-3} and the gain is of the order of $G \simeq 2 \ 10^8$ cm^{-1} correspondingly.

4. Conclusion

In conclusion, in the paper, we have developed and demonstrated a new mechanism of manipulation of population in molecular rotational levels in a weakly aligned molecules. Treating molecules as simple rotators, we have described their behavior using the density matrix taking into account the relaxation processes. We have considered the interaction of the weakly aligned molecules with a microwave field in a high finesse cavity. Using the mechanism based on an adiabatically changing electric field interacting with the molecules with dipole moments, we have shown that, on one hand, the population inversion can be reached in the ensemble of the weakly aligned molecules to be used for the maser operation at room temperature. On the another hand, we have found that the enhancement of the absorption can reach the theoretical limit and be used for gas sensing with high sensitivity and selectivity.

It is interesting to note here that the same idea of manipulation of population in molecular rotational levels can be used not only to produce the maser effect, but also to detect signal from molecules and do spectroscopy or gas sensing. This technique can work as a sensitive and very selective molecular detector. The resonance depends on the dc electric field applied and the molecular parameters such as rotational constants and the magnitude of the molecular dipole moment. This opens an interesting opportunity of molecular detection at the chosen frequency [7, 8]. Different molecules have their resonances at the chosen frequency but at the different magnitudes of the electric field. Knowing electric field and frequency allows researcher to identify the molecules. We estimate the sensitivity of the technique for some simple molecules such as [25], using $\wp \simeq 1D$ as a typical value, and for frequencies $\omega_0 = (2\pi)5$ MHz and 10 GHz, and obtain $N \simeq 2\,10^{11}$ cm^{-3} and $N \simeq 2\,10^8$ cm^{-3} correspondingly. Such sensors can efficiently analyze the multi-gas mixtures and be used for a huge range of applications – stretching from technology, sciences, control of environment, biology and medicine.

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[25] Parameters of some molecules are the following: CO, $\wp_{CO} = 0.122$ D, $B_e = 1.93$ cm^{-1}; and HCN, $\wp_{HCN} = 2.98$ D, $B_v = 1.47$ cm^{-1}, N$_2$O, $\wp_{N_2O} = 0.17$ D, $B_e = 1.93$ cm^{-1}; and NO, $\wp_{NO} = 0.16$ D, $B_v = 1.67$ cm^{-1}. NO$_2$, $\wp_{NO_2} = 0.316$ D, $A = 8.0$ cm^{-1}, $B = 0.43$ cm^{-1}, $C = 0.41$ cm^{-1}, H$_2$O, $\wp_{H_2O} = 1.85$ D, $B_v = 15.$ cm^{-1}.